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GAS BUBBLE AND BLISTER NUCLEATION IN METALS FOLLOWING LOW-ENERGY ION IRRADIATION.

by

R.J.K. Nicholson, B.Sc.

A Doctoral Thesis submitted in partial fulfilment of the requirements for the award of Doctor of Philosophy of the Loughborough University of Technology.

ABSTRACT

Plasma surface interactions play an important role in the operation of a fusion reactor. One phenomenon is that of radiation blistering, where gas-filled blisters are formed on the surface region of the irradiated solids. The mechanisms for bubble formation have been extensively studied at higher energies (>10keV), although comparatively little work has been done on the effects of low-energy (<5keV) ion irradiation.

A field-ion microscope (FIM) study has therefore been conducted to investigate the nucleation and growth of radiation induced bubbles following low-energy ion bombardment. The FIM is particularly suited for the study of low-energy radiation damage, since not only does the technique possess atomic resolution, but also allows the precise nature and depth of the damage to be determined. The application of the FIM technique to low-energy irradiation studies is reviewed and the use of controlled field evaporation assessed as a depth of damage analysis method, where experimental measurements are correlated with theoretical models to deduce a semi-quantitative damage sizing technique.

The FIM is concerned with the irradiation of tungsten and molybdenum by low-energy (0.2 to 3keV) hydrogen and helium ions with total ion doses ranging from $1 \times 10^{15}$ to $5 \times 10^{17}$ ions.cm$^{-2}$. The investigation of the low-energy damage has primarily determined the effect of increasing ion dose for given projectile energies. These experiments have enabled the nature of the surface and sub-surface damage to be defined on the atomic scale. In addition, since the direction of irradiation has been along the axis of the field-ion emitter, subsequent field evaporation has allowed precise depth of the damage to be determined. The relevance of these results to the current theories of bubble and blister formation is discussed.
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INTRODUCTION

In the intensive search for an efficient, low cost, energy producing system, nuclear energy has become a vitally important source of energy. The proposed successor to the nuclear breeder reactor, as a low cost energy producing device is the controlled thermonuclear fusion reactor (CTR). This device will operate with a net power gain from the thermonuclear fusion of light nuclei. The choice of reacting nuclei will initially be limited to the well known deuterium-tritium fusion reaction:

\[ D + T \rightarrow \text{He}^4 (3.5 \text{ meV}) + n (14.1 \text{MeV}) \]

Since tritium is not naturally available, it must be bred within the reactor using a suitable reaction such as neutrons with lithium.

The main process inside the reactor will involve a plasma of deuterium and tritium which will be heated to high temperatures in the range 100-500 million Kelvins. The hot plasma is contained by magnetic fields to prevent the plasma touching the reactor wall. Approximately 80% of the energy released will be transferred to the emergent neutrons, which in turn will be converted into heat by slowing down collisions in the so called "blanket" material surrounding the first wall, enabling the energy to be extracted.

The development of fusion reactors as a major power source presents many scientific and technological challenges. One major problem involves
the surfaces exposed to the intense radiation from the plasma where plasma surface interactions can occur. During the operation of thermonuclear devices and reactors, the surfaces of major components such as the container, limiter and diverter walls will be exposed to both the primary plasma radiations and the secondary radiations which include charged particles, neutrons and photons. The main components will be 14.1 MeV neutrons, hydrogen (D,T) ions and atoms with a mean energy of 0.5 to 20 keV and helium ions with an ion energy distribution in the approximate range of 100 eV to 50 keV (Martel et al (1977)) with an energetic tail up to 3.5 MeV. The flux rates of these particles are expected to be in the range $10^{13}$ to $10^{18}$ ions cm$^{-2}$ sec$^{-1}$ (Roth et al 1975).

The energetic particles bombarding the surfaces of the major components can cause a variety of surface phenomena such as sputtering, evaporation, blistering, secondary electron and X-ray emission, backscattering and nuclear reactions (see for example McCracken and Stott (1979)), resulting in surface erosion and a flux of wall material into the hot plasma. One such phenomenon is radiation blistering, where gas filled blisters are formed on the surface region of irradiated solids and the gas released on bursting contaminating the plasma, and peeling of blister skins, resulting in wall erosion.

The blistering phenomena was initially observed in irradiated glass by Stark and Wendt (1912), and then later by Primak (1963); Kaminsky (1964) also observed the bursts of gas released by rupturing blisters using a mass spectrometer. The blistering phenomena has been studied on a wide range of materials as suitable candidates for first wall materials. The materials
considered have ranged from stainless steels, alloy steels, various alloys and metals (for example, molybdenum, niobium, vanadium, tungsten and nickel), carbon and sintered aluminium product (SAP). These materials have been bombarded with mainly hydrogen (D,T) and helium ions especially at the higher energies (> 10 keV) where relatively large blisters are observed (see for example, Das and Kaminsky (1973)). Extensive studies have since shown the effects of the radiation damage as functions of the main experimental parameters i.e. dose, ion energy, temperature and surface roughness, (Evans (1978)), (Das and Kaminsky (1974)), (Bauer and Thomas (1973)) at these higher energies.

At the lower energies (< 5 keV) the damage volumes produced by the incident hydrogen and helium ions in large doses (>10^{15} ions cm^{-2}) are considerably smaller. Also the depth of damage is small (typically < 20 nm) due to the low mean projected ranges of the light ions in heavier solids. Consequently it is difficult to investigate low-energy radiation damage with the techniques used for such surface studies (e.g. electron microscopy). The field-ion microscope (FIM) is particularly suited for the study of the radiation damage produced by low energy ions. Since not only does the technique possess atomic resolution, but it also allows the precise nature and variation of the damage with depth to be investigated by field evaporation where successive atomic layers are removed from the damaged surface.

In this thesis the basic principles concerned with the application of the FIM technique to the study of low-energy irradiation damage are considered. The interpretation of image contrast associated with void or bubble formation is discussed and the moiré analogy is used to deduce
new information regarding non-spherical planar facets on the emitter surface. A simple theoretical model is used to deduce the relative amounts of material removed from the different regions of the emitter surface and correlated with extensive experimental measurements. The field evaporation model forms the basis of a semi-quantitative void sizing technique which enables the void size and depth distributions to be determined in the irradiation study.

The production of low-energy ion beams is discussed in detail, and several different ion sources have been utilised for ion irradiation. The design and development of a mass-analysed ion source is described. A novel irradiation source is mentioned which may allow the observation of radiation damage during simultaneous irradiation and operation of the FIM.

The general aspects of radiation damage are reviewed from the literature, especially those factors concerned with the nucleation and growth of gas bubbles and blisters. The theoretical treatment of range and damage distributions of low-energy ions is described, based on computer calculations (LSS theory), and appropriate values are obtained for low energy hydrogen and helium ions in tungsten and molybdenum respectively. The conditions necessary for bubble formation and blistering are reviewed in terms of the current theories used to describe bubble and blister formation.

The FIM technique has been used to investigate the low-energy radiation damage produced by hydrogen and helium ions (0.2 to 3 keV) in tungsten and molybdenum by various irradiation techniques. The helium irradiation of tungsten has been examined in detail in the ion dose range $1 \times 10^{15}$ to
5 \times 10^{17} \text{ ions.cm}^{-2}. The ion irradiation has been performed along the axis of the emitter and thus allows the void size and depth distributions to be determined. These measurements enable new information to be deduced regarding the nucleation and growth of the gas filled voids leading to blister formation. The helium irradiation of molybdenum is also described. The general trends and relevance of this work for low energy irradiation is discussed in terms of the current theories of bubble and blister formation.
CHAPTER ONE

Field-ion Microscopy

1.1 Basic principles of field-ion microscopy

The field ion microscope (FIM) can be considered as a point projection device which has high resolution and magnification properties and permits direct observation of the atomic lattice. The specimen consists of a sharply pointed needle, the end point of which is approximately hemi-spherical and has a radius of about 10 to 100 nm. The specimen is mounted inside a vacuum chamber facing a phosphor screen. In operation the microscope is evacuated to a background pressure of less than \(5 \times 10^{-7}\) Torr; the specimen is cooled to liquid nitrogen temperature (\(\sim 78^\circ\text{K}\)) and has a high positive potential applied to it via mounting leads. Imaging gas (usually helium or neon) is then admitted to the specimen chamber to a pressure of \(\sim 5 \times 10^{-5}\) Torr.

The imaging gas atoms approaching the specimen tip are polarised by the high electric field (\(\sim 10^{10}\) V m\(^{-1}\)) due to a high positive potential applied to the specimen which is cooled to liquid nitrogen temperature (\(78^\circ\text{K}\)). The extremely sharp curvature of the specimen enables an electric field of several volts per Angstrom to be generated by the application of a few kilovolts to the specimen. The gas atoms are attracted to the emitter surface due to dipole effects and ionisation takes place close to the surface by electron tunneling in the highest regions of the field, usually the field associated with atoms located at protruding sites. The resulting ion is then repelled from the tip, down a potential gradient to an earthed screen. The beams of ions produced in this manner form a projected pattern of bright spots, which are representative of the atomic location of the protruding atoms on the emitter surface.
The intersection of a curved emitter surface with a crystal lattice produces a faceted structure in which most of the protruding atoms occur on ledges in concentric rings as shown in Fig. 1.1. This concept was illustrated by Müller (1960) using ball models, where the characteristic ring structure of field-ion micrographs was readily observed. Each of the imaging atoms lies on the edge of a crystal plane, the atoms within that plane will not produce an image due to decreasing field strengths. This arises because only those atoms on the step edges or on high index (i.e. non closed packed) planes protrude sufficiently from their neighbours to act as individual ionisation centres. Wald (1963) has shown using a ball model, that the fraction of visible atoms decreases with increasing tip radius. Typically for a 50 nm radius tip only 5% of the atoms on the emitter surface are observed.

The rings of atoms constituting various crystallographic planes can be indexed by reference to the symmetry of the pattern for different specimens. Once the prominent planes are identified by symmetry, self consistent sets of Miller indices for the different planar facets exhibited by the tip may be determined.

1.2 Field ionisation

The concept of ionising a free atom by the tunnelling of an electron in the presence of a high electronic field was first proposed by Oppenheimer (1928). Field ionisation can be considered as a quantum mechanical process where an electron is transferred from the thermally accommodated gas atom close to the metal surface. A neutral atom of the imaging gas is influenced by the large fields around the emitter and becomes polarised, acting as an orientated dipole. As the polarised atom
Fig. 1.1 The intersection of the curved emitter surface with the crystal lattice producing a faceted topography, in which the protruding atoms occur on the ledges of concentric rings.
approaches the surface the potential well about the gas atom becomes distorted, influenced by the large field of the emitter. Ionisation is therefore possible by the tunneling of an electron through the potential barrier at the metal surface, provided there is an available electronic energy level.

Figure (1.2) schematically illustrates the mechanism of electron tunneling, where the Figs. (a) and (b) show the potential energy diagrams of an outer electron of an atom with and without an applied external field. The electron is shown at a potential \( I \) above the zero energy level. In the latter case ionisation can occur by the electron tunneling through the potential barrier along the path A B. The tunneling probability increases as the atom approaches the metal surface as shown in (c). The potential barrier has decreased in height due to the image forces which attract the electron to the image of the ion-electron dipole in the metal surface.

However if the gas atom approaches closer than a critical distance \( X_c \), the tunneling probability is greatly reduced due to the low density of vacant energy levels to which the electron may tunnel. Müller (1973) has shown through energy considerations that the critical distance for ionisation \( (X_c) \) neglecting image forces, could be found from an expression of the form:

\[
X_c = \frac{I - \phi}{F e}
\]  

(1.1)

where \( I \) is the ionisation potential, \( \phi \) the effective work function of the metal surface, \( F \) is the applied electric field and \( e \) the electronic
Fig. 1.2 Schematic diagram of the mechanism of electron tunnelling.

(a) Potential energy diagram for an electron in a helium atom as a function of the distance, $x$, from the nucleus

(b) The effect on the potential energy curve when the external field is applied. Tunnelling occurs from A to B.

(c) Ionisation at the emitter surface for a helium ion.

(Courtesy H.N. Southworth).
charge. In the case of tungsten ($\phi = 4.5\text{eV}$) imaged with helium gas (where $I = 24.6\text{eV}$) and a field of $0.45\text{V/nm}$ it is found that $X_c$ is $0.45\text{nm}$. Although these simple theories lead to an understanding of the field ionisation phenomena, several effects cannot be adequately explained. These include regional brightness variations (i.e. regional variations in ionisation rate), hydrogen promotion effects and gas promoted field desorption and these have been extensively discussed by Müller (1973).

1.3 Field evaporation

The process of stripping away and removing the surface atoms from the specimen, by the application of an electric field is known as field evaporation. If the applied electric field is raised sufficiently above that required to obtain an image, the removal of surface atoms is observed. The process begins at a well defined field, and the rate of removed of atoms increases rapidly with field. The phenomenon is manifested by the collapsing to the centre of the circular facet structure composed of imaged atoms around each surface plane. Continued field evaporation of a freshly prepared specimen preferentially removes any topographical irregularities and eventually reveals the characteristic field evaporated end form which is free from contamination. The field evaporation process is essential for the production of a nearly perfect, almost hemispherical crystal surface from the rough end form produced by specimen preparation.

The field applied during the evaporation process can be accurately controlled so that single atom or layer removal enables a full bulk investigation to be made. This fact makes the use of the FIM technique
powerful tool for research. However, although field evaporation allows the structure of the surface and sub-surface regions to be examined, it also restricts the wide scale application of the technique. This restriction occurs since the surface atoms of some metals can be removed by a field lower than that necessary for the ionisation of the imaging gas, thus preventing stable image formation. This problem can be overcome to some extent by the reduction of specimen temperature and the use of suitable imaging gases having lower ionisation potentials (such as neon and argon), and can extend the range of materials available for examination (see for example, Boyes (1974)).

The process of field evaporation is a complex problem which is not fully understood, although several theories have been proposed to explain the phenomenon. Only a brief review will be given here, indicating the principles involved with the two basic mechanisms. These are the Image Force Model (Müller, (1956)) and the Charge Exchange Model (Gomer, (1961)). The image force model considers the escape of a metal ion over a potential energy barrier and from the superposition of a potential resulting from the applied field, together with an image force attracting the ion to the surface. The superpositioned potential reduces the maximum in the potential energy of the ion. The energy \( Q_o \) required to remove a surface atom as a multiple charged ion is given by:

\[
Q_o = A + E_n I_n - n\phi \tag{1.2}
\]

where \( A \) is the sublimation energy and \( E_n I_n \) the total ionisation energy. The activation energy \( Q_n (F) \) for field evaporating a surface atom as a multiple charged ion in an applied field \( F \) is then given by:
Muller (1960) has shown that if a surface atom is evaporated thermal activation over a potential barrier $Q$, reduced slightly by the applied field, then the critical field of evaporation is given by:

$$F = n^{-3} e^{-3} \left[ A + \sum_n I_n - n\phi - kT \ln \frac{t}{t_o} \right]^2$$

where $t$ is the time constant for evaporation and $t_o$ the reciprocal of the lattice vibration frequency.

The charge exchange model was initially proposed for the desorption of electronegative gases in high fields. The model considers a transition between bound states to multiple charged states for a field desorbed atom $A$ to a metal atom $M$ as the transition process shown:

$$A + M \rightarrow A^{n+} + M^{n-}$$

The model is valid for locally bonded adatoms but has been extended by Tsong (1970) to the field evaporation of metal atoms.

Field evaporation is an important process for the analysis of sub-surface radiation damage and depth investigations. The practical application of controlled evaporation will be dealt with in greater detail in Section 3.7.
1.4 Resolution

Müller (1951) initially observed a hydrogen image of tungsten but with low resolution. Later individual atoms on a tungsten surface were observed (Müller, 1960) with helium imaging gas at liquid hydrogen temperature, with sufficiently low imaging gas pressure to prevent mean free path collision scattering of the ions. Müller (1960) and Gomer (1961) have shown that the resolution of a F IM image should be dependent on the specimen temperature, applied voltage, imaging gas and the shape of the emitter.

The resolution of the F IM is mainly limited by two factors, namely (1) the diffraction effect due to the wave nature of the ions and (2) the tangential component of the initial velocity of the ion generated by field ionisation. Müller (1960) has shown that the potential resolution can be determined by an expression of the form:

\[ \delta = \left[ 4.5 \times 10^{-23} \left( \frac{T}{m} \right)^{\frac{1}{2}} + 5.2 \times 10^{-7} \left( \frac{T}{F} \right) \right]^{\frac{1}{2}} \]  

(1.5)

where, \( m \) is the mass of the imaging gas atom, \( r \) the tip radius, \( F \) the best image field for each gas, and \( T \) the imaging gas temperature. This expression has been modified (Müller and Tsong (1969)), to take into account the dimensions of imaging gas atoms and ionisation zone \( g(D) \), so that \( \delta \) becomes:

\[ \delta = g(D) + \left[ 9 \times 10^{-23} \left( \frac{T}{mF} \right)^{\frac{1}{2}} + 2.07 \times 10^{-6} \left( \frac{T}{F} \right) \right]^{\frac{1}{2}} \]  

(1.6)
Adachi and Nakamura (1972) have experimentally measured the resolution of the FIM image for a tungsten surface as a function of the temperature, tip curvature and imaging gas molecules, and found reasonably good agreement with equation (1.6). However, taking typical imaging parameters for a tungsten specimen with helium gas, the resolution is found to be about 0.12 nm (Southworth (1970)) with a tip temperature of 210 K. As a general rule, better resolution is obtained at lower specimen temperature. The resolution is such that interatomic spacings of 0.3 nm can be resolved at liquid nitrogen specimen temperatures.

1.5 The effect of field induced stress.

The specimen in the FIM is subjected to high electric fields which can produce elastic deformation and leads to the formation of stress distributions in the specimen. The magnitude of the stress \( \sigma \) produced by a field \( F \) can be determined (Müller, (1958)) from the relation:

\[
\sigma = \frac{F^2}{8\pi} = 44.3 F^2
\]  

Under typical imaging conditions with helium at \( F = 0.45 \text{ V mm}^{-1} \) the stress is about \( 10^6 \text{ N mm}^{-2} \). A stress distribution is produced due to the considerable variation of field over the emitter surface, as a result of the differing radii of curvature of various crystallographic regions. A second factor is the influence of the shank on the emitter field distribution producing field screening effects. Generally the stress distribution has two components, which are of a hydrostatic and shear nature. Both components can be generated by non-spherical
end forms, and can lead to differential dilations, with resultant displacement of the imaged atoms. This effect although very small has been measured (Rendulic and Müller, (1967)), who showed that the overall effect for a uniform and hydrostatic stress resulted in a dilation of the specimen.

The application of the field to the specimen produces an elastic deformation and an increase in tip volume due to an assumed negative hydrostatic pressure of about $10^5$ (Müller and Tsong, (1969)). Therefore field-ion microscope observations are always made upon a strained material, where the strain is considered to be elastic. The relative decrease in volume can be calculated from the relation:

$$\frac{\Delta V}{V} = -\frac{3(1-2\nu)}{E} p$$  \hspace{1cm} (1.18)

where $\nu$ is Poisson's number and $E$ is Young's modulus about $2 \times 10^7$ N.cm$^{-2}$ (Bowkett and Smith, (1970)). The relative changes in volume for molybdenum and tungsten are about 3.5 and 3.2 per cent respectively.

Smith and Smith (1970) have shown that the tensile stress in a specimen under helium imaging conditions decreases along the shank of the specimen away from the surfaces. Although the stress levels necessary for fracture are not generally exceeded due to the absence of dislocation sources. Yielding processes in the shank leading to fracture can occur, but the effect can be minimised by low temperature operation.

The effect of field induced stress is important in the present
study, due to the presence of gas filled bubbles existing close to the surface following high dose bombardment. The presence of the sub-surface bubbles will produce a weakening effect on the tip structure which may cause fracture to occur. An estimate of the stress involved can be obtained using the Griffith crack formula (Bowkett and Smith, 1970).

\[ \sigma = \left( \frac{Ey}{r} \right)^{\frac{1}{2}} \]  

(1.19)

where \( \gamma \) is the surface energy (about \( 10^{-3} \text{ J cm}^{-2} \)). For a void of radius \( r \geq 2 \times 10^{-2} \text{ nm} \) fracture would occur, thus smaller voids can be imaged without fracture, although the large internal gas pressure associated with a bubble will increase the stress and the likelihood of fracture. The effect of stress on bubbles will be discussed in more detail later, together with vacancy and interstitial observation in the FIM.

1.6 Experimental details

The experiments were carried out in stainless steel ultra-high vacuum (UHV) field-ion microscopes designed and built by Vacuum Generators Ltd. The microscopes could be fully baked up to 200°C together with ancillary equipment to facilitate sufficient outgassing, to that base pressures lower than \( 5 \times 10^{-10} \text{ Torr} \) (measured with V.I.G. 10 ionisation gauges) could be routinely achieved.

The UHV microscope initially used in the preliminary experiments utilised three separate types of vacuum pumps, comprising a 50 \( \text{s}^{-1} \)
triode Ferranti ion pump (including a mu-metal cover for stray magnetic field shielding). A 2 inch mercury diffusion pump with a double liquid nitrogen trap (type LNT 2) was employed and backed by a 50 l s\(^{-1}\) rotary pump together with a 6 inch titanium sublimation pump (TSP). This combination allows the system to be pumped by the ion pump during baking and by the trapped diffusion pump during imaging with helium or neon gases. This ensures a continuous flow of imaging gas, which is difficult to achieve by ion pumping at high inert gas pressures. The position of the titanium sublimation pump allows active contaminant gases to be removed from the system by chemical reaction with the freshly activated titanium, thus the dynamic gas supply technique prevents the build up of impurity gases.

The second UHV microscope was pumped by a single air cooled 6 inch oil diffusion pump, with a large capacity liquid nitrogen trap and backed by a 50 l s\(^{-1}\) rotary pump. The use of additional water cooling enabled the system to be baked and pumped simultaneously. The schematic layouts of the microscopes are shown in Fig. (1.3).

The imaging gases used in all the experiments were of research (grade X) quality and supplied by B.O.C. Ltd. Up to three gases could be used on the microscopes, usually helium, neon and hydrogen gases. The gases could be admitted into the system at a finely controlled rate by the use of calibrated leak valves (MD6) having an adjustable leak rate of < 10\(^{-9}\) Torr litre sec\(^{-1}\).

The internal construction of the microscopes is shown in Fig. (1.4). The specimen, usually held in a copper block, is connected to metal pins,
Fig. 1.3 The schematic layouts of the field-ion microscopes used in the study.
Fig. 1.4 The internal construction of the main chamber of a field-ion microscope.

A Specimen
B Support insulating cylinder
C Copper gripping jaws
D Inner dewar
E Outer dewar
F Insulated electrical conductor
G Electrical feedthrough
H Image intensifier assembly
I Viewport
which pass through a ceramic insulation to prevent any high voltage leaks and for reasonable heat transfer. The insulator is clamped by means of copper jaws onto the main cooling assembly commonly known as a "cold finger". This consists of a double dewar, the inner one having a copper base (directly connected to the copper jaws) welded to the stainless steel dewar body. The coolant, usually liquid nitrogen is placed in both inner and outer dewars enabling a fast cooling rate of the specimen to be achieved.

A large voltage, up to 30kV may be applied to the specimen, supplied by a Brandenburg high voltage power supply, via a high voltage glass feedthrough and insulated leads to the pins. The colder finger is arranged so that the specimen points directly at the image intensifier and screen assembly located at a central view port.

1.7 Imaging gases

A major problem with the application of field-ion microscopy to the observation of a large range of materials is the removal of surface atoms at fields, lower than that necessary to produce ionisation of the imaging gas. The most common imaging gas for the refractory metals is helium, which has a large ionisation potential (24.6eV) and consequently a large field (typically about 450 MV cm⁻¹). Helium possesses a small atomic diameter and low polarisability so that high atomic resolution can be achieved. Helium imaging gas is particularly useful in non UHV microscopes, where due to the large field, contaminant gas species are ionised at some distance from the emitter surface, thus avoiding any contamination effects (e.g. field etching of the emitter surface).
The other gas commonly used for imaging is neon which possesses a lower ionisation potential (21.6eV) than helium; subsequently lower electric fields are required for ionisation (about 350 MV cm⁻¹) resulting in lower intensity images observed on the screen. The field associated with neon imaging although high enough to prevent corrosion of the emitter surface to occur, does allow the observation of contaminant atoms on the emitter surface. This becomes apparent at fields below the 'best image voltage' (BIV), where the surface rapidly becomes covered with adsorbed species indicated by bright image spots distributed over the emitter surface.

The use of helium-neon gas mixtures, however combines the imaging properties of both gases, giving better overall contrast on tungsten and molybdenum specimens. This is achieved by the helium imaging high field regions, and neon preferentially imaging low field regions. The use of helium-neon mixtures produces little change in the image intensity normally associated with neon imaging with a small reduction (about 5%) in the 'best image voltage'. Both tungsten and molybdenum specimens were imaged in helium-neon mixtures of 1 x 10⁻⁵ Torr Ne and 5 to 8 x 10⁻⁵ Torr helium.

1.8 Image recording

The use of the channel plate is now almost universal in field-ion microscopy and has considerably reduced the exposure time necessary for image recording. The channel plate image intensifier essentially consists of a hexagonal matrix of up to 10⁶ parallel fine tubes of semi-conducting glass, in the form of a 2 inch diameter disc. It is princi-
pally an amplifying device and when suitably biased can produce a gain of about $10^2$ to $10^5$ (Boyes (1974)).

Any incoming charged particle entering a channel and striking the channel wall will initiate secondary electron emission. The applied field will thus amplify the resulting electron cascade before reaching the output side. The maximum secondary electron current is limited by the plate resistance which usually lies in the range $1 - 10 \times 10^7$ ohm. In practice the channel plate is mounted directly in front of the specimen with a 3 to 10 cm separation. The front face of the channel plate is normally earthed and a positive voltage applied to the reverse side via current limiting high voltage resistors (about $10^8$ ohm). An applied voltage between 500 to 2000 volts is sufficient to produce a bright image suitable for image recording dependent on the imaging gas pressure.

The channel plate forms the major component of the image intensifier assembly although different forms of focusing of the electrons onto the screen are employed in each microscope. In both cases the channel plates were supplied by Mullard Ltd and subsequently fitted into the converter assembly by Vacuum Generators Ltd. The first UHV microscope has a proximity focusing arrangement similar to that of Brandon (1966), where the electrons are directly focused by the channel plate onto a P8 type (green) phosphor screen. The second UHV microscope has a focusing arrangement combining electric and magnetic fields to produce a well defined image (Turner et al, (1969)). A fixed magnetic field (approximately 150 Gauss) is supplied by an external permanent magnet, and together with a series of concentric ring electrostatic lenses operated by a potential dividing system, enables the electrons to be focused onto the P11 type
(blue) phosphor screen. In each case the phosphor is deposited onto a tin oxide coated glass surface, and covered with a thin film of aluminium to improve image brightness and contrast by the reduction of reflected light produced by electrons incident on the phosphor.

The image on the screen was photographed using a Micro-Nikkor Auto 55 mm f/3.5 lens with a Nikon F camera. An extension ring (Nikon M2) is used to produce an image magnification of 1:2. The camera together with a binocular microscope is mounted on a table in front of the screen with a separation distance of about 17 cms. The table could be traversed for either close image examination or image recording. Typical exposure times ranged from 5 to 10 seconds at f 5.6 with Kodak TRI-X (ASA 400) and Ilford HP4 (ASA 400) 35 mm films. Films developed in Universal developer or Microdol X gave reasonably good quality contrast negatives. A closed circuit T.V. recording system has also been used to record transient images on the screen, especially in the controlled field evaporation experiments. This will be discussed in detail in the section dealing with field evaporation.
CHAPTER TWO

Production of low-energy ions.

2.1 Introduction

In recent years there has been a large increase in the application of low energy (≤ 5 keV) ion beams for surface analysis and surface processing. The interaction of ion beams with surface can provide useful information concerning the elemental surface composition and the variation of the surface constituents with depth. The more important applications enclose a variety of surface techniques such as ion scattering spectroscopy (ISS) (Honig and Harrington, (1973)), secondary ion mass spectrometry (SIMS) (Lieble, (1975)) and ion bombardment studies. These studies can be separated into two main areas, the first dealing with the low-energy sputtering phenomena, the second concerning sub-surface radiation damage. Thermal desorption techniques have been successfully utilised for the investigation of the radiation damage produced by inert gas ions incident on metal surface and subsequent sub-surface damage (Kornelsen (1970)). Most of these techniques require ion beams with known characteristics so that the interaction between the bombarding ions and the surface and bulk atoms can be more fully understood.

The ion sources used in the investigation of ion-bombardment phenomena consist of two main types, (1) gas discharge ion sources, and (2) electron impact ion sources. In the first type, the source pressure is generally greater than $10^{-3}$ Torr (Khan and Schroer, (1971)) with gas flowing through the source, so that differential pumping is required to obtain a low pressure in the target chamber. The electron
impact source was originally developed by Dempster (1921) and subsequently improved by Nier (1974), and does not require differential pumping. However this type of source produces rather low ion currents (about $10^{-8}$) although high current densities can be achieved under suitable focusing conditions. However for certain types of experiments under UHV conditions low ion current densities are required (Suurmeier and Boers (1971)) and are particularly suited to the electron impact type source.

General reviews on various types of ion sources have been published, these include Carter and Colligon (1968), Hurley (1972), Wilson and Brewer (1973) and Green (1974), where different ion beam systems are discussed together with their applications. The performance of low-energy ion sources suitable for surface studies, in terms of the source ion-optical system have also been discussed by Hurley (1975), Kornelsen (1976) and Liebl (1978).

In the present study several ion sources have been used where ions have been generated using different techniques. The ion sources are required to produce positively charged hydrogen and helium ions in a UHV environment. For controlled 'in-situ' radiation damage experiments at low energies, the bombarding ion beam is very important and consequently the experimental parameters have to fulfil certain requirements. These are listed as follows:

1) The ion beam must be monoenergetic with a narrow energy distribution. The energy range of interest is from 200 eV to 3 keV.

2) The ion beam should be mass analysed so that the ion current
and the resulting damage at the target will be produced by only one ion species. The mass resolution need not be too large \((M/\Delta M < 10)\), since the ion current must be as high as possible.

3) The use of X, Y deflector plates is required to allow the ion beam to be directed off axis, not only to locate the precise area for bombardment, but also to separate any neutral atoms from the ion beam.

4) The ions need to be collimated or focused into a fine beam (< 0.5 cm diameter) so that relatively high current densities (> 1 µA cm\(^{-2}\)) with uniform beam profiles can be achieved.

2.2 Production of ion beams

Generally any process which on impact can impart energy greater than the first ionisation potential of the required gas atom species can produce positively charged ions. One of the most common methods of imparting this energy is by means of an electrical discharge in a gas of chosen atom species. This results in a plasma being formed where positive ions are produced by electron bombardment. In this type of hot cathode electron impact type source a tungsten thermionic cathode is used. Oxide coated cathodes have been successfully used (Dowretsky et al. (1968)), although coated cathodes need to be frequently activated and may produce contaminant background gases unsuitable for some surface studies.

Electrons produced by a hot tungsten filament are accelerated to an energy greater than the first ionisation potential of the gas atoms.
(viz. 24.6 eV for helium and 15.6 eV for hydrogen) towards a cylindrical grid anode usually at 150V for maximum efficiency. This is achieved by mounting the filament next to a grid assembly similar to that used in a Bayard–Alpert type ionisation gauge. During the passage of the electrons through the gas, the electrons can transfer energy to the gas atoms by means of elastic and ionising collisions, excitation and dissociation. The increase in kinetic energy of the gas atoms during these processes is usually small due to the large mass imbalance of the momentum transfer. This can lead to low ion energy distributions due to the small energy spread of the accelerated electrons.

With increased electron ionisation energy (dependent on the accelerating voltage) the excess energy can appear in different forms as multiple ionisation, excitation and increased kinetic energy of the secondary electron after collision. Thus electrons travelling through the gas atoms suffer ionising collisions producing positively charged gas ions. These ions are then trapped inside the grid anode assembly which is positively biased to prevent the neutralisation of the ions by space charge effects. The ions are effectively screened by an equipotential field and can only escape by extraction at one end of the cylinder.

The positive ion current produced in the source is dependent on several parameters, the main factors are the operating gas pressure, $p$, the electron current $i^-$ at the grid, the mean path length of the electrons $\ell$ and the differential ionisation coefficient $\sigma$. The number of positive ions produced per second $i^+$ can then be determined (Wilson and Brewer (1973)) from the expression:
\[ i_+ = \frac{i_{-} \lambda p \sigma}{e} \] (2.1)

where \( e \) is the electron charge. The differential ionisation coefficient is primarily dependent on the electron energy and the gas species (von Engel (1965)). Figure 2.1 shows the variation of the differential ionisation coefficient with electrons of increasing energy for different gases (Lafferty (1971)). Electrons with energies less than or much greater than the atomic and molecular ionisation energies do not ionise the gases effectively. Each particular gas has a curve with a maximum at certain electron energies. (From Fig. 2.1 helium \( \sigma_{\text{max}} \) is about 100 eV), so that suitable selection of the electron energies will produce the maximum possible ion current.

The grid operates at an applied voltage which governs the ion energy, since ions produced inside the grid structure will assume the grid potential. Any applied voltages will be relative to the grid potential (including the filament supply) to produce any biasing voltages necessary for extraction and focusing. Ions can be extracted from the grid structure through a negatively biased extracting aperture which can accelerate the ions into a electrostatic lens focusing system or directly onto the target. The electron impact type source is thus particularly suited for this study and has a low energy spread with a FWHM less than 1 eV (Hartley (1975)). This type of source together with an effective ion-optical focusing system has enabled relatively large ion current densities (\( > 1 \mu \text{A cm}^{-2} \)) to be produced; hence allowing sufficient numbers of ions necessary in achieving the relatively high doses (\( > 10^{15} \) ions cm\(^{-2} \)) to be achieved, for the study of the nucleation and growth of the bubbles and blisters.
Fig. 2.1 The variation of the differential ionisation coefficient, $\sigma$, with electron energy, $E$, for several different gases (Lafferty (1971)).
In this chapter the various ion source configurations used are discussed, each one employing the same principle of gas ionisation by electron impact from a hot cathode, except for the case of cathode sputtering which will be dealt with separately at the end of the chapter. The first section deals with the basic principles involved with the production of ions and subsequent sections discuss the various source configurations and ion-optical components including mass separation of the ions.

2.3 A simple-ion source

The ion source (AG1) was supplied by Vacuum Generators Ltd as a source suitable for the sputter cleaning of specimens and for pressure measurement as an ionisation gauge in the pressure range $10^{-3}$ to $10^{-8}$ Torr. Although normally utilised as an argon sputter cleaning source for the removal of surface contamination prior to surface analysis, the ion gun has been used to produce low-energy (200 to 500 eV) hydrogen and helium ions.

The ion gun is constructed of non-magnetic stainless steel, copper and nickel components, and supported on an 8 pin metal to ceramic feedthrough flange (FC 38) allowing baking up to 400°C. The ion source is similar to the Bayard-Alpert type with a filament situated close to a cylindrical wire grid, with a small separation of several mm, and surrounded by a second cylindrical mesh. One end of the outer mesh has a small aperture forming an accelerating lens. The whole structure is enclosed by an earthed steel can, having a fine mesh circular aperture through which the ions are finally extracted as shown schematically in Fig. 2.2.
Fig. 2.2 A schematic diagram of the modified simple ion source (Y.G. - AGl).
Ions are produced by ionising collisions between the gas atoms and the electrons accelerated to about 140 eV from the hot tungsten filament. The filament power is usually 20 to 30 Watts, although during outgassing 60 Watts can be achieved. The ion gun is controlled by a separate unit (supplied by Vacuum Generators Ltd) which supplies the required current to the filament at a constant voltage (about 10V) dependent on the electron emission current. The electron emission is controlled by a feedback transistor amplifier which ensures a constant emission (500 μA for pressure measurement and 5 mA for ion-bombardment) against any variation in chamber pressure or mains supply. The controlled emission (typically < 1%) enabled a stable ion current to be measured at the target. Contaminant effects due to stray electrons are considerably reduced by the non-line of sight orientation of the filament.

Ions produced in the inner grid are extracted through the aperture of the second grid by a negatively biased potential (about -50 volts). The whole assembly is raised to the ion energy with respect to earth, by the application of suitable voltages supplied by the control unit. Variable ion energies are achieved by the use of constant biasing voltages applied to the filament and grids, which are separately raised relative to the ground potential. The ion gun does not have any focusing elements, so that the ions produced in the inner grid emerge at various angles in a general conical shape from the exit aperture in the earthed can. Similarly the ions cannot be deflected onto the target, thus the orientation and position of the target is important.

Ion current density measurements were performed by in-situ experiments in the FIM, where ion currents were measured on metal targets of
known areas. This was achieved by positioning small nickel flags (about 1 x 1 and .5 x .5 cm² respectively) at the normal position of the field-ion specimen on the cold finger, shown schematically in Fig. 2.3. The current density measured at the flag would then be representative of the ion dose received by the field-ion specimen under identical bombardment conditions. (The ion current was measured with a Keithley 602 Picoammeter.)

Several experiments were performed to establish the optimum gas pressure by ion current measurements at different gas pressures (from $10^{-6}$ to $5 \times 10^{-4}$ Torr), these results are shown in Fig. 2.4 for helium ions are different ion energies with a 1 cm² flag. The optimum gas pressure was found to occur at $4 \times 10^{-4}$ Torr, where the ion beams attained a high current density, further increases in pressure caused non-linearity due to space charge effects. The gas pressure readings are nitrogen equivalent pressures measured by a Bayard-Alpert ionisation gauge. For true pressure a conversion factor is required, however the equivalent pressures will be assumed in all subsequent measurements. The overall linear relationship between the ion current and the gas pressure is in agreement with equation (2.11). The measurement helium current densities ranged from about 0.9 μA cm⁻² at 200 eV to over 1μA cm⁻² at 500 eV.

The ion gun was modified by the addition of an extra earthed collimating aperture (8mm diameter) situated 8mm in front of the existing earthed aperture to reduce the conical nature of the beam. This resulted in higher current densities at the target. These results are shown in Figs. 2.5 and 2.6 for hydrogen and helium ions respectively using a .25 cm² target.
A schematic diagram of the experimental layout in the PM for the measurement of the ion beam characteristics.

FIG. 2.3
Fig. 2.4  Operating characteristics of the simple ion source. The ion current as a function of the helium gas pressure for several ion energies. (0.25 cm² target).
Fig. 2.5 Operating characteristics of the modified simple ion source. The ion current as a function of the helium gas pressure (0.25 cm$^2$ target).
Fig. 2.6  Operating characteristics of the modified simple ion source.
The ion current as a function of the hydrogen gas pressure (0.25 cm$^2$ target).
The additional aperture produced higher current densities although space charged effects producing non-linearity occurred at lower pressures, especially at the lower ion energies. The distance between the ion gun and target is important as increased current densities were measured with smaller separation distances, indicating that small separations would result in higher current densities. At separations of about 4 cm and with an additional aperture ion current densities of about 10 μA cm\(^{-2}\) and 8 μA cm\(^{-2}\) can be attained for 500 eV hydrogen and helium ions. The close agreement between current densities measured on targets with different dimensions indicated that the beam profile was relatively uniform.

One novel use for this type of ion source, is the technique of raising the ion gun to the tip potential and operating the source during the imaging of the specimen with suitable imaging gas. The electrically isolated ion source technique has several advantages over other types of ion sources since the irradiation takes place during actual imaging conditions within the F I M. Thus allowing the direct observation of surface damage as a function of irradiation time (dose). The technique is still in a development stage but may be particularly promising method for irradiation studies, particularly coupled with closed circuit television (CCT) monitoring. The technique would be especially useful for blistering studies and other surface techniques dependent on ion bombardment (e.g. ISS and SIMS) in which surface damage may have an important role.

In the electrically isolated ion source technique the source control unit (0 - 0,5 kV) has to be isolated from voltages up to 20 kV with respect to earth potential. The source itself is further raised
Vacuum chamber

ION GUN

CONTROL UNIT
(0-5 keV)

(0-2 keV)
Supply

STEP-UP TRANSFORMER

30kV ISOLATING TRANSFORMER
(10V, 30A)

MAINS

Fig. 2.7
A block diagram of the electrical circuit used in the electrically isolated ion source.
above the isolated voltage via an additional separate voltage supply (0-2 kV) which also governs the ion energy. The electrical circuit is schematically shown in Fig. 2.7. Ion energies, which are variable up to 2.5 kV can be obtained and superimposed on the floating voltage (i.e. specimen imaging voltage). The control unit and voltage supply are isolated from earth by a 300 VA isolating step down transformer up to 30 kV with a maximum output of 10V, 30A and subsequently stepped up to mains via an intermediate transformer. The ion gun is electrically isolated from the earthed vacuum chamber by a special UHV extension incorporating a glass tube about 3cms in length joined by glass to metal seals onto suitable flanges. The isolated ion-gun arrangement is shown in Fig. 2.8 and is aligned directly onto the field-ion specimen.

Ion current measurements for this ion gun with helium ions at different ion energies and gas pressures are shown in Fig. 2.9. The ion gun to target separation distance is 14 cms, with a target consisting of a 0.25 cm² nickel flag. At the lower pressures (> 5 x 10⁻⁵ Torr) the ion current is nearly constant for higher ion energies (> 1keV), however, at the lower ion energies a large variation in ion current is observed due to space charge effects. The low current density observed is about 3 x 10⁻⁹ A cm⁻² and extremely low for blistering study purposes. Increased ion current density can be achieved by decreasing the separation distance (to about 4 cms), although electrical breakdown problems are encountered due to the proximity of the vacuum chamber.

2.4 Mass-analysed ion source.

2.4.1 Design and construction

The mass-analysed ion source was developed for the production of
Fig. 2.8 The experimental arrangement of the electrically isolated ion source within the field-ion microscope. The source is aligned directly onto the field-ion specimen.
Fig. 2.9 Operating characteristics of the electrically isolated ion source. The ion current as a function of the helium gas pressures shown below (0.25 cm$^{-2}$ target): $\circ$ $1 \times 10^{-5}$ Torr, $\bullet$ $5 \times 10^{-5}$ Torr, $\times$ $1 \times 10^{-4}$ Torr, $\triangle$ $5 \times 10^{-4}$ Torr.
a particular ion species (i.e. singly charged ions), thus minimising
the effects of any damage which may be produced by different ion
species during ion irradiation studies. The ion source was constructed
with consideration given to the features outlined in section 2.1 for
ion sources operating UHV environments. Consequently the mass
analysed source is almost entirely fabricated from stainless steel
(AISI 304). This also includes the 'top hat' shaped mesh making up
the grid structure. Stainless steel is utilised as a UHV compatible
material, which is non-magnetic and has low outgoing rates. The
filament grid assembly is similar to the previous ion gun (AGI),
although two tungsten filaments (0.025 cm diameter) were incorporated
to increase the electron current density, and arranged in series as
shown in Fig. 2.10(a) and (b). Both filament and grid are surrounded
by a shield which can be negatively biased to repel electrons.

The construction of the ion gun is shown schematically in Fig. 2.11.
The plate electrodes making up the grid, extractor and focusing lens
elements have a small separation to ensure optimum current transfer
(Hurley, (1972)). The plate electrodes are electrically insulated from
one another by sapphire balls. These small spherical beads (0.3 cm
diameter) are placed alternatively in peripheral holes in the plates.
The whole assembly is then spring loaded using tantalum springs on three
supporting M4 studs to reduce any heating effects. This arrangement also
allowed the different electrodes to be interchanged so that the optimum
multiple lens system can be easily achieved.

The ions formed inside the grid assembly will automatically
assume the potential applied to the grid with respect to earth so that
Fig. 2.10 The electron and ion source of the mass-analysed ion gun.

(a) plan view
(b) sectional view
Fig. 2.11 A schematic diagram of the mass-analyzed ion source.
the ion energy can be accurately controlled. The ions are then extracted from the grid by a negatively biased electrode which accelerates the ions into an electrostatic lens focusing system. This system consists of three lenses forming an Einzel lens, the focusing properties of which have been extensively studies by Klemperer (1953), Grivet (1965) and Kanaya et al, (1966). The Einzel lens acts as a converging lens without changing the ion energy in a similar way to that used by Hurley (1972) and focuses the ion beam into the shielded collimating plates of the Wien filter. The ion beam then passes through the filter and after separation is deflected onto the target. The assembled mass-analysed ion gun is illustrated in Fig. 2.12.

The deflection plates consisted of 3cm long insulated nickel plates with a separation of 1.5cm. The whole X, Y assembly is positioned directly after the Wien filter on the 3 supporting studs. Normally each pair of deflecting plates have equal and opposite applied voltages to produce high stability beams (as in the Wien filter). However, in this case one side was earthed and the necessary deflecting voltage applied to the other plate. Suitable polarity allowed the beam to be steered in either direction. The ion beam deflection produced by an applied voltage \( V_x \) on a plate length \( l_x \) with separation \( d_x \) to a target positioned at a distance \( L_x \) is given by the relation:

\[
\Delta x = \frac{V_x \cdot l_x}{2d_x} \tag{2.2}
\]

An identical relationship applies for the y deflector plates, so that the beam steering can be accurately controlled by the applied voltages.
Fig. 2.12 The assembled mass-analysed ion source.
2.4.2 The electrical circuit

The ion source voltages are supplied by a specially insulated control unit which is raised off ground to the ion energy (i.e. isolated). Thus avoiding the problems associated with the large number of high voltage supplies required. The electrical circuit required to operate the ion source, Wien filter and deflector plates is illustrated in Fig. 2.13. The control unit, insulated in a perspex box, is floated at the ion energy which corresponds to the voltage applied to the ion forming grid. The floating or grid voltage is supplied by a 6 mA (0-5kV) power supply type VGMKI (VG Electronics Ltd) which is adjustable to 0.1V with a 0.005% drift and an accuracy of 0.25%. A 6 KV isolating transformer (about 100 KVA) was used to supply the control unit from the mains.

The filament current (0-15A, 0-10V), electron acceleration voltage (0-150V, 50mA), Einzel lens voltage (0-500V) and the extractor voltage (0-350 V) are all supplied by the insulated control unit. The required voltages could be suitably biased by using the relevant polarity of the small power supplies with respect to the grid voltage in the control unit. The filament current is supplied by a variable output transformer with currents of up to 15A and can be monitored together with the voltage across the filament. A transistor feedback amplifier controls the electron emission via the filament current so that stable ion currents can be produced, although oscillatory effects were observed during initial heating of the filament.

The electron emission could be monitored, and is measured between the filament and grid with electron currents of up to 50 mA. Similarly,
Fig. 2.13
A schematic diagram of the electrical circuit required to operate the mass-analysed ion source.

- Deflector plates
- Wien filter
- Einzel lens
- Ion source
- Transformer

Key voltages:
- (0-1 kV)
- 0-650V
- (0-350V)
- (0-150V)
- (0-50 mA)
- (0-5 kV)

To Isolating Transformer
with suitable switching the electron accelerating and extracting voltages together with the Einzel lens voltage can be monitored, allowing suitable adjustment to maximise the ion current on the target. The Wien filter voltages were supplied by a Solatron CRT control unit with a combined positive and negative output of 650 V adjustable to 1V with less than 0.1% drift. The use of opposite polarity voltages produces more stable ion beams (Wilson and Brewer, 1973). The deflector voltages were supplied by Keithley (0-1 keV) power supplies which could be controlled to 0.1V, so that beam steering could be easily achieved. All the electrical connections were made by 10 KV insulated cables via a specially constructed nylon adaptor, ensuring that electrical breakdown did not occur at the 8 pin glass to metal feedthrough to the vacuum chamber.

2.4.3 Wien filter

The concept of utilising crossed electric and magnetic fields was originally developed by Wien (1897) as a particle analyser. The filter has subsequently been used for many purposes involving isotope separation (e.g. Wahlin 1964) and ion implantation (e.g. Wilson and Jamba 1971) as an alternative to the sector magnet. The Wien filter has also been used as a type of mass spectrometer by Jordan (1941) and Holmlid (1973).

The Wien filter essentially consists of crossed electric and magnetic fields, usually achieved by placing non-magnetic deflector plates between the poles of a magnet. Electromagnets can be used, with a variable magnetic field strength dependent on the coil current, although a fixed magnetic field is used in the present study. The type of magnet used is an Alcomax 3 permanent magnet, bakable to 200°C, and
in the form of a C shape, 3 cm in length. The magnetic field strength between the poles was measured using the Hall probe technique and found to be 1.9K Gauss, with a field strength variation of less than 15% between the centre and ends of the magnetic. The magnet is clamped between two earthed collimating plates which have 0.5 cm diameter apertures. The insulated deflector and collimating plates are manufactured from stainless steel. Earthed tubes about 0.8 cm in diameter are used to shield the ion beam from any stray electric and magnetic fields. The whole assembly is positioned directly in front of the last earth aperture of the ion source as shown schematically in Fig. 2.13

The principle of operation of the filter concerns the effect of the perpendicularly inclined electric and magnetic fields on the trajectories of the ions passing through the filter. The application of suitable electric and magnetic fields $E_o$ and $B_o$ respectively, which produce deflecting forces within the device which are balanced for ions with an axial velocity $V_o$. Ogilvie et al (1968) have shown that ions will pass undeviated through the fields when:

$$V_o = \frac{E_o}{B_o}$$ (2.3)

These ions will emerge from the filter undeflected, whereas ions with velocities other than $V_o$ will be deflected away from the axis and become dispersed in space.

The velocity of an ion entering the filter can be determined from the principle of conservation of energy to be:
where $e$ is the ionic charge of an ion of mass number $m$, and $V_I$ is the ion energy referred to the ion source grid voltage. The electric field strength for two parallel plates is simply given by the ratio of the applied voltage $V$ and the separation $d$ between the flat plates, so that

$$E = \frac{V}{d} \quad (2.5)$$

For the fixed magnetic field $B$, the applied voltage necessary to allow an ion of energy $V_I$ undeflected through the filter will thus be given by:

$$V = Bd \left( \frac{2e}{m} V_I \right)^{\frac{1}{2}} \quad (2.6)$$

the applied voltages are recorded in table 2.2 for singly charged hydrogen, helium and neon ions, where relatively large applied voltage differences occur between hydrogen and helium, enabling easier separation.

The ion-optical properties of the Wien filter have been investigated by several authors. Herzog (1934) and Henneburg (1934) have determined the first order ion-optical properties and aberration effects. Wahlin (1964) has examined the astigmatism produced in the ion beam although the effect can be minimised by the use of inhomogeneous electric fields. Seliger (1972) has determined the stigmatic and focusing properties of filters with planar and inclined poles. Ioanoviciu (1973, 1974) has investigated the effects due to fringing fields and field inhomogeneities.
by the use of second order ion-optical calculations. In the present study the Wien filter is only required to separate hydrogen and helium ions, so that no attempt has been made to deduce the different ion-optical parameters (viz. astigmatism and other aberrations), except for the most important factor, which is that of dispersion.

The mass separation capability of the Wien filter essentially depends on the dispersion of ions through the filter. The dispersion $D$, or the distance off-axis that an ion of mass $M_1$ is deflected can be determined (Wilson and Brewer, (1973)) for a given set of experimental parameters by:

$$D = \frac{E}{2V_L} \left( \frac{l^2}{2} + \&L \right) \left( 1 - \sqrt{\frac{M_1}{M_2}} \right)$$  \hspace{1cm} (2.7)

where $l$ is the length of the separator, $L$ the distance to the target, $M_1$ and $M_2$ are the undeflected and deflected ion mass numbers. Under typical conditions of $L = 5$ cms for 1 keV hydrogen and helium ions with $E$ about $4.11 \times 10^4$ V m$^{-1}$, then $D$ is about 2 cms.

The use of small apertures can increase the mass separating capability of the filter (Ioanoviciu and Cuna, (1978)), however this produces a large reduction in ion current, thus large apertures are employed which results in low dispersion. This difficulty is offset by the nature of the ions involved (see Table 2.2) where large differences in applied balancing voltages are apparent for light ions. The voltage difference decreases with mass number so that the dispersion effect will become smaller for large mass numbers.
2.4.4 Ion beam characteristics

A bakable UHV system was used in a series of experiments to determine the characteristics of the mass-analysed ion gun, and hence as shown schematically in Fig. 2.3 representative ion doses could be obtained, similar to those under bombarding conditions. The ion species used were hydrogen, helium and neon, each gas being introduced by means of an MD6 leak valve. After bake-out the system attained a base pressure of about $5 \times 10^{-10}$ Torr. The ion gun filament and grid assembly were thoroughly outgassed by operating the source at low pressures $\sim 1 \times 10^{-8}$ Torr for approximately 5 hours.

The electron emission current is controlled by the filament current, Fig. 2.14 shows this variation for different accelerating voltages, where increasing accelerating voltages produce higher electron emission currents at lower filament currents. Normally an emission current of 10 mA was used to reduce any heating effects and to prolong the filament lifetime. Higher emission currents reduced the filament life especially at high gas pressure ($> 10^{-4}$ Torr) although high emission currents ($> 30$ mA) can be used for a short duration.

The ion current drawn from the source can be increased by optimising the potentials applied to the extractor and Einzel lens. Fig. 2.15 shows the variation of ion current at the target with increasing negative extractor voltage (relative to the grid voltage). A linear region extends to nearly 50 V which then sharply forms a plateau at 200 V. A low extraction voltage is desired to reduce any spread in ion energy which occurs at higher extraction voltages. For this reason the extraction voltage was normally chosen to be 50 V negative to the grid voltage.
The variation of electron emission current with filament current for different electron accelerating voltages.
Fig. 2.15  The total ion current from the mass-analysed ion source as a function of the extractor voltage (negative relative to the grid potential).
The optimum Einzel lens voltage for a given ion energy in the range 500 eV to 2keV for helium ions is shown in Fig. 2.16. Each point corresponds to a maximum ion current at the target, a linear relationship is evident between the lens voltage and the ion energy, similar to that found by Kornelsen (1973) with argon ions.

The ion current density can be determined in-situ by measuring the ion current incident on a target plate of known dimensions. Any secondary electrons produced by the ion beam striking the target material can lead to anomalous ion current measurements. Suppression of the secondary electrons is achieved by applying a positive potential (usually + 70 volts.) to the target. The results are shown for hydrogen, helium and neon ions in Fig. 2.17, (a, b and c) respectively with ion beam energies ranging from 0.5 to 2.5 keV. In these experiments a target plate of over 1 cm² was used to determine the ion current densities. Fig. 2.18 (a and b) show very similar curves for hydrogen and helium ion beams with a target plate of area 0.25 cm². The two values of the ion beam current densities are in close agreement. Figs. 2.17 and 2.18 show that at low pressures (< 10⁻⁴ Torr) the ion current is nearly directly proportional to the gas pressure in agreement with equation (2.1).

At higher pressures (> 10⁻⁴ Torr) the linearity discontinues as positive ion space charge effects lead to ion beam saturation (Lafferty, (1971)). At these higher pressures the ion current reaches a maximum value which is different for each gas and then decreases. To achieve large current densities (up to 1 μA cm⁻²) the ion gun is operated at the pressure producing the maximum ion current. Figs. 2.17 and 2.18 also show that the ion beam current density increases with higher ion.
The optimum applied Einzel lens voltage for a given helium ion energy in the range 0.5 to 2 keV.
Fig. 2.17 (a) Operating characteristics of the mass-analysed ion gun. The ion current as a function of the gas pressure for a range of ion energies (1 cm² target).

(a) hydrogen  
(b) helium  
(c) neon
Fig. 2.17 (b) helium
Fig. 2.17 (c) neon
Fig. 2.18(a) Operating characteristics of the mass-analysed ion gun. The ion current as a function of the gas pressure for a range of ion energies (0.25 cm$^2$ target).

(a) hydrogen
(b) helium
Fig. 2.18(b) helium
beam energies. This is shown separately in Fig. 2.19 for hydrogen, helium and neon ions. The optimum working pressure to achieve maximum ion current is therefore $5 \times 10^{-4}$, $4 \times 10^{-4}$ and $2 \times 10^{-4}$ Torr for hydrogen, helium and neon gases respectively.

The deflection plates can be employed to determine the ion beam profile. The variation of the deflected beam with ion energy is illustrated in Fig. 2.20, where a linear relationship is observed which is in good agreement with equation (2.2). This can be utilised by scanning the beam over the displaced target. The resultant deflecting voltage will therefore correspond to the deflection distance, and is illustrated in Fig. 2.21 (a and b) at target separation distances of 8 cm and 20 cm respectively. The first beam profile (a) shows reasonable symmetry with a second unresolved peak in the form of a hump due to low dispersion in the Wien filter. The second beam profile (b) is symmetrical, and both indicate a beam diameter of approximately 1 cm. All the profiles are of mass-analysed ion beams.

The source and Wien filter operating parameters are recorded in Table 2.1. With the ion source parameters optimised for maximum ion current, the required ion species could be obtained by suitably adjusting the deflecting voltages of the Wien filter. These results for singly charged hydrogen, helium and neon ions are shown in Table 2.2 for different ion energies. The high balancing voltages necessary for the mass separation of the hydrogen ions limited the ion energy to about 1 to 1.5 keV. Similar results were obtained with helium (to 2.5 keV) although neon ions of up to 3 keV were separated, the low dispersion of the filter prevented the observation of well resolved peaks.
The optimum ion beam current density as a function of ion energy for singly charged hydrogen, helium and neon.

Fig. 2.19
Fig. 2.20 The variation of the optimum deflector voltages with ion energy for a maximum ion-current within the field-ion microscope.
Fig. 2.21 Ion beam profiles of 1 keV helium (He$^+$) ions for a source/target separation of: (a) 8 cms (b) 20 cms.
### TABLE 2.1

<table>
<thead>
<tr>
<th>Source operating parameters</th>
<th>Nominal</th>
<th>Max Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Electron Energy $V_e$</td>
<td>150V</td>
<td>200V</td>
</tr>
<tr>
<td>Electron Emission $I_e$</td>
<td>10mA</td>
<td>30mA</td>
</tr>
<tr>
<td>Filament Power $P_f$</td>
<td>20W</td>
<td>30W</td>
</tr>
<tr>
<td>Source Voltage (Ion Energy) $I_I$</td>
<td>200 to 4,000V</td>
<td></td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Wien filter operating parameters</th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Type</td>
<td>Alcomax 3</td>
</tr>
<tr>
<td>Pole Separation</td>
<td>1 x 1 cm</td>
</tr>
<tr>
<td>Magnet Length</td>
<td>3 cms</td>
</tr>
<tr>
<td>Mean Magnetic field</td>
<td>1.89 kG</td>
</tr>
<tr>
<td>Deflector plate dimensions</td>
<td>0.9 X 3 cms</td>
</tr>
<tr>
<td>Separation of deflection plates</td>
<td>0.9 cm</td>
</tr>
<tr>
<td>Deflecting Voltage</td>
<td>0 - 650V</td>
</tr>
<tr>
<td>Ion species</td>
<td>Ion Energy keV</td>
</tr>
<tr>
<td>-------------</td>
<td>----------------</td>
</tr>
<tr>
<td></td>
<td>Experimental</td>
</tr>
<tr>
<td>hydrogen</td>
<td>0.5</td>
</tr>
<tr>
<td></td>
<td>1.0</td>
</tr>
<tr>
<td></td>
<td>1.5</td>
</tr>
<tr>
<td>helium</td>
<td>0.5</td>
</tr>
<tr>
<td></td>
<td>1.0</td>
</tr>
<tr>
<td></td>
<td>1.5</td>
</tr>
<tr>
<td></td>
<td>2.0</td>
</tr>
<tr>
<td></td>
<td>2.5</td>
</tr>
<tr>
<td>neon</td>
<td>0.5</td>
</tr>
<tr>
<td></td>
<td>1.0</td>
</tr>
<tr>
<td></td>
<td>1.5</td>
</tr>
<tr>
<td></td>
<td>2.0</td>
</tr>
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<td></td>
<td>2.5</td>
</tr>
<tr>
<td></td>
<td>3.0</td>
</tr>
</tbody>
</table>
Fig. 2.22 illustrates the variation of the Wien filter voltage against the ion energy for both the experimentally determined and theoretical values. The results are in good agreement with equation (2.16), although it is apparent that low mass ion species, can be more easily separated to form reasonably resolved peaks than the higher mass ion species. This is reflected in mass spectrum of 1.4keV ions which was obtained with a target positioned in front of the ion gun, as illustrated in Fig. 2.23. The Q7 mass spectrometer revealed that the chamber contained a mixture of residual gases; namely, nitrogen, water vapour etc., together with small amounts of neon and helium introduced by means of a leak valve to a pressure of $6 \times 10^{-7}$ Torr. The helium peak can be clearly resolved although the peaks corresponding to the nitrogen, neon etc., have merged to form one large unresolved peak. The mass resolution can be found by comparing the ratio of the Wien filter voltages for different mass peaks, which indicates that $(M/\Delta m \lesssim 5)$. The low dispersion is mainly due to the large diameter collimating apertures and the size of the target, together with the short deflecting length of the filter and drift length of the separated ions. The filter is capable of separating the low mass number ions effectively, although the beam current densities are low, typically about $1 \times 10^{-7}$ A cm$^{-2}$, resulting in large bombardment times to achieve the required doses. Difficulties are involved with higher energy hydrogen ions (>$1.5$ keV) due to large applied electrode voltages due to the fixed magnetic field strength. At higher ion energies (>$3$ keV) breakdown problems are encountered.
Fig. 2.22 The variation of Wien filter voltage for different ion energies of hydrogen, helium and neon.
The mass spectrum of 1.1 keV helium ions produced by the ion source under optimum conditions at a pressure of $6 \times 10^{-7}$ Torr. The presence of residual gases illustrates the low dispersion of the Wien filter.
2.5 High current density ion gun

The ion gun is incorporated in a commercial UHV Varian Auger spectrometer and is normally utilised in a surface cleaning and depth profiling mode for Auger surface analysis. The ion gun is capable of producing ion current densities of more than 300 μA cm⁻² of 3 keV Argon ions are pressures of 5 x 10⁻⁵ Torr, although the gun can operate up to 10⁻⁴ Torr of inert gas pressure. A special feature of this ion bombardment gun is the scanning facility, which allows the ion beam to be scanned over a large sample surface, thus producing a uniform sputtering effect.

The ion gun has an electron impact ionisation source as previously discussed in the preceding sections, together with an electrostatic lens system for accelerating, focusing and deflecting the ion beam. The ions formed in the grid cage are accelerated towards the three element lens by the extractor and then steered onto the target by means of the deflector plates. The ion gun is illustrated in Fig. 2.24, together with the operating voltages which are derived from the beam energy power supply.

The ion energy is variable from 0 to 3 keV with high stability ion beams produced by a regulated feedback control of the electron emission. The emission current during ion-bombardment is typically about 20 to 30 mA, although electron currents of up to 40 mA can be attained. The ion gun has a variable focus control, with a focal range of 4 to 10 cms from the end of the source. The size of the ion beam can be controlled, with a typical value of about 0.25 cm for 3 keV argon ions focused about 5 cms from the end of the source.
The electrical circuit (Warren - model 981-2043) is also indicated. The schematic diagram of the high current density ion source is shown in Figure 2.24.
The ion current density can be determined by means of a probe, consisting of Faraday collector cup positioned behind an earthed molybdenum plate with a 500 μm diameter circular aperture. Secondary electron suppression is achieved with a positive bias (about 90V) applied to the Faraday cup. Figure 2.25 shows the variation of the ion current as a function of the ion energy and gas pressure measured by the probe for helium ions in the energy range 0.1 to 3 keV. Space charge saturation is evident at the lower energies (< 1 keV) which is not observed at higher beam energies. The ion current density can be calculated from a knowledge of the size of the aperture for any ion energy and gas pressure combination, thus defining the bombardment time for ion dose experiments.

2.6 Cathode sputtering.

Cathode sputtering is a convenient 'in-situ' method of bombarding field-ion specimens. Unfortunately this technique does not produce a homogeneous ion energy distribution and suffers a further disadvantage, in that the applied field produces large stresses in the specimen during bombardment. In this technique the FIM specimen is operated in the field emission mode by reversing the polarity of the applied electric field. The specimen thus acts as a field electron emitter in an inert gas (in this case hydrogen or helium) of known pressure.

Ions are formed by electron bombardment and are subsequently attracted to the negatively biased specimen, typically of the order of 1 to 5 keV. The energy of the individual ion depends on its spatial position on formation relative to the emitter. Muller and Tsong (1969) have estimated that there
Fig. 2.25 Operating characteristics of the high current density ion source at an emission current of 16 mA (500 μm diameter aperture target).
is a low possibility of an ion with energy exceeding that corresponding to half the applied emitter voltage striking the imaged portion of the specimen. Higher energy ions can escape the field lines near the emitter and strike the unimaged regions of the specimen.

The determination of the number of ions striking the emitter surface is complex due to the nature of ion formation, since the ion current is superimposed on the field emission current, both processes occurring simultaneously. An approximate value for the ion dose (N) incident on the emitter surface has been determined by Walls et al (1976) as:

\[ N = 7.2 \times 10^{23} \sigma ipt \]  

(2.8)

where \( \sigma \) is the differential ionisation coefficient of the gas species, \( i \) the current measured in the specimen, \( p \) the gas pressure and \( t \) the bombardment time.

Therefore large doses of low-energy hydrogen and helium ions can bombard the surface, although only an approximate order of magnitude value of the dose can be made. The ions produced have a large energy spread and bombard the specimen surface at different incident angles. This technique is therefore useful for a preliminary study but not suitable for precise dose and ion-energy dependent studies.

2.7 Summary

The production of low-energy (< 5keV) hydrogen and helium ion beams has been considered and the important ion beam characteristics of several
ion sources has been investigated. The ion sources generally employ a hot filament type electron excitation source to generate ions with a low-energy spread, although one irradiation technique involves a field emission electron source. The simple ion source (AG1) was found to produce reasonably high ion current densities (up to $10^{-15}$ A.cm$^{-2}$), although a large beam divergence is observed. However, the source has been modified by the inclusion of a collimating aperture which results in higher beam current densities under the experimental conditions for irradiation. A novel type of source incorporating an electrically isolated ion gun (AG1) has been developed, which may enable simultaneous irradiation during the imaging of the FIM specimen. This promising ion source may allow the direct observation of low-energy radiation damage on the surface of the specimen during irradiation.

A mass-analysed ion source has been designed and constructed which has good ion beam characteristics. The 'in-situ' ion source employs both ion-optical focusing and deflection elements. The inclusion of a Wien filter enables the preferred ion species to be chosen (hydrogen and helium) although the filter has an overall low mass resolution and dispersion properties. The mass-analysed ion source however, has the disadvantage of low total current densities ($\sim 1 \mu$A.cm$^{-2}$) which give rise to long irradiation times to achieve the required ion doses on the target (up to $5 \times 10^{17}$ ions. cm$^{-2}$). Furthermore for higher hydrogen ion energies, where large Wien filter applied voltages are needed, electrical breakdown problems may occur.

A high current density commercial ion source with beam energies (0.2 to 3 keV) was found to have good overall beam characteristics including a small spot size ($\sim$ several mm) which could be accurately positioned
by means of focusing and deflecting elements. This source is pre-
dominantly suited for the study of low-energy radiation damage, since
the incident ion beam may be aligned along the axis of the emitter. A
feature not available with the other devices, although the ion source may
not be utilised for 'in-situ' irradiation experiments.

Finally the 'in-situ' field emission indeed irradiation source
or cathode sputtering technique has been considered, where high current
densities of hydrogen and helium ions are produced. Unfortunately this
irradiation source has several disadvantages in that the irradiated
specimen undergoes large stresses due to the applied electric field.
Furthermore, the technique does not produce a homogeneous ion energy
distribution and the ions are incident over the entire specimen surface.
For this reason the total ion dose is difficult to determine precisely.
Therefore the cathode sputtering technique is useful only in a preliminary
study to evaluate the conditions necessary to produce the nucleation and
growth of the voids.
Interpretation of Field-ion Microscopy

3.1 Introduction.

A field-ion micrograph contains a large quantity of information concerning the arrangement of the atomic structure of the specimen under investigation. In this chapter the information contained in field-ion micrographs is examined and the different aspects of the interpretation of micrographs are discussed.

In the first instance the crystallography and information concerning the emitter topography are described. This is followed by a detailed review of the projection geometry of the FIM. The moiré analogy is introduced and a detailed analysis is presented of non-spherical zone plates representing non-spherical planar facets on the emitter surface. New information is deduced from the moiré patterns produced by non-spherical zone plates. The different types of defects (such as point and line defects) observed in the FIM are reviewed, especially those defects which may occur in irradiation damage studies.

The application of the field evaporation process in previous studies is reviewed and an idealised theoretical model is developed and compared with extensive experimental measurements. The measurements correlate the removal rates of the different crystallographic regions of the emitter surface and enable the FIM to be used in semi-quantitative analysis of features present within the specimen.
by means of controlled field evaporation sequences. This is particularly useful in radiation damage studies since this technique permits the measurement of the size and distribution of defects within the irradiated surface.

3.2 Crystallography

In a field-ion micrograph, the image points corresponding to surface atoms, form a complex network of rings, each ring relating to a particular crystallographic plane. Thus forming a one to one correspondence between surface atoms and the image points. The distance between the rings on the emitter surface are determined by the interplanar spacing of the individual planes. The prominence of each plane in the field-ion image also depends on the interplanar spacing (Dreschler and Wolf (1958) and Moore and Ranganathan (1967)).

The most prominent planes in a field-ion micrograph can be indexed by inspection of the pattern displaying a symmetry characteristic of a given orientation of the crystal lattice. The micrograph of a tungsten specimen shown in Fig. 3.1 illustrates the various planes indexed in a manner which is crystallographically consistent. The indexing of planes is usually performed by an assignment of Miller indices to two planes so that high indexed planes can be subsequently defined using the Weiss zone law and relative prominence rule. The higher indexed planes \((h_3k_3l_3)\) can be identified using the lower indexed planes and the Weiss zone law given by:

\[
a (h_1k_1l_1) + \beta (h_2k_2l_2) = (h_3k_3l_3)
\] (3.1)
where \((h_1k_1l_1)\) and \((h_2k_2l_2)\) corresponds to the low index planes, \(\alpha\) and \(\beta\) are integers which can take zero and positive values.

The interplanar spacing for any \((hk\ell)\) plane can be determined from the geometry of cubic crystal, thus the planar step heights \((d_{hk\ell})\) on the emitter surface around any plane can be found from the well known relation:

\[
d_{hk\ell} = \frac{a}{\delta(h^2 + k^2 + \ell^2)^{\frac{1}{2}}}
\]

where \(a\) is the lattice parameter obtained from x-ray diffraction data which for tungsten and molybdenum are 0.316 nm and 0.314 nm respectively. The \(\delta\) term can assume two values for different \(hk\ell\) indices, viz \(\delta = 1\) for \((h+k+\ell)\) being even and \(\delta = 2\) for \((h+k+\ell)\) being odd. The interplanar spacings or step heights for the most prominent low index planes in both tungsten and molybdenum are recorded in Table 3.1

The interplanar angle \(\Theta\) between two planes \((h_1k_1l_1)\) and \((h_2k_2l_2)\) can be similarly determined from the geometry of a cubic crystal using the relationship:

\[
\cos \Theta = \frac{h_1h_2 + k_1k_2 + \ell_1\ell_2}{(h_1^2 + k_1^2 + \ell_1^2)^{\frac{1}{2}}(h_2^2 + k_2^2 + \ell_2^2)^{\frac{1}{2}}}
\]

The interplanar angles for some prominent low index planes are also recorded in Table 3.1 for an [011] orientated emitter, where the apex \((011)\) plane is centred on the tip axis.
TABLE 3.1

Interplanar angles and spacings for tungsten and molybdenum

<table>
<thead>
<tr>
<th>Plane  (hkl)</th>
<th>Interplanar spacing (nm)</th>
<th>Interplanar angle between (hkl) and (011) plane (degrees)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>tungsten</td>
<td>molybdenum</td>
</tr>
<tr>
<td>011</td>
<td>0.223</td>
<td>0.222</td>
</tr>
<tr>
<td>134</td>
<td>0.062</td>
<td>0.061</td>
</tr>
<tr>
<td>123</td>
<td>0.084</td>
<td>0.084</td>
</tr>
<tr>
<td>031</td>
<td>0.10</td>
<td>0.099</td>
</tr>
<tr>
<td>121</td>
<td>0.129</td>
<td>0.128</td>
</tr>
<tr>
<td>213</td>
<td>0.084</td>
<td>0.084</td>
</tr>
<tr>
<td>020</td>
<td>0.158</td>
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<tr>
<td>211</td>
<td>0.129</td>
<td>0.128</td>
</tr>
<tr>
<td>110</td>
<td>0.223</td>
<td>0.222</td>
</tr>
</tbody>
</table>
Zone lines can also be used to identify crystallographic planes on the emitter surface due to the symmetry exhibited in the field-ion micrograph. A zone line corresponds to a line drawn on the micrograph which passes through the centres of all planar regions lying on a crystallographic zone. A zone line [UVW] containing two planes \((h_1k_1l_1)\) and \((h_2k_2l_2)\) can be defined by the relationships:

\[
U:V:W = (k_1l_2 - k_2l_1) : (h_2l_1 - h_1l_2) : (h_1k_2 - h_2k_1)
\]  

(3.4)

Furthermore any plane \((hkt)\) forming a zone line will also satisfy the relation:

\[
hU + kV + lW = 0
\]

(3.5)

The field-ion image usually possesses a central pole defined by the property that all zone lines passing through the pole are straight lines providing a point projection is assumed. The general types of zone lines observed in a field-ion image of tungsten are shown in Fig. 3.1, where the tungsten emitter has an [011] orientation. The zone lines are an important feature of the micrograph since they correspond to planar regions of the emitter surface which have a constant azimuthal angle. This effectively reduces any tip shape analyses to the determination of the step height variation across the emitter surface as a function of the interplanar angle dependent on the chosen type of projection. Thus detailed quantitative analysis of field-ion micrographs is difficult due to the nature of formation of the image which is dependent on knowing the projection relationship.
Fig. 3.1 A helium field-ion micrograph of tungsten with indexed field-ion planes. The principal zone lines are illustrated, where [111] zone lines are solid, [100] zone lines are short dashed, [110] zone lines are long dashed.
3.3 **Emitter topography.**

In a classic paper Dreschler and Wolf (1958) described a method, now most commonly used to determine the local radius of curvature of a field ion emitter. The method consists of counting the number of net plane rings \( n \) between two planes of known angular separation \( \Theta \). Fig. 3.2 illustrates the geometric arrangement, from which it can be seen that if the interplanar spacing or step height \( d_{hkl} \) is known, then the local radius of curvature \( R \) is given by the formula:

\[
R = \frac{nd}{1 - \cos \Theta}
\]  

(3.6)

This is a particularly convenient method to determine the local radius although inaccuracies can occur due to relative phase factors of the planes (Walls (1973)). A further problem can arise due to unresolved atoms in the rings of atoms forming the planes, thus making an estimate of \( n \) difficult. Large inaccuracies can also occur for deviations in tip shape away from the spherical end form. To counteract these effects Dreschler and Wolf (1958) have proposed an 'average tip radius' \( R_{av} \) which is determined by an empirical relationship of the form:

\[
R_{av} = 3.3n_{(011-123)} \text{ nm}
\]  

(3.7)

where \( n_{(011-123)} \) is the number of net plane rings between the (011) and (123) planes on the emitter surface. This method of radius
The geometric model of the field-ion emitter to calculate the local radius of curvature, $R$, by the method of Drechsler and Wolf (1958) - The number $n$, of net plane rings between the centres of two planes ($h_1k_1l_1$) and ($h_2k_2l_2$) of known angular separation. The radius may then be determined from $R = nd/1 - \cos \theta$. 

Fig. 3.2
measurement thus reduces any large variation in local radii which may exist over the emitter surface.

Several other methods of emitter radius measurements are also possible. One of these involves the measurement of the tip voltage applied when imaging a specimen and correlating this voltage with the field necessary for the ionisation of a particular imaging gas. The field $F$ necessary for gas ionisation can then be determined from a relation of the form (Gomer (1961)):

$$F = \frac{V}{kR}$$  \hspace{1cm} (3.8)

where $k$ is a geometrical factor which varies slowly with tip radius $R$. A further method involves the measurement on the micrograph of an arc distance $D$ along a given zone line within a known subtended angle $\theta$ so that the local radius $R$ can be found from the relation:

$$D = R\theta$$  \hspace{1cm} (3.9)

In all the above methods an approximate spherical end form has been implicitly assumed although this may not be the case in practice. Evidence that the spherical end form approximation is inadequate has been obtained by electron microscopy observation of field-ion specimens (Norden and Bowkett (1967)). Fortes (1971) has shown that the surface of a field-ion emitter can be approximately determined from the micrograph. This method essentially assumes a stereographic projection together with a tip shape that can be approximated by a cubic expression.
Bolin et al (1976) however have developed a method for determining tip shapes from field-ion micrographs by the numerical analysis of image points. The use of a data fitting high order polynomial expression together with a computer-graphic technique allows the shape of the emitter surface to be determined. The local radius of the emitter surface has also been determined by the use of the ring counting moiré technique (Southworth and Walls (1977)). Recently Webber and Walls (1978) have developed a ring counting technique which can determine the shape of the emitter through the crystallographic nature of the ring structure on the emitter surface.

3.4 **Projection geometry of the FIM**

A knowledge of the projection geometry of the FIM is essential for a crystallographic analysis of the emitter surface, such as boundary misorientations and accurate plane indexing. Qualitative analysis of topographical features observed in field-ion images can be readily performed, whereas quantitative analysis is a complex problem; and essentially dependent on the type of projection assumed.

The projection in the FIM originates with the imaging ions leaving the ionisation zone close to the specimen surface. The trajectory of the ions are in turn determined by the electric field distribution between the specimen and the screen. The shape of the trajectories over the distance between the emitter and the screen are approximately linear, except near to the specimen (Smith and Walls (1978)). The greatest curvature of the ion trajectory thus occurs in the region close to the emitter. This therefore leads to the concept of an apparent projection point between the image point and the corresponding atom on the emitter.
surface.

The projection geometry of the field-ion image is represented schematically in Fig. 3.3. The field-ion emitter in this case is assumed to have a spherical end form facing a screen separated by a distance T. A given point P on the emitter surface is imaged at the point P'. The ions that form the image will take some non-linear trajectory between the points P and P'. The projection relationship can now be determined by extending the straight line PP' back to the point O, where it joins the axis of the emitter. The location of the point A can then be defined by the distance NR (where N is a non-integer) behind O. The values of N such as infinity, zero and unity thus apply to standard crystallographic projection relationships of orthographic, gnomonic and stereographic respectively.

Müller (1960) originally suggested that the field-ion image projection relationship was gnomonic, although Brenner (1962) proposed that a stereographic projection gave a better approximation. Brandon (1964) considered the image to be found on a plane tangential to the specimen apex, and parallel to the image plane (shown schematically in Fig. 3.4). A point P on the surface of the projection sphere of radius r is projected from the point A, at a separation distance d, onto a projection plane which is tangentially located to the projection sphere. For this idealised case, geometrical considerations show that:

$$d = \frac{r(N+1) \sin \theta}{N+ \cos \theta} \tag{3.10}$$

Brandon subsequently showed, that as \( \theta \) tends to zero, all the projections become equivalent, so that:
Fig. 3.3 The projection geometry of the field-ion image.
Fig. 3.4 The model for the projection relationship proposed by Brandon (1964).
\[ d = r \theta \lim_{\theta \to 0} \] (3.11)

This model was applied to the actual geometry of the FIM by assuming that the separation \( D \) between the central pole of a tungsten micrograph to poles of various planes (of known interplanar angle \( \theta \)) was related to \( d \). Measurements of \( D \) determined for planes of known angle \( \theta \) inferred (by the use of equation 3.11) that the projection based on \( N = 2 \) would give a better approximation than a stereographic (\( N=1 \)) projection.

A similar approach towards defining a projection relationship has been proposed by Newman et al (1967). In this model, illustrated in Fig. 3.5, a projection point is arbitrarily chosen such that the arc \( CP \) subtending an angle \( \theta \) at the centre of a projection sphere, is projected onto a tangent plane similar to that of Brandon's model, so that:

\[ d = r \theta \] (3.12)

The information contained in the projection plane is then projected orthogonally onto a plane screen so that:

\[ D = d = r \theta \] (3.13)

Southworth and Walls (1978) have reviewed the projection geometry of the FIM and concluded that the projection relationship could not be specified by a general \( N \) value, but by an approximate range of \( N \) values. In this model, shown in Fig. 3.6, a spherical field-ion emitter of radius \( R \) is considered as a projection sphere forming a magnified image on a screen separated by a distance \( T \). The model can be used to derive an
Fig. 3.5 The model for the projection relationship proposed by Newman et al (1967).
Fig. 3.6 The model for the projection relationship proposed by Walls and Southworth (1978).
expression relating the projected distance \( D \) with physical imaging parameters, analogous to that derived by Brandon (see equation 3.10) viz:–

\[
D = \frac{MT \sin \theta}{N+\cos \theta}
\]  

(3.14)

where \( M \) is the micrograph (print) magnification factor. Furthermore in the limiting case, the value of \( D \) for \( \theta \) tending to zero is given by:

\[
D = \frac{MT \theta}{N+1} \lim_{\theta \to 0}
\]  

(3.15)

The value of \( D \) therefore varies with each projection and is thus a function of \( N \). It follows that:–

\[
D = k \theta
\]  

(3.16)

where \( k = MT/N+1 \). Numerical calculations based on equation (3.16) have shown that the parameter \( D/MT \) has an approximate linear relationship with \( \theta \) for values of \( N \) between 0.5 and 5 (Southworth and Walls (1978)). Moreover these authors have experimentally determined a value of the projection \( N \) for a tungsten emitter from the gradient \( (k) \) of a plot of \( D \) against \( \theta \). An \( N \) value of 0.83 was determined which has a close agreement with theoretically derived estimates of the projection geometry (Rose (1956)).

In general it is experimentally found that a plot of \( D \) against \( \theta \) gives an approximate linear relationship over a large angular range \( (\theta \approx 60^\circ) \). Typical plots of \( D \) against \( \theta \) for two different tungsten
emitters are shown in Fig. 3.7. The D values correspond to the separation between the central (011) plane and planes of known interplanar angle θ along different zone lines ([111] orientated) and illustrates the approximate linearity between D and θ.

The relationship between D and the tip to screen distance T has been investigated by Lewis et al (1975), where D was measured for a number of plane positions as T was varied over the range 0.5 to 4 cm. It was found that D was a linear function of T suggesting that N remains constant as T was varied. However, for N to be invariant with θ, the angular deviation from the radial projection must increase with θ. This observation has been attributed to a cap focusing effect due to the proximity of the conical emitter shank of the specimen (Southworth and Walls (1978)).

The effect of field evaporation on the relation D = kθ has been studied by Fortes (1971), where it was observed that the constant k remained unchanged during field evaporation. Thus the projection relationship remains unchanged, implying that the value of N is constant even though the emitter radius has increased. This is observed experimentally since no apparent change in D occurs (measured for a given plane) during field evaporation. The observation of constant D values has been explained in terms of imaging ion trajectories, where the ions follow parallel paths with small lateral shifts. Therefore no displacement of the poles are observed, since due to the parallelism, the translation is not magnified (Southworth and Walls (1978)). However, in assymmetrical emitters, Fortes (1971) has shown that a variation of k is observed along a given zone which suggests that N may vary along assymetric zones.
Fig. 3.7 A plot of the linear separation, D, between two known planes against the interplanar angle, $\theta$, determined for two tungsten field-ion micrographs.
The projection geometry of the field-ion emitter can also reveal information regarding the pattern formed in the image by the projection of areal features corresponding to planar regions (or defects) on the emitter surface. Several features of the field-ion image may be simulated by the use of moiré patterns (see for example Walls et al (1973)). The moiré patterns enable quantitative analysis of the field-ion images since it is only a numerical relationship between the ring patterns. However, the shapes of the rings and their interrelationships used in conjunction with projection geometry allow the image patterns to be better understood.

Moiré patterns are produced by the intersection of a spherical projection representing the lattice planes parallel to the image projection and a grid which corresponds to lattice planes perpendicularly included to the projection plane. Thus projection onto a flat plane transforms the intersection into the superposition of a spherical projection and the grid. The surface geometry of the emitter is then assumed to be a hemispherical surface which is intersected by three sets of orthogonally orientated planes. These intersections determine the ledge-step geometry of the surface which is seen projected onto a plane in atomic detail. Thus the geometry of successive rings of plane edge atoms around low index poles in field-ion images resembles the moiré patterns around the low index poles.

The arrangement of the moiré patterns on the emitter surface can be determined analytically by considering the intersection of a three-dimensional grid representing the crystal lattice and the hemispherical emitter surface. Three orthogonal planes form the grid structure and the general axis of the emitter. Thus the grid periodicity may be
represented by equations of the form:—

\[
\begin{align*}
x &= m \, d \\
y &= n \, d \\
z &= p \, d
\end{align*}
\] (3.17)

where \(m\), \(n\) and \(p\) are non-zero positive integers and \(d\) the lattice spacing. The resulting moiré pattern will be determined from the fundamental INDICIAL equation (Oster (1964)):—

\[hm + kn + lp = q\] (3.18)

The \(q\) value corresponds to a non-positive integer sequentially increasing and depends on the values of \(m\), \(n\) and \(p\) for a given plane \((hkl)\). It follows that for the hemisphere of radius \(R\) intersecting with the grid network we have:—

\[hx + ky + pz = Rd(hkl)\] (3.19)

where \(d(hkl)\) is interplanar spacing of the \((hkl)\) plane. The hemispherical surface intersecting the crystal lattice may take spherical co-ordinates so that:—

\[
\begin{align*}
x &= R \cos \Theta \cos \phi \\
y &= R \sin \Theta \sin \phi \\
z &= R \cos \Theta
\end{align*}
\] (3.20)

Thus the pattern on the hemispherical emitter surface will be given by:—
The projection of the moiré pattern on the surface of the emitter is shown schematically in Fig. 3.8. It may be shown that the projected image on the screen will be transformed from the pattern in the emitter by the transformation relations:

\[ \tan \psi = \frac{\sin \Theta}{N + \cos \Theta} \]

and

\[ \rho = (T + (N+1)R) \tan \psi \]

where \( x = \rho \cos \phi \) and \( y = \rho \sin \phi \). It should be noted that these transformation relations are only valid for hemispherical emitter surfaces. Thus the moiré pattern projected onto the screen may be determined by eliminating \( \Theta, \phi, \psi, \) and \( \rho \). The solution of these equations although tedious can be shown to be of the form:

\[
\frac{(hx + ky)}{a} \left[ 1 + N \left( 1 + \frac{(1 - N^2)(x^2 + y^2)}{a^2} \right)^{\frac{1}{2}} \right] = \frac{dq}{R} \left[ 1 + \frac{(x^2 + y^2)}{a^2} \left( 1 - N^2 + N^2 \frac{(x^2 + y^2)}{a^2} \right) \right] \]

\[
2N \frac{(x^2 + y^2)}{a^2} \left( 1 + \frac{(x^2 + y^2)}{a^2} (1 - N^2) \right) \left[ 1 + \frac{N^2(x^2 + y^2)}{a^2} - 1 \right]^{\frac{1}{2}} + \epsilon \left[ \frac{N^2(x^2 + y^2)}{a^2} - 1 \right]
\]

where \( a = (T + (N + 1)R) \). Therefore this equation (3.23) although complex, describes completely the pattern on the screen which is the
Fig. 3.8 The model for the projection geometry of the moiré pattern on the emitter surface.
projected image of the pattern on the emitter. The form of the
projected image pattern (such as circular shaped planar regions)
as a function of the coordinates \( x \) and \( y \) depends on three main
parameters. These include the tip to screen distance \( T \), the
emitter radius \( R \) and the type of projection, characterised by the
\( N \) term.

The two main parameters (\( T \) and \( R \)) are fixed for a given
specimen, thus the projected image pattern will have a strong dependence
on the projection term \( N \). The values of \( N \) can thus be chosen to
correspond with different types of projection (for example \( N = 1 \)
would give a stereographic projection), and the resultant moiré
pattern may be analytically determined. However, although the general
relations are complex, and increase with complexity for higher \( N \) values
(\( N > 1 \)), some general features regarding the image patterns produced on
the screen may be deduced.

The planar facets on a hemispherical emitter correspond to
circles on the emitter surface. These low index planes (such as the
central (011) plane in tungsten) are found to correspond to circles
when gnomonic and stereographic projections are used. For \( N \) values
corresponding to different projections the general equation is complex
(second order curves) and non-spherical in shape; the form of the
second order equations are described in detail in the following section.

Tyson (1964) and Doerr and Ownby (1975) have also investigated the
projection of field-ion images and found that elliptical or even
pear-shaped moiré patterns are formed when circular planes facets are
projected non-stereographically. Moiré analysis of the projected
emitter surface has been reported by Ownby et al (1975) for bcc and
fcc lattices, where similar results were recorded. However, differences between field-ion images and moiré patterns would be expected for non-spherical emitters, and further work is required to elucidate the properties of the projection relationships.

3.5 Image interpretation using the Moiré analogy.

3.5.1 Introduction

The use of moiré analogy enables a better understanding to be developed of the complex relationships between the atoms arranged on the surface of field-ion emitter and the image observed on the screen. The moiré analogy numerically relates the structure of a plane in a field-ion image to that of other planes, thus moiré relationships can be developed to obtain quantitative information of the specimen tip geometry (Southworth and Walls (1977)). The interpretation of field-ion micrographs using the moiré analogy has basically considered the emitter of idealised spherical end form with corresponding circular planar facets.

One interesting feature of the emitter topography which is apparent in a field-ion micrograph, is the relationship between the radius of the apex planar rings $r_n$ and the planar ring number $n$. Fortes and Ralph (1967) considered a spherical emitter of radius $R$ and from the geometry showed that:

$$r_n^2 + (R - nd)^2 = R^2$$

(3.24)

where $d$ is the step height of the apex plane, and providing that $R \gg d$
we have:

\[ r_n = (2nR)^{1/2} \]

Measurements of \( r_n \) from micrographs for different \( n \) values have confirmed that the \( r_n \propto \sqrt{n} \) relationship is valid over small angular distances from the apex plane. This relationship is not observed over the entire emitter surface due to variation of the emitter surface deviating from the idealised spherical end form. However, this relationship forms the basis of the moiré optical analogue (Oster (1964), Vali and Gordon (1973), and Walls (1973)) that exists between the geometry of a planar facet on the emitter surface and that of a binary zone plate.

A moiré pattern is formed by the superposition of two periodic structures, and which for overlapping circular zone plates produces a moiré pattern closely resembling a field-ion image (Walls et al (1973)). Oster (1964) originally suggested that circular regions of the field-ion micrograph, corresponding to high indexed crystallographic planes on the emitter surface, were moiré patterns formed by more prominent planes located on either side. Thus moiré patterns are associated with the configuration of the imaging atoms on the surface of the emitter where the moiré overlap points corresponds to individual atoms.

This approach although yielding quantitative analysis of symmetrical emitters with circular planar facets, does not apply to non-circular planar facets associated with assymmetrical emitters. Furthermore any deviation from symmetry can produce distortions in the image moiré pattern which is mathematically intractable with the patterns
formed by overlapping circular zone plates. In the following section the effect of non-circular moiré patterns produced by elliptical zone plates (EZP) are discussed in detail to deduce further information regarding the topography of the emitter surface.

3.5.2 General moiré theory.

In this section a systematic analysis of the theory of moiré patterns produced by two overlapping EZP's is carried out. The major and minor axis of each ellipse has a square root dependance on the ring number. In a cartesian co-ordinate system an EZP centred at the origin can be generally described by a relation of the form:-

\[
\frac{u^2}{a^2} + \frac{v^2}{b^2} = m + \delta
\]  

(3.26)

where \(a\) and \(b\) are the major and minor axes respectively. The ring number \(m\) is an integer running from 1, 2, ..., and \(\delta\) is the phase of the EZP which can take values \(0 < \delta < 1\). The phase term corresponds to the size of a planar facet which can vary during field evaporation in the moiré analogue. In the general analysis the second EZP is considered to be rotated about some arbitrary point \((x_0, y_0)\) by some angle \(\phi\), as illustrated in Figure 3.9. The moiré patterns located at the points \((a, \beta)\) have principal axes which are orientated at an angle \(\gamma\) to the major axes.

The rotation of the EZP can be determined relative to the origin by the use of the well known transformation co-ordinates shown below:-

\[
u = x \cos \phi - y \sin \phi
\]

\[
v = x \sin \phi + y \cos \phi
\]  

(3.27)
Fig. 3.9  A schematic diagram indicating the location and orientation of the elliptical zone plates on which the general analysis is based.
where $\phi$ is the angle of rotation. Thus the two periodic functions generating the moiré patterns will be of the form:

$$\frac{x^2}{a_1^2} + \frac{y^2}{b_1^2} = m + \delta_1$$

(3.28)

$$\frac{(x-x_o^2)\cos \phi - (y-y_o)\sin \phi}{a^2} + \frac{(x-x_o)\sin \phi + (y-y_o)\cos \phi}{b^2} = n + \delta_2$$

(3.29)

The fundamental relation which defines the moiré patterns produced by overlapping periodic structures is the Indicial equation (Oster et al. (1964)) which for two generating functions is of the form:

$$+ n m + kn = q$$

(3.30)

where $h$ and $k$ are indices taking integer values. The $h$ and $k$ indices define a series of moiré patterns which become increasingly less prominent with higher values of $h$ and $k$. The resultant $q$ will run over some subset of the integers. The positive and negative sign of each term produces additive or subtractive moiré patterns although only additive moiré patterns are considered here. Thus eliminating the ring number $m$ and $n$ from equations 3.29 and 3.30 gives:

$$h \left[ \frac{x^2}{a_1^2} + \frac{y^2}{b_1^2} \right] + k \left[ \frac{(x-x_o^2)\cos \phi - (y-y_o)\sin \phi}{a^2} + \frac{(x-x_o)\sin \phi + (y-y_o)\cos \phi}{b^2} \right] = q + h\delta_1 + k\delta_2$$

(3.31)
the above equation can be rearranged to give an expression of the form:

$$A x^2 + Bxy + C y^2 + 2Dx + 2Ey + F = 0$$

(3.31)

This is a general equation of the second degree and represents a conic section, thus the moiré pattern produced is a system of second order curves. The values of each coefficient is determined by:

$$A = \frac{h}{a_1^2} + k \left( \frac{\cos^2 \phi}{a_2^2} + \frac{\sin^2 \phi}{b_2^2} \right)$$

(3.32)

$$B = k \cos \phi \sin \phi \left( \frac{1}{b_2^2} - \frac{1}{a_2^2} \right)$$

(3.33)

$$C = \frac{h}{b_1^2} + k \left( \frac{\sin^2 \phi}{a_2^2} + \frac{\cos^2 \phi}{b_2^2} \right)$$

(3.34)

$$D = k \left( y_0 \sin \phi \cos \phi \left( \frac{1}{a_2^2} - \frac{1}{b_2^2} \right) - x_0 \left( \frac{\cos^2 \phi}{a_2^2} + \frac{\sin^2 \phi}{b_2^2} \right) \right)$$

(3.35)

$$E = k \left( x_0 \sin \phi \cos \phi \left( \frac{1}{a_2^2} - \frac{1}{b_2^2} \right) - y_0 \left( \frac{\sin^2 \phi}{a_2^2} + \frac{\cos^2 \phi}{b_2^2} \right) \right)$$

(3.36)

$$F = k \left( x_0 y_0 \sin \phi \cos \phi \left( \frac{1}{b_2^2} - \frac{1}{a_2^2} \right) + k x_0 \frac{\cos^2 \phi}{a_2^2} + \frac{\sin^2 \phi}{b_2^2} \right)$$

(3.37)

$$+ k y_0 \frac{\sin^2 \phi}{a_2^2} + \frac{\cos^2 \phi}{b_2^2} - q - h \delta_1 - k \delta_2$$

(3.38)
The general equation describes the moiré pattern produced by the overlapping EZP's and in the following analysis the patterns will be characterised in terms of the coefficients of the general equation. Thus the shape, location, size, orientation and phase of each of the second order curves determined by the h,k values can be examined. Furthermore, if more than two overlapping EZP's of different locations are considered, only the complexity of the coefficients increases, and the general methods and formulae in the analysis are still applicable.

In the following analysis two specific examples are used to illustrate the general trends. The examples consist of an overlapping circular zone plate (CZP) and an EZP, in the second example two overlapping EZP's are considered. The analysis is limited to the region between the two zone plates, since this is the main region of interest in the moiré analogy to field-ion micrographs.

3.5.2.1 Shape of the moiré pattern.

The general form of the moiré pattern can be determined from the evaluation of the discriminant \( \Delta \) of the general second order equation (3.31) given by Moser et al (1975) as:-

\[
\Delta = AC - B^2
\]

where

\[
\Delta = \left( \frac{h}{a_1} + k \left( \frac{\cos^2 \phi}{a_2} + \frac{\sin^2 \phi}{b_2} \right) \right) \left( \frac{h}{b_1} + k \left( \frac{\sin^2 \phi}{a_2} + \frac{\cos^2 \phi}{b_2} \right) \right)
\]

\[- k^2 \cos \phi \sin^2 \phi \left( \frac{1}{b_2} - \frac{1}{a_2} \right)^2 \quad (3.39)\]
There are three possible cases where different forms of the moiré patterns can be deduced, these are when:

\[ \Delta > 0 \]

- **ellipses**
- **parabolas**
- **hyperbolas**

Thus the forms of the moiré patterns can be deduced for any values of the coefficient in the general equation. For a CZP at the origin and an EZP located at \((x_0, y_0)\) for which \(a_1 = b_1 = r\), we have:

\[
\frac{h}{r^2} + k \left( \frac{\cos^2 \phi}{a_1^2} + \frac{\sin^2 \phi}{b_1^2} \right) \left( \frac{h}{r^2} + k \left( \frac{\sin^2 \phi}{a_2^2} + \frac{\cos^2 \phi}{b_2^2} \right) \right) > k^2 \sin^2 \phi \cos^2 \phi \left( \frac{1}{b_2^2} - \frac{1}{a_2^2} \right)^2
\]

Thus elliptical moiré patterns are formed for \(\phi = n\pi\) where \(n = 0, 1, 2, \ldots\) etc and for any values of \(\phi\) from 0 to \(2\pi\). A similar analysis for two EZP's shows that:

\[
\frac{h}{a_1^2} + k \left( \frac{-\cos^2 \phi}{a_1^2} + \frac{\sin^2 \phi}{b_1^2} \right) \left( \frac{h}{b_1^2} + k \left( \frac{\sin^2 \phi}{a_2^2} + \frac{\cos^2 \phi}{b_2^2} \right) \right) > k^2 \cos^2 \phi \sin^2 \phi \left( \frac{1}{b_2^2} - \frac{1}{a_2^2} \right)^2
\]

again elliptical moiré patterns are produced for all \(\phi\) and \(h, k\) values.

### 3.5.2.2 Location

The position of the moiré pattern can be determined if parallel axes are transferred to the origin of the second order curve \((a, \beta)\).
This transforms the general equation (3.31) to the form:

\[ A (x + \alpha)^2 + 2B (x + \alpha)(y + \beta) + C (y + \beta)^2 + 2D (x + \alpha) + 2E (y + \beta) + F_1 = 0 \]  

(3.43)

where \( F_1 \) are \( A\alpha^2 + 2B\alpha\beta + C\beta^2 + 2D\alpha + 2E\beta + F \)

If \( \alpha \) and \( \beta \) are now chosen so that:

\[ A\alpha + B\beta + D = 0 \]  

(3.44)

and

\[ B\alpha + C\beta + E = 0 \]  

(3.45)

Equation (3.43) then becomes:

\[ A x^2 + 2B xy + C y^2 + F_1 = 0 \]  

(3.46)

and solving equations 3.44 and 3.45 gives:

\[ \alpha = \frac{BE - CD}{BE - CD} = \frac{BD - AE}{AC - B^2} = \frac{1}{\Delta} \]

thus the values of \( \alpha \) and \( \beta \) are given by:

\[ \alpha = \frac{BE - CD}{\Delta} \text{ and } \beta = \frac{BD - AE}{\Delta} \]  

(3.47)

The values of \( \alpha \) and \( \beta \) therefore determine the positions of the
centre of the second order curves forming the particular \((h,k)\) moiré pattern.

The important consequence of equation 3.47 is that the moiré patterns will not have the same location \((\alpha,\beta)\). Thus each value of \(\alpha\) and \(\beta\) will depend only on the values of \(h\) and \(k\) for a given set of parameters (viz., \(a, b, x_0, y_0\) and \(\phi\)). Only moiré patterns with the same indexing parameters \((h = k)\) will have identical locations. Therefore, the centre of each moiré pattern will not be linearly related to the origin as was originally suggested by Moser et al (1975). The relationship between the two location points \((\alpha, \beta)\) can be determined by equating relations (3.46) and (3.47) so that:

\[
\beta = \frac{a(DB-AE)}{(BE-CD)} \tag{3.48}
\]

substitution of the values of the coefficients in the above equation shows that a complex non-linear relationship exists between \(\alpha\) and \(\beta\) primarily dependent on the values of \(h\) and \(k\) for a given set of parameters. The equations determining the locations for the moiré patterns produced by overlapping CZP and EZP and two EZP's are complex. However, the use of computer calculations allows the exact location of each moiré pattern formed to be determined. This has been performed with \(h, k\) values in the range 1 to 4 for the locations of the moiré patterns with different values of \(\phi\) from 0 to 180°.

For an overlapping CZP and EZP located at \((10, 10)\) shown in Fig. 3.10, a symmetrical location of the moiré pattern is observed as the EZP is rotated through 180° about the line joining both origins. The
Fig. 3.10 The location of the moiré patterns resulting from the overlapping CZP and EZP as a function of rotation angle, $\phi$. The (1,1) moiré pattern is illustrated.
reverse occurs on further rotation to $360^\circ$ as the moiré patterns retain their original positions. A linear relationship is observed for $\phi = 45^\circ$ for the particular values chosen (viz., $a = b = 1; a_2 = 2$ and $b_2 = 1$).

In the case of two overlapping EZP's, one located at the origin the other at $(10, 0)$ and shown in Fig. 3.11 a symmetrical location of the moiré patterns occurs for $\phi$ the range $0$ to $180^\circ$ and $180^\circ$ to $360^\circ$. However changes in location occur with rotation angle $\phi$ as shown in Fig. 3.11, where a high degree of symmetry is observed about the x axis. The maximum effect occurring at $\phi = 90^\circ$ where the location of the major moiré patterns shift towards the rotating EZP, a fact which is observed experimentally. The only linear relationship of the location of the moiré pattern with $\phi$ is observed at $\phi = 0$ and $180^\circ$ where moiré patterns exist on the ordinate axis. However, the positions of the moiré patterns are non-coincident, a fact which is also observed experimentally. This is due to the effects of the ZP rings of different sizes interacting to produce different locations. Thus the angular orientations of the overlapping ZP's have a direct influence on the location of the resultant moiré patterns.

3.5.2.3. Orientation

The series of second order curves representing the moiré pattern about the points $(a, b)$ have principal axes which are orientated at an angle $\gamma$ to the major axis as illustrated previously in Fig. 3.9. The determination of the angle $\gamma$ involves the rotation of the principal axis of the conic section by the same angle $\gamma$ so that the form of the equation
Fig. 3.11 The location of the moiré patterns resulting from the two overlapping EZP's as a function of the rotation angle, \( \phi \). The (1,1) moiré pattern is illustrated.
governing the shape of the pattern reduces to a simpler form. Thus transformation relations similar to equation 3.27 are used which remove any cross product \((X \cdot Y)\) terms so that equation 3.46 becomes:

\[
A (x \cos \gamma - y \sin \gamma)^2 + 2B (x \cos \gamma - y \sin \gamma) (x \sin \gamma + y \cos \gamma) + C (x \sin \gamma + y \cos \gamma)^2 + F_1 = 0
\]

thus we have:

\[
A_1 x^2 + 2B_1 xy + C_1 y^2 + F_1 = 0 \tag{3.49}
\]

where

\[
B_1 = B (\cos^2 \gamma - \sin^2 \gamma) - (A - C) \cos \gamma \sin \gamma
\]

If \(\gamma\) is now chosen so that \(B_1 = 0\), then:

\[
B (\cos^2 \gamma - \sin^2 \gamma) = (A - C) \cos \gamma \sin \gamma \tag{3.50}
\]

This is equivalent to rotating the principal axes parallel to the co-ordinate axis, thus the relative angle of rotation corresponding to the orientation of the moiré pattern can be found from:

\[
\tan 2 \gamma = \frac{2B}{A - C} \tag{3.51}
\]

so that:

\[
\tan 2 \gamma = \frac{h k \cos \phi \sin \phi \left( \frac{1}{b_2^2} - \frac{1}{a_2^2} \right)}{h \left( \frac{1}{a_1^2} - \frac{1}{b_1^2} \right) + k \left[ \cos^2 \phi \left( \frac{1}{a_2^2} - \frac{1}{b_2^2} \right) + \sin^2 \phi \left( \frac{1}{b_2^2} - \frac{1}{a_2^2} \right) \right]} \tag{3.52}
\]
Thus the orientation of the moiré pattern only depends on the indices \( h \) and \( k \) for a given set of dimensions of the overlapping elliptical zone plates. For a CZP and EZP combination \( a_1 = b_1 \), thus equation 3.52 reduces to:

\[
\tan^2 \gamma = \frac{\sin^2 \phi}{2 \sin^2 \phi - 1}
\]

(3.53)

and so becomes independent of any \( h, k \) values. The results are plotted graphically in Fig. 3.12, where all the moiré patterns have the same orientation for different rotation values.

The orientation of the moiré pattern produced by two EZP's is described by equation (3.52) and has a complex form. The orientation angle \( \gamma \) is dependent primarily on the indices \( h \) and \( k \) for given values of size of the major and minor axes of the EZP's.

The actual orientation is symmetrical about the rotation angle \( \phi \) for \( \phi = 90^\circ \) but each moiré pattern follows a different orientation angle \( \gamma \), which has a maximum value at different values of \( \phi \) as shown in Fig. 3.13, for the three main \( h, k \) values of two identical EZP's. More complex relationships are produced for non-identical EZP which could lead to assymmetrical forms of variation of orientation angle \( \gamma \) with rotation angles \( \phi \).

3.5.2.4 Size of a moiré pattern

The size of the moiré pattern for a given \( h, k \) value can be determined from equation (3.52), by the removal of the cross term \((XY)\), and usually achieved by taking \( B_1 = 0 \) to determine the orientation of
Fig. 3.12 The orientation angle, $\gamma$, of the principal (1,1) moiré pattern as a function of rotation angle, $\phi$, for an overlapping CZP and EZP. The (h,k) moiré patterns have a similar orientation as a function of rotation angle.
Fig. 3.13  The orientation angle, $\gamma$, of several $(h,k)$ moiré patterns as a function of the rotation angle, $\phi$, for two overlapping EZP's.
the moiré pattern. Thus equation 3.49 reduces to the form:

$$A_1 x^2 + C_1 y^2 = - F_1$$

(3.53)

where

$$A_1 = A \cos^2 \gamma + C \sin^2 \gamma + 2B \cos \gamma \sin \gamma$$

$$C_1 = A \sin^2 \gamma + C \cos^2 \gamma - 2B \sin \gamma \cos \gamma$$

$$F_1 = A \alpha^2 + 2B \alpha \beta + C \beta^2 + 2D \alpha + 2E \beta + F$$

Equation (3.53) therefore describes the moiré patterns and together with the conditions (equation (3.40)), which determines the shape of the pattern, allows the size of the moiré pattern to be determined from the coefficients $A_1$ and $C_1$ shown below:

$$A_1 \left( \frac{a_1}{a_2} + k \left( \frac{\cos^2 \phi}{a_2^2} + \frac{\sin^2 \phi}{b_2^2} \right) \right) \cos^2 \gamma + \frac{h}{b_1} \left( \frac{\sin^2 \phi}{a_2^2} + \frac{\cos^2 \phi}{b_2^2} \right) \sin^2 \gamma$$

$$+ 2k \cos \phi \sin \phi \left( \frac{1}{b_2} - \frac{1}{a_2} \right) \sin \gamma \cos \gamma$$

(3.54)

$$C_1 = \left( \frac{h}{a_1} + k \left( \frac{\cos^2 \phi}{a_2^2} + \frac{\sin^2 \phi}{b_2^2} \right) \right) \sin^2 \gamma + \frac{h}{b_1} \left( \frac{\sin^2 \phi}{a_2^2} + \frac{\cos^2 \phi}{b_2^2} \right) \cos^2 \gamma$$

$$- 2k \cos \phi \sin \phi \left( \frac{1}{b_2} - \frac{1}{a_2} \right) \sin \gamma \cos \gamma$$

(3.55)

It is clear from the above relations that the size of the moiré
pattern is independent of the relative positions \((x_o, y_o)\) of the two EZP's. However for a given set of parameters a complex relationship exists between the \(h, k\) indices and the size of the moiré pattern.

For the overlapping CZP and EZP the size of the resultant moiré pattern for \(a_1 = b_1 = r\) is given by:

\[
A_1 = \frac{h}{r^2} + k \left[ \left( \frac{\cos^2 \phi}{a_2^2} + \frac{\sin^2 \phi}{b_2^2} \right) \cos^2 \gamma + \left( \frac{\sin^2 \phi}{a_2^2} + \frac{\cos^2 \phi}{b_2^2} \right) \sin^2 \gamma \right. \\
\left. + \cos \phi \sin \phi \left( \frac{1}{b_2^2} - \frac{1}{a_2^2} \right) \sin^2 \gamma \right]
\]

and

\[
C_1 = \frac{h}{r^2} + k \left[ \left( \frac{\cos^2 \phi}{a_2^2} + \frac{\sin^2 \phi}{b_2^2} \right) \sin^2 \gamma + \left( \frac{\sin^2 \phi}{a_2^2} + \frac{\cos^2 \phi}{b_2^2} \right) \cos^2 \gamma \right. \\
\left. - \cos \phi \sin \phi \left( \frac{1}{b_2^2} - \frac{1}{a_2^2} \right) \sin^2 \gamma \right]
\]

Both \(A_1\) and \(C_1\) refer to the major and minor axes of an elliptical zone plate representing the moiré pattern, which corresponds to the first ring size. The subsequent ring sizes will follow the \(r_n = n \frac{\pi}{2}\) relationship along both the axes.

The size of the moiré pattern produced by two overlapping EZP's is given by equations (3.56) and (3.57), where the size is a complex function of the indices \(h\) and \(k\), the orientation angle \(\gamma\) and rotation angle \(\phi\). One interesting feature is the special case when, \(\phi = 0\) and thus \(\gamma = 0\), then for identical EZP's equations and reduce to the form:
Thus the size of the elliptical moiré patterns is a simple fraction of the size of one main EZP, dependent on the h, k values chosen. For example the (1,1) moiré pattern will have a size equal to half the original generating EZP size and the moiré patterns on each side (viz (2,1) and (1,2) will have identical sizes, namely one third that of the original. In this way a whole series of moiré patterns of decreasing size will be observed between each main EZP.

3.5.2.5 Phase

The phase of the moiré pattern is a complex function, but dependent on the shape of the pattern can take values from zero to the maximum size of the particular pattern. The phase $\delta_m$ of the moiré pattern can be determined by comparison of equation 3.53 to be:

$$\delta_m = A\alpha^2 + 2B\alpha\beta + C\beta^2 + 2D\alpha + 2E\beta + F$$

The equation above is a general equation describing the phase of a moiré pattern and substitution of suitable values of the coefficients, location terms ($\alpha, \beta$) and the phases of the EZP's allows the exact value to be determined. The resultant generalised equation is complex and has not been developed further in this investigation.

3.5.3 Discussion

The work performed in this investigation of the effects of non-
circular overlapping zone plates has established that the resultant 
moiré patterns formed are non-simply related to the position and 
orientation of the generating zone plates. However, the generalised 
equations characterising the moiré patterns have been developed in 
terms of pattern size and shape, location, orientation and phase. 
Several important features have emerged indicating that the moiré 
patterns produced by non-circular zone plates are not generally located 
on a line drawn between the two centres of the zone plates as had been 
previously supposed. Furthermore the location and orientation of the 
moiré pattern is dependent on the indices \((h,k)\) values) of a given 
moiré pattern. The shape of the pattern will also be dependent on 
the mathematical form of the generating zone plates. The two examples 
chosen of overlapping zone plates (viz, CZP and EZP, EZP and EZP) have 
shown these salient features, which could be experimentally measured 
(eg. location, orientation etc) with further work and compared with 
the moiré theory.

The main application of this work, and the reason for the investi-
gation, is the interpretation of field-ion micrographs of emitter surfaces 
having non-circular planes. The theoretical moiré simulation of non-
circular planes developed in this section together with experimental 
measurements on well defined overlapping zone plates may be used to 
determine the location of severely distorted planes (eg. surface dis-
tortion due to radiation damage). This application would be particularly 
useful in field evaporation studies of damaged surfaces due to ion-
bombardment, where the location of the damaged plane on the micrograph 
would enable the precise depth of damage to be established.
3.6 Defect Observation in the FIM

The FIM is a useful tool as a direct method of studying individual point defects and line defects such as dislocations and grain boundaries. The FIM has potentially greater advantages over other techniques such as resistivity methods and electron microscopy which are unable to distinguish unambiguously the type of defect present in the specimen. However, due to the rigorous imaging conditions present in the FIM, the possibility of any surface artefacts has to be considered. Thus, although vacant lattice sites may represent genuine vacancies and bright spots indicate interstitials, considerable care must be employed to distinguish between these and artefact point defects.

Several methods have been employed to produce point defects in field-ion specimens, vacancies have been introduced by irradiation, by heating and subsequent quenching (Schultz (1964)). Substitutional and interstitial impurity atoms may be studied by the use impure material of different impurity content.

Vacancies are detected in the FIM by the contrast observed in the image usually in the form of a dark spot on an atomically resolved plane or on planar ring edges. The observation of vacancies over the entire emitter surface is difficult since the vacancy concentration is dependent on the imaging field (Wald (1963)). The effect of the field stresses in the production of vacancies was suggested by Müller (1965), where vacancies were spontaneously produced by the volume expansion of the emitter during imaging. However, Bowkett and Ralph (1969) found
that large numbers of vacancies were not induced in tungsten by field imaging stresses.

In a study of the vacancy concentration in tungsten Attardo and Galligan (1966) found that the observed fraction of vacant sites in annealed tungsten specimens was less than $10^{-5}$, indicating that tungsten was a suitable material for vacancy studies. Considerable effort has been employed in counting vacancies in tungsten specimens, particularly on the (211) plane edges which are considered the most suitable regions for these measurements. Even so, the number of vacancies counted has been found to be extremely subjective even though the number of artefacts in tungsten is considered to be low (Bowkett and Smith (1970)).

Interstitials are manifested on field-ion images as additional bright spots on the surface of the emitter in unusual imaging atom positions. Müller (1959) suggested that the bright spot contrast arose from sub-surface interstitial atoms influencing the surface topography, and not directly by interstitial atoms situated on the surface; an interpretation adopted by many authors. Moore (1962), using the 'thin shell' computer model in field-ion image simulation found that small atomic displacements resulted in positional changes of the imaging atoms. Thus surface atoms appear to be sensitive to small atomic displacements, giving rise to visible effects.

In ion-bombardment studies several types of interstitials may be encountered. An interstitial atom can be formed by the repositioning of a normal lattice atom due to an atomic collision event, and is re-
ferred to as a self interstitial atom (S.I.A.). Interstitials can also be formed by inert gas atoms trapped in vacant lattice sites or interstitial positions. These trapped gas atoms may cause sufficient local lattice distortion to produce S.I.A.s in the surrounding lattice structure. An extensive study of the S.I.A. contrast observed in the field-ion microscope has been performed by Seidman and Lie (1972) and also reviewed by Seidman (1973). It was found that extra bright spot contrast may be caused by S.I.A.s several atomic layers beneath the surface and that bright spots also occurred in normal lattice positions. Furthermore, they proposed that S.I.A.s may be preferentially field evaporated to leave vacant lattice sites.

Bright spot contrast can also be exhibited by impurities in the specimen (Fortes (1968)), thus self interstitials may give rise to the same type of contrast as those of impurity atoms. Distinction between S.I.A. and impurity atoms can be achieved using an "Atom-Probe" field-ion microscope (Paintz (1973)). The observation and interpretation of interstitials is further complicated by the zone decoration phenomenon, a striking feature observed as bright image spots arranged along a particular crystallographic zone in the image. In tungsten this occurs on the [100] zone by single or multiple rows of bright atom spots marking the edges of (011) net plane edges. The spots are wide and bright adjacent to the (011) plane, but decrease in brightness to give normal contrast between the (021) and (031) planes. The brightness of the zone decorating atoms has been attributed to atoms occupying protruding low co-ordination number sites (Bowkett and Smith (1970)) not usually associated with actual lattice sites.
The observation of dislocations in a field-ion micrograph is usually manifest as a spiral-like structure which occurs on planar regions of the emitter surface. In fcc and bcc metals, a perfect dislocation intersecting the emitter surface at a pole \((hk\bar{l})\) produces a spiral with a pitch of an integer number of \((hk\bar{l})\) spacings. However, for non-integer values of pitch, partial dislocations may be formed; especially those associated with stacking faults, where stepped spirals may be produced. Generally the detailed analysis of dislocations is complex and difficult to be unambiguously interpreted. A detailed review of this subject has been given by Bowkett and Smith (1970).

3.7 Field Evaporation

3.7.1 Introduction

The phenomenological theories dealing with field evaporation have been discussed previously and in this section the physical parameters controlling the rate of field evaporation are investigated. Previous theories dealing with field evaporation are discussed and a simple semi-quantitative theory is described which enables the depth of damage of the irradiated specimens to be determined.

The field evaporation phenomena is manifested as a collapsing of rings around the major crystallographic planes. Each time a ring of atoms forming a plane disappears, a thickness of material equal to the inter-planar spacing associated with that plane will have been removed. The rate of evaporation or removal of planes is not constant over the
emitter surface for a stable end form. Brandon (1966) found that the evaporation rate varied over the surface of the specimen, with a maximum usually at the apex and falling to zero at the extremes of the image. The low index planes tend to evaporate initially due to the kink site atoms which have the lowest activation energy required to field evaporate. The higher indexed planes located around the apex are then observed to collapse and disappear as successive close packed planes are field evaporated.

Brandon (1965) considered the field evaporation of a tungsten emitter and the dependence of the evaporation rate on the applied field. The rate of evaporation of the emitter was determined in terms of an evaporation parameter developed from the image force theory of field evaporation. The measurement of the time for each plane to evaporate allows the evaporation rate to be found. The evaporation parameter was evaluated by measurements of the evaporation rate either as a function of the applied voltage at constant emitter radius, or of the tip radius at constant voltage.

In the determination of the tip radius, two simple approximations for the shape of the tip were used to relate the number of planes removed to the tip radius during field evaporation. The first tip shape considered consisted of a hemisphere located on a cone of half angle, \( \psi \), with an emitter radius given by:

\[
R = \sin \psi \left( N_d + R_o \right)
\]  

(3.59)

where \( n_d \) is the number of apex planes removed of inter-planar spacing \( d \).
The initial tip radius is denoted by \( R_0 \). The second tip shape was that of a paraboloid with an inscribed hemisphere, in which case the emitter radius is given by:

\[
R^2 = a^2 + 2a \text{Nd} \tag{3.60}
\]

where \( a/2 \) is the distance of the focus from the apex of the paraboloid. The use of the approximate tip shapes produced reasonable agreement for the comparison of the rate of evaporation with the change in evaporation voltage. However, a discrepancy was observed between the experimentally measured field dependence of the evaporation and the theoretically calculated value (Brandon (1964)).

A simple model relating the amount of material removed from the apex by field evaporation as a function of the emitter radius has been developed by Bowkett and Smith (1970). The emitter is considered as a spherical cap on a truncated cone. The cone has a tip taper angle \( \alpha \) with the emitter of initial and final radius \( R_1 \) and \( R_2 \) respectively. Removal of \( N \) apex planes of inter-planar spacing \( d \) during evaporation produces an increasing emitter radius given by:

\[
N_d = \frac{(R_2 - R_1)(1 - \sin \alpha)}{\sin \alpha} \tag{3.61}
\]

Thus for a constant tip taper angle the number of apex planes removed is simply related to the initial and final tip radii. An identical relationship has been deduced by Tsong and Müller (1970) to determine the electric field on the idealised emitter surface during field evaporation.
at constant voltage to account for the change in emitter radius.

Analysis of tungsten micrographs with about 200 apex planes removed by field evaporation indicated a linear increase in tip radius and gave a value of the tip taper angle of 23°, (Bowkett and Smith (1970)). The tip radius measurements were performed using the Dreschler and Wolf relation. Therefore this method could be used to determine the tip taper angle simply from field evaporation data and compared with actual tip shapes using electron microscopy.

A similar argument has been developed by Bowkett and Smith (1970) for the analysis of crystallographic line defects such as grain boundaries and dislocations. A number of simplifying assumptions are made, which include a spherical tip shape and stereographic projection, both invariant during field evaporation with an equal thickness of material removed from all areas of the emitter surface. However as it will be shown later, these assumptions are not strictly correct, in that the amount of material removed has a cosine dependence with the tip axis. Moreover subsequent changes in the emitter radii during field evaporation can be correspondingly large for the removal of significant numbers of apex planes. The model is only generally applicable for a limited field evaporation sequence where a small amount of material is removed from the emitter surface (eg. analysis of dislocations).

The variation of emitter shape with field evaporation has been considered by Fortes (1971). The thickness of material, \( t \), removed at each plane is related to the interplanar spacing by considering two parallel planes tangential to the surface. Assuming no change in tip
size and shape during field evaporation, the amount of material removed at any pole on the emitter surface can be related to the amount of material removed from the apex, \( n \). For a known interplanar angle \( \Theta \), Fortes showed that:

\[
t = n \cos \Theta
\]

(3.62)

and further deduced that the relationship was valid for any tip shape.

Thus the accurate determination of the number of planes removed from each pole of the emitter can be achieved by the use of small evaporation increments between successive micrographs. Subsequent analysis of the micrographs revealed a close agreement with equation (3.62) for both tungsten and iridium specimens for large values of \( \Theta \) (up to 70°).

A similar analysis of field evaporation has been considered by Faulkner (1969) where the specimen consists of a spherical cap situated on a cylindrical shank. Field evaporation of \( N \), layers from a plane of known interplanar spacing, \( d \), some angle \( \Theta \) away from the centre of projection is assumed to be equivalent to removing \( Nd/\cos \Theta \) layers from a plane at the centre of the projection.

The use of field evaporation in analysis of particle sizes has been developed by Schwartz and Ralph (1969), who considered the removal of successive layers of an emitter with a small tip taper angle. They suggested that the evaporation of a small amount of material from the emitter surface generated a new surface almost identical to the original. In this analysis it was assumed that the emitter radius did not vary significantly during the field evaporation sequence. Furthermore within
the volume studied, no anomalous denuded zones were present, but a reasonable sample of the bulk micro-structure.

Thus during field evaporation the tip surface will recede along the tip axis a distance $\Delta L$. The amount of material removed at a point on the surface where the normal is at an angle $\Theta$ to the tip axis is assumed to have a cosine dependence. The number of rings, $\Delta n$, which are seen to collapse at a predetermined counting plane is then given by the relation:

$$\Delta n = \frac{\Delta t}{d} = \frac{\Delta L \cos \Theta}{d} \quad \text{(3.63)}$$

where $\Delta t$ is the amount of material removed normal to the surface of the counting plane of interplanar spacing $d$.

3.7.2. Field evaporation: theory

A simple idealised model is developed in this section to establish a general relationship between the amount of material removed from the apex plane and other planar regions of the emitter surface. The model is based on the concept of differing planar removal rates relative to the apex planar removal rate during field evaporation. In the first instance a spherical emitter end form is assumed and analysed, a subsequent model determines the effect of non-spherically shaped emitters.

A simple model is considered, consisting of a spherical cap situated on a truncated cone of tip taper angle $\psi$. During field evaporation the emitter end form becomes enlarged due to the removal
of atoms from the surface to produce a higher radius spherical tip as illustrated in Fig. 3.14. The amount of material removed from any planar region \( N_{hkl} \) can be characterised by the number of apex planes ((011) planes in tungsten and molybdenum) removed during field evaporation. In Fig. 3.14 (a) the increase in tip radius is shown for emitter radii \( R_1 \) and \( R_2 \) after \( N \) apex planes have been removed from the emitter surface. From the geometry it is clear that:

\[
\Delta x = (R_2 - R_1) \sin \theta
\]  

(3.64)

The number of \((hkl)\) planes removal for a known number of apex planes \( N \) can be related via the inter-planar angle \( \theta \), which can be shown from Fig. 3.14(b) to be of the form:

\[
(N_{hkl} + \delta_{hkl}) = Z \cos (\theta + \psi)
\]  

(3.65)

where \( d_{hkl} \) is the interplanar spacing and \( \delta_{hkl} \) the phase of a particular \((hkl)\) plane which can take a value in the range \( 0 < \delta_{hkl} < 1 \). Equation 3.65 can be expanded to give:

\[
(N_{hkl} + \delta_{hkl})d_{hkl} = Z (\cos \theta \cos \psi - \sin \theta \sin \psi)
\]

It can further be shown from geometry in Fig. 3.14(b) that:

\[
\sin \psi = \frac{\Delta x}{z} \quad \text{and} \quad \cos \psi = \frac{h}{z}
\]  

(3.66)

where \( h = (N_{\text{apex}} + n_2 - n_1) d_{\text{apex}} \).
Fig. 3.14 A diagram illustrating the field evaporation model used to determine the relative planar removal rates.
Here the integers $n_2$ and $n_1$ correspond to the numbers of apex planar rings which extend to the (hkl) planar region prior to and after field evaporation, including the edge of the emitter. The edge can be considered as the intersection of the imaging emitter surface and the non-imaging truncated cone. Substitution of the above equation into equation (3.65) gives:

$$(N_{hkl} + \delta_{hkl}) \ d_{hkl} = (N_{\text{apex}} + n_2 - n_1) \ d_{\text{apex}} \ \cos \theta - \Delta x \ \sin \theta$$

Further substitution for the $\Delta x$ term using equation and the Dreschler and Wolf relation $R = nd/(1-\cos \theta)$ produces:

$$(N_{hkl} + \delta_{hkl}) \ d_{hkl} = (N_{\text{apex}} + n_2 - n_1) \ d_{\text{apex}} \ \cos \theta$$

$$- \frac{(n_2 - n_1) \sin^2 \theta \ \ d_{\text{apex}}}{(1- \cos \theta)} \quad (3.67)$$

and on rearranging gives:

$$(N_{hkl} + \delta_{hkl}) \ d_{hkl} = N_{\text{apex}} \ d_{\text{apex}} \ \cos \theta - (n_2 - n_1) \ d_{\text{apex}}$$

so that:

$$N_{hkl} = N_{\text{apex}} \ d_{\text{apex}} \ \cos \theta - (n_2 - n_1) \ d_{\text{apex}} - \delta_{hkl} \ d_{hkl} \quad (3.68)$$

This equation therefore represents the number of planes removed at a known interplanar angle $\theta$ as a function of the number of apex planes removed during field evaporation. The last two terms correspond to the number of apex planar rings to the edge of the emitter for the initial and final field evaporated surfaces and the phase of the hkl plane. This
equation is in agreement with the previous attempts in using field evaporation to determine the amount of material removed and shows the cosine dependence. Moreover if the ratio of (hkl) planes to apex planes is taken, the last term in equation 3.68 can be neglected, leaving:

\[
\frac{N_{hkl} d_{hkl}}{N_{apex} d_{apex}} = \cos \theta - \frac{(n_2 - n_1)}{N_{apex} d_{apex}}
\]

Thus a plot of the ratio \(N_{hkl} d_{hkl}/N_{apex} d_{apex}\) against \(\cos \theta\) will produce a linear relationship whose line will not intercept the origin. Furthermore, for a zero ratio value corresponding to the non-removal of planes on the emitter surface we have:

\[
\cos \theta_0 = \frac{(n_2 - n_1)}{N_{apex} d_{apex}}
\]

This means that at the intersection of the emitter surface and the truncated cone, no field evaporation occurs, ideally this intersection corresponds to the maximum angle \(\theta_m\). The use of equation 3.69 will be described more fully in the next section dealing with the experimental results.

The effects of non-spherical end forms on the amount of material removed during field evaporation has also been considered. For this an elliptical emitter shape was considered which can be described by a relation of the form:
\[ R_\theta = R_0 \left( 1 \pm \epsilon \sin \theta \right) \]  \hspace{1cm} (3.71)

where \( \epsilon \) is the eccentricity of the ellipse. The variation of the emitter surface with angle \( \theta \) is given by \( R_\theta \) and \( R_0 \) corresponds to the radius of an inscribed hemisphere.

In this model the elliptical cap is situated on a truncated cone of half angle \( \psi \) in a similar manner as the spherical emitter. The derivation is identical to the spherical emitter so that from 3.67, we have:

\[
\cos \theta \left( N_{\text{apex}} + (n_2-n_1) d_{\text{apex}} - (R_2-R_1) \sin^2 \theta \right) = (N_{hkl} + \delta_{hkl}) \cdot d_{hkl} \]  \hspace{1cm} (3.72)

similarly it can be shown that:

\[
R_\theta = \frac{nd_{\text{apex}}}{1-\cos \theta \left( 1 \pm \epsilon \sin \theta \right)} \]

so that:

\[
R_2 - R_1 = R_{\theta_2} - R_{\theta_1} = \frac{(n_2-n_1) \cdot d_{\text{apex}} \left( 1 \pm \epsilon \sin \theta \right)}{1-\cos \theta \left( 1 \pm \epsilon \sin \theta \right)} \]  \hspace{1cm} (3.73)

thus from 3.72 and 3.73 we have:

\[
N_{hkl} \cdot d_{hkl} = N_{\text{apex}} \cdot d_{\text{apex}} \cos \theta - (n_2-n_1) \cdot d_{\text{apex}} \frac{1-\cos \theta \pm \epsilon \sin \theta}{1-\cos \theta \pm \epsilon \sin \theta \cos \theta} \]

\[
- \delta_{hkl} \cdot d_{hkl} \]  \hspace{1cm} (3.74)
Taking the ratio of $(hk\ell)$ planes and apex planes we have:

\[
\frac{N_{hk\ell} \, d_{hk\ell}}{N_{apex} \, d_{apex}} = \cos \theta - \frac{(n_2 - n_1)(1 - \cos \theta \mp \epsilon \sin \theta)}{N_{apex} \, (1 - \cos \theta \pm \epsilon \sin \theta \cos \theta)} \tag{3.75}
\]

Therefore for an idealised elliptical end form the amount of material removed from the surface has a cosine dependence with the apex of the tip. When $\epsilon$ has a zero value a spherical end form is produced, and reduces to:

\[
\frac{N_{hk\ell} \, d_{hk\ell}}{N_{apex} \, d_{apex}} = \cos \theta - \frac{n_2 - n_1}{N_{apex}}
\]

which is identical to equation 3.69. Therefore the cosine relationship for the amount of material removed from the emitter surface is valid for tip shapes which may have large deviations from the spherical end form, although the extra term becomes more complex with increasing deviation.

Evaporation sequences of a number of planes from the apex of the emitter surface will enable the number of $(hk\ell)$ planes to be found if the end term is small enough to be neglected, which is the case when $N_{apex} \, d_{apex} \cos \theta \gg (n_2 - n_1)$, so that:

\[
N_{hk\ell} \, d_{hk\ell} = N_{apex} \, d_{apex} \cos \theta \tag{3.76}
\]

Equation (3.76) can therefore be used to determine the relative rates of evaporated planes over the emitter surface. This relation is
then useful in the determination of the depth of damage below the original surface. Furthermore, controlled field evaporation sequence allows the size of a void or similar feature to be determined from a knowledge of the angle $\theta$ and the number of apex planes removed during field evaporation through the void.

3.7.3. Field evaporation: experimental

The relative removal rates of field evaporated (hk\$) planes over the emitter surface can be determined by the use of a closed circuit television (CCT) system. In a series of experiments symmetrical tungsten emitters were imaged in the field-ion microscope and the apex (011) planes slowly evaporated under controlled conditions.

The experimental layout used in the experiments is illustrated in Fig. 3.15. A CCT camera (Sony, model HV 405) fitted with a f2-1 Cannon zoom lens of variable focal length (11.5 to 90mm) is positioned directly in front of the screen. The CCT camera is focussed onto the screen and the TV signal fed into a Sony video tape recorder (VTR) which enables the field evaporated sequence to be simultaneously observed and recorded on a high resolution TV monitor.

The evaporation sequences are controlled by the voltage applied to the field-ion emitter which in turn is controlled by a high precision ten turn potentiometer. The voltage is supplied by Brandenburg power supplies (Alpha 1 series) having a regulation of 0.05% and a maximum ripple of 0.05%. Field evaporation sequences of up to 200 apex planes of tungsten emitters were performed at a removal rate of about 0.5 apex
Fig. 3.15  Block diagram of the experimental layout used in the recording of the controlled field evaporation sequences.
plane sec

The evaporation sequences were recorded and subsequently analysed using the VTR.

This procedure thus allows the number of a given \((hkl)\) plane to be determined for a known number of removed apex planes. The evaporation sequence could be continuously replayed using the VTR and the removal rates for different \((hkl)\) planes determined. In all the experimental work the removal rate was kept constant at a slow evaporation rate, since the shape of the emitter may be influenced at higher removal rates (Hren et al. 1967). In a series of experiments two types of field evaporation analysis were performed, in the first analysis the number of prominent planes removed along a particular zone line \((\{111\})\) type) were determined. In the second and more general analysis, a large number of evaporated low and high index planes were recorded from several principal zone lines which extend over the emitter surface. In the course of the experiments nearly 4500 field evaporated planes were analysed in the determination of the evaporation rates of the different \((hkl)\) planes.

In the first experiment a total of 191 \((011)\) apex planes were removed from the tungsten emitter surface. The number of \((hkl)\) planes removed from the emitter surface for a particular zone line is shown in Table 3.2. Several other planes situated on different zone lines are also included. The total number of each \((hkl)\) plane is correlated with the number of apex planes removed by field evaporation and the appropriate interplanar spacings, shown in Table 3.3. The ratio \(N_{hkl} \frac{d_{hkl}}{N_{011} d_{011}}\) has been determined and plotted against \(\cos \Theta\) as shown in Fig. 3.16, where \(\Theta\) is the interplanar angle between the apex \((011)\) plane and a given \((hk\ell)\) plane.
### Table 3.2

Field evaporation data

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TABLE 3.3
Planar removal rates of different (hk\ell) planes.

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<tr>
<th>hk\ell</th>
<th>cos Θ</th>
<th>d_{hk\ell}</th>
<th>N_{hk\ell}</th>
<th>Ratio (N_{hk\ell} \cdot d_{hk\ell}) / (N_{011} \cdot d_{011})</th>
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<td>031*</td>
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<td>211*</td>
<td>0.578</td>
<td>1.292</td>
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<td>222*</td>
<td>0.817</td>
<td>0.9136</td>
<td>59*</td>
<td>0.803</td>
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* These planes are on different type zone lines
+ Only analysed for 30 (011) planes removed.
A plot of the ratio $\frac{N_{hkl} d_{hk\ell}}{N_{011} d_{011}}$ against $\cos \theta$ for a single [111] zone line of a tungsten emitter. A total of 191 (011) apex planes have been removed during controlled field evaporation.
A linear relationship is observed between the ratio and \( \cos \theta \) which indicates that a cosine dependence exists between the amount of material removed from different regions of the emitter surface and the apex. The linear relationship formed in the range \( \theta = 0 \) to \( 60^\circ \) is in agreement with equation 3.76 deduced from theory for an idealised end form. Moreover, the extrapolation of the straight line does not intersect the origin, again in agreement with equation 3.76. A least squares linear regression analysis of the data plotted in Fig. 3.16, gives a straight line of gradient 1.12 and an intercept value of -0.154. For an ideal spherical end form a gradient of unity would be expected, however deviations from the spherical end form over the emitter surface could result in different values of the gradient.

The intercept on the abscissae reveals information about the change in the number of apex plane rings at the edge of the emitter during the course of field evaporation. Similarly the intercept on the abscissae axis provides information about the region of the emitter surface where no field evaporation occurs. Care however is required in the extrapolation of the data plotted in Fig. 3.16, which give tentative results for the intercept values, which should only be considered as approximate values. The data indicates that field evaporation does not occur beyond \( \Theta_m \sim 80^\circ \) as determined from the intercept on the \( \cos \Theta \) axis.

A similar analysis of a field evaporated tungsten emitter has been performed for a series of different principal [111] orientated zone lines. A total of 70 apex planes were removed by a controlled field evaporation sequence, the number of prominent planes removed from the emitter surface were recorded as shown in Table 3.4. The ratio
### TABLE 3.4

Field evaporation data for several principal zone lines

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cont.
TABLE 3.4 (cont).

Field evaporation data for several principal zone lines

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TABLE 3.5

Planar removal rates of (hkl) planes from several principal zone lines

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(N_{hkl} \frac{d_{hkl}}{N_{apex} d_{apex}}) for each plane is determined in Table 3.5 and the data plotted against \( \cos \theta \) in Fig. 3.17. A linear relationship indicating a cosine dependence between the amount of material removed at the apex and a given (hkl) plane is observed for the four principal zone lines considered. There is however, a considerable spread in the data points, especially at higher values of \( \theta \).

The variation of the data points also indicates that each zone line has a slightly different gradient which suggests that the end form is not symmetrical. This is also indicated by the different intercept values on the abscissae axis giving an approximate range of angular \( \theta_m \) values of 70° to 88°. These angles correspond to surface regions where field evaporation does not occur on the edge of the emitter surface. However, a least squares linear regression has been fitted to the data shown in Fig. 3.17, where a straight line of gradient 1.155 is found and an extrapolated intercept on the abscissa indicates an average value of \( \theta_m = 82° \) is found for the limit of field evaporation on the emitter surface.

The most important fact to emerge from this series of experiments, however, is the cosine relationship which exists between the amount of material removed from the apex and any planar region on the emitter surface. This forms the basis of the relation:

\[
D = N d \cos \theta
\]

where \( D \) is the amount of material removed by field evaporation at a given angle \( \theta \) to the apex plane. The above relation can be derived theoretically as shown in equation 3.68, where it is assumed that the last two terms can
Fig. 3.17  A plot of the ratio \( \frac{N_{\text{hkl}}}{N_{\text{011}}} \) against \( \cos \theta \) for several [111] zone lines of a tungsten emitter. A total of 70(011) apex planes have been removed during controlled field evaporation.
The gradient of the data plotted in Fig. 3.17 also gives information regarding the typical spread in the results involved in these measurements between the different zone lines. These results indicate that the error involved in the use of the relation $D = N \cdot d \cdot \cos \theta$ is less than 20% for a typical field-ion emitter. The corresponding depth below the original surface may also be determined for different $(hk\ell)$ planes as a function of the number of apex planes removed. This is illustrated in Fig. 3.18, where the amount of material removed from each $(hk\ell)$ planar region (in terms of depth from Table 3.2) has been plotted against the number of apex planes removed by field evaporation. This corresponds to the depth scale since the amount of material removed from each planar region is known when compared with the original emitter surface prior to field evaporation. Therefore a relation of the form $D = N \cdot d \cdot \cos \theta$ can be used as a quantitative technique to determine the size and depth of any interesting feature observed in the field-ion image. This will be achieved in practice by measuring the persistence of the features as a function of the number of apex (monitor) planes removed during successive field evaporation.
Fig. 3.18 Depth of material removed from (hk\&i) planes as a function of the number of (011) apex planes removed by field evaporation.
3.8 Particle and void sizing techniques

The FIM has some unique advantages for the observation and analysis of small finely dispersed particles (<10nm) and low-energy radiation damage in metals such as voids or bubbles. Apart from the high resolving power of the technique to detect small microstructures on the emitter surface; specimens with high particle or void number densities are more readily examined in the FIM than the TEM. Furthermore the use of field evaporation enables an in-depth analysis not possible to achieve in the TEM. However, the determination of any particle size distribution by electron microscopy is relatively straightforward, performed by analysing plane sections (Goldsmith, 1967). In the FIM, however, the problem is more complex due to the curved shape of the emitter surface.

In principle there are two basic methods of obtaining a particle or void size distribution from field-ion micrographic data. The first method employs the projection relation deduced by Brandon (1964) and Newman et al (1967) viz:-

\[ D = k \theta \]

where the size of the void \( D \) can be determined directly from the micrograph by measurement of the void image width. The variation in magnification across the image is then determined by suitable measurement of the constant \( k \) along a zone line close to the void.

This method, however, suffers from several disadvantages, in
that the chosen value of \( k \) must be representative of the region of the emitter surface containing the void. Furthermore large numbers of voids make this method tedious and extremely difficult on heavily damaged emitters having highly distorted surfaces.

The second and more useful method of analysis is the application of controlled field evaporation sequences to establish the particle or void size distribution. A series of micrographs representing a sequence of field evaporated surface layers of known layer thickness can thus yield quantitative data on the particle or void size distribution. The void sizes, for example can be found from the number of layers over which the voids are seen to persist on the sequence of micrographs. A knowledge of the variation of thickness of each layer removed from different regions of the emitter surface enables the precise void size to be established. Thus the size of a void \( D \) persisting for \( n \) apex planes removed will be given by:

\[
D = nd \cos \theta
\]

The angle \( \theta \) can be determined not only from the \( D = k \theta \) relation but also by inspection of the void close to prominent planes on the micrograph, thus not susceptible to variations in magnification across the image. Furthermore, the number of apex planes removed at which the void is initially observed may also be recorded. This together with the persistence of the damage (at a measured angular orientation) permits the determination of the depth of void beneath the emitter surface. This method can therefore convert the depths at which the voids occur beneath the curved emitter surface to a linear depth scale.
Schwartz and Ralph (1969) have successfully employed this method to obtain particle size distributions from field-ion data in studies on the precipitation and growth of second phase particles in several alloy systems. The size and number density of precipitates in copper alloys has also been investigated with this method (Goodman et al (1973)). In this study the particle size measurements were restricted to particles located within 30 degrees of the apex plane. The cos θ term in the size relation was neglected resulting in an overestimate of particle size by 13%. However close agreement was found for particle sizes in both FIM and TEM techniques for the larger particle sizes studies (> 10nm). The number density of the particles was determined by counting the number of particles observed within a volume of material, V, removed by field evaporation. The volume of material removed by evaporation was calculated by approximation to a solid frustrum of a cone given by:

\[ V = \frac{\pi d}{3} (A_1 + A_2 + (A_1 A_2)^{\frac{1}{2}}) \]  

(3.77)

where n is the number of field evaporated apex planes of interplanar spacing d. \(A_1\) and \(A_2\) correspond to the initial and final area of the imaged surface under examination.

Brenner and Seidman (1975) have similarly used this method to determine the sizes of voids produced by neutron irradiation of molybdenum specimens. The void sizes were measured from the number of apex planes removed between the appearance and disappearance of a void for a calculated value of θ. The void number density was determined by the use of equation 3.77 and \(A_1\) and \(A_2\) found from the average radius
of curvature. They were also able to show that the void number density \( N_d \) implied an average spacing between the voids given by

\[
2R = \left( \frac{4}{3} \pi N_d \right)^{-\frac{1}{3}}
\] 

(3.78)

thus allowing a direct comparison with similar electron microscopy studies.

This technique can therefore not only determine void or particle size distributions by the choice of a suitable counting pole with sufficient small field evaporation increments, but can also be used to perform depth profile analysis beneath the emitter surface. Thus this method, together with a knowledge of the variation of the amount of material removed over the emitter surface, represents a powerful technique for the analysis of void or damage distributions within the emitter in low-energy ion bombardment studies using the FIM technique.

3.9 Summary

In this chapter the crystallography and emitter topography of field-ion specimens is reviewed, and the basic relations concerning the crystal structure are reported. The radius and surface topography of specimens may be deduced from ring counting measurements obtained directly from field-ion micrographs. The projection geometry of the FIM has been extensively reviewed in terms of the current models describing the projection relations. The introduction of the moiré analogue enables further information to be discerned, concerning the shape of projected
ring systems from an idealised emitter surface of known surface topography.

The application of the moiré analogy has been reported in detail, especially concerning the patterns produced by non-spherical zone plates, which represent non-spherical planar facets evident in field-ion micrographs. The theoretical investigation indicates that the location and orientation of the resultant moiré patterns are not simply related to the generating zone plates. Two specific examples illustrate the main features of the investigation, in which the shape, location, size and orientation of the patterns are described. The application of the moiré patterns for non-spherical planar facets may be particularly useful in irradiation damage studies, especially in the analysis of severely damaged specimens.

The field evaporation phenomena is considered in detail, particularly as an analytical tool, and several theoretical models reviewed from previous studies. An idealised model for field evaporation of a hemispherical emitter is reported. A relationship has been derived for the relative removed rates of different planar regions of the emitter undergoing field evaporation. The model is correlated with extensive experimental measurements during controlled evaporation sequences. Good agreement is observed between theoretical prediction and experimental results.

The field evaporation model forms the basis of a particle or void sizing technique using controlled evaporation rates and is compared with previous analysis methods employed with the FIM technique.
The model not only provides information concerning the size of damage or defects but also the depth at which such features are present beneath the original surface of the emitter. The controlled field evaporation analysis method therefore provides a powerful technique for the study of irradiation damage using the FIM.
CHAPTER FOUR

Low-Energy Radiation Damage

4.1 Introduction

The irradiation of metal surfaces with energetic particles causes a variety of surface and sub-surface phenomena such as sputtering, channeling, trapping and re-emission of trapped particles, and radiation damage. Thus energetic ions which penetrate the surface into the metal lattice may displace lattice atoms from their normal sites to create vacancies and interstitials. When the incident ions have slowed down sufficiently they may be trapped in either interstitial or substitutional lattice sites. Under certain conditions (see for example McCracken (1975)) hydrogen and helium ions can combine with the vacancies created by lattice displacements and nucleate gas bubbles in the bulk material. If the gas bubbles are located close to the surface with high gas pressure, then the bubbles may plastically deform the surface layers by extreme deformation to produce rupturing. Therefore the phenomenon of surface deformation associated with gas bubbles produced by irradiation is known as "radiation blistering".

In this chapter the processes involved in low-energy irradiation damage are considered, initially at an atomic collision level where single displacement events occur such as Frenkel pair production. The Kinchin-Pease model concerning the number of displaced atoms is briefly reviewed and the effects of collision sequences and channeling are discussed. The role of the implanted atoms in trapping sites is discussed from previous studies using techniques such as thermal desorption, which reveal infor-
mation concerning the different forms of trapping sites together with the atomistic behaviour of the implanted gas atoms. The damage and range distribution of low-energy implanted helium is determined using numerical calculations based on LSS theory, which is briefly described. The total and mean projected range of low-energy hydrogen and helium in tungsten and molybdenum have also been determined. The conditions necessary for radiation blistering are considered and previous studies outlining the main experimental parameters and observations are reviewed. The mechanisms proposed for bubble formation and growth are discussed, together with a review of the three main types of theories dealing with radiation blistering. The work described in this chapter attempts to give an overall view of all the relevant factors concerning the radiation blistering phenomena, which will aid the study of low-energy irradiation damage.

4.2 Production of defects.

Energetic ions incident on the surface and travelling through the irradiated target undergo an energy loss by collision processes with the target atoms. Eventually the implanted atoms are brought to a minimal energy value within the target. In elastic collisions, energy is conserved between the incident projectile and the target atoms, although inelastic collision events can convert the transmitted energy into different forms (i.e. atom excitation). When an energetic projectile initially makes contact with the target atom, a fraction of its energy is dissipated in a primary collision event, which can produce a secondary cascade of atomic collisions resulting in the displacement of the surrounding atoms.
The maximum energy transferred $E_T$ between the projectile and target atom can be determined from classical theory from the simple relation:

$$E_T = \frac{4M_1M_2}{(M_1+M_2)^2}E$$  \hspace{1cm} (4.1)

where $M_1$ and $M_2$ are the masses of the projectile and target atoms and $E$ is the incident ion energy. The energy transferred to the target atom may be sufficient to produce further displacements. The distribution of energies imparted during irradiation depends in the collision cross section of the type of interaction between the particles. At certain ion energies, however, the primary event can occur close to or at the surface and may cause the ejection of some surface atoms. The incident ion collision processes resulting in the ejection of surface atoms is known as sputtering. Several extensive reviews of this subject have been given by Carter and Colligon (1968) and Sigmund (1975).

One of the most simple damage mechanisms between the energetic projectile and the metal lattice is the displacement of an atom situated at a normal lattice site into an interstitial position, the so called Frenkel defect. The amount of energy required to form a Frenkel pair is usually termed the displacement energy $E_d$ and has a bulk value in the range 25 to 40 eV for bcc metals. The average number of Frenkel pairs $N(E)$ produced in a solid by an energetic projectile can be estimated from a simple model originally developed by Kinchin and Pease (1955) as:

$$N(E) = \frac{E}{2E_d}$$  \hspace{1cm} (4.2)
where $E$ is the projectile energy and $E_d$ is the effective displacement threshold energy. The recoiling primary atom usually possesses sufficient energy to produce a cascade of secondary displacements at high projectile energies, although this effect may diminish at lower ion energies ($<10$ keV). Dworschak et al (1967) have pointed out that a large recombination volume may exist (~100 atomic volumes) about an isolated point defect, so that 'self healing' by recombination of vacancies and interstitials can occur. Furthermore if the energy of the projectile falls below a certain value (i.e. $< 2E_d$), then the primary atom may simply replace the displaced atom producing no net-displacement. Thus the average number of Frenkel pairs produced per projectile $N(E)$ increases from zero at $E = E_d$ to unity at $E = 2E_d$.

The Kinchin-Pease model is based on the random slowing down of the projectile by elastic collisions and hard-sphere scattering with energy constrictions about a defined transition energy $E_d$ has been studied by Sigmund (1969). Thus for low-energy projectiles, using a power approximation in the Thomas-Fermi collision cross-section, the average number of displaced atoms $N(E)$ has an upper limit of:

$$N(E) = \frac{6}{\pi^2} \frac{E}{U} \ln \left( 1 + \frac{V}{E_d} \right)$$

(4.3)

where $U$ is the bonding energy of the metal lattice atom. The loss of defects by replacement collision is not considered and for a small bonding energy equation (4.3) reduces to:

$$N(E) = 0.61 \frac{E}{E_d}$$

(4.4)
indicating a greater number of displaced atoms than the simple Kinchin-Pease model. Furthermore, the threshold displacement energy can have a range of values due to the crystalline nature of the irradiated target, where a directional dependence would be expected. Although the Kinchin-Pease model takes no account of the crystalline nature of the irradiated target, the lattice structure can produce significant effects on the range and distribution of damage.

One effect of lattice structure is that of focused collision sequences, which can result in the long range transfer of energy and matter, and thus influence the distribution and total number of defects produced (Silsbee (1957)). This can occur in collisions where the knock-on atom receives an energy below a certain focusing limit and within a certain solid angle, and thus transfers energy along a close packed line of atoms. Energy is dissipated by interactions with neighbouring lines of atoms and if a row intersects the surface, an atom may be ejected. In this case no replacement occurs and the energy is transmitted along the line as a focused energy packet. The focused collision sequence is not only able to transfer energy, but also atoms, thus in replacement collision sequences an interstitial atom can be produced some distance from the primary collision (Thompson (1969)). The range of focusing sequences are determined by the focusing energy $E_f^{hk\ell}$ and the replacement energy $E_r^{hk\ell}$. Simple focusing can occur only along close packed directions for transmitted atom energies in the range $E_r^{hk\ell} < E < E_f^{hk\ell}$. Assisted focusing can also occur in certain crystal directions ([110] directions in bcc metals) where the range of the focusing sequences is dependent on the temperature (lattice vibrations) and on the distribution of defects. A schematic diagram illustrating some of these energy transfer collision processes is shown in Fig 4.1 after Venables.
A schematic diagram illustrating some of the collision processes which can occur during low-energy ion irradiation (Venables (1970)).
A second phenomenon which can significantly alter the range and distribution of damage is that of channeling. In this phenomenon the projectile atoms can travel along the open channels which exist between the close packed rows of atoms, Gemmell (1974) has written an extensive review of channeling. For monocrystalline target materials, channeling is strongly dependent on the relative orientations of the beam and target. In bcc metals channeling occurs most favourably along the [110] direction and can also occur in any low index planar direction. The average number of displaced atoms can be reduced by channeling, although long range effects can increase the damage volume.

The damage processes occurring in irradiated materials described above have only considered the damage formation as collision and energy transfer processes. However in light ion irradiation the role of the implanted atoms can have a significant effect in the production of defects by the formation of complex defects (Baskes and Wilson (1976)). This can occur when the implanted gas atom penetrates beyond the last vacancy it has produced, due to insufficient energy for single vacancy displacement. Thus the implanted atoms at the end of their range will reside as interstitials unless the interstitial mobility is sufficient for the implanted atoms to move substitutionally into the vacancies it has created. The presence of implanted gas atoms within a vacancy is sufficient to prevent Frenkel pair recombination (Baskes and Wilson (1976)) and so prevent 'self-healing' within the metal lattice. The resultant self interstitials of the target can therefore combine with any unfilled vacancies or become trapped in interstitial sinks such as dislocations. Thus the dominant defect created by
light ion implantation may be the trapped implanted atoms existing in substitutional sites within the metal lattice.

Considerable experimental and theoretical effort has been directed towards the study of the migration and trapping of inert gases; especially helium in metals. The thermal desorption technique has been widely used to establish the nature of the trapping sites and the disposition of the implanted gas atoms (Kornelsen (1970, 1972), Erents and McCracken (1972), Bauer et al (1972) and Cavaleru et al (1972). This technique utilises low-energy helium implantation (<250eV) at low doses (<10^{15} ions cm^{-2}) as a probe after previous irradiation with the higher energy implanted ions. The population of the individual trapping sites can be determined from the deconvolution of the desorption spectra. Kornelsen (1972) found that the population of individual sites varied in some cases directly as the square and in two cases as the cube of the incident ion dose. This evidence has been used to suggest multiple occupation of sites, furthermore Kornelsen has postulated that the lattice vacancy constitutes the main trapping site. Higher desorption temperature peaks have been assigned to several forms of multiple helium-vacancy complexes, where up to six helium atoms can exist in a single vacancy complex (Baskes and Wilson (1976)). Kornelsen has proposed that at low temperature, the strain in the lattice increases as the vacancy becomes occupied with more gas atoms, until at some point the arrival of another implanted gas atom displaces a second lattice atom, resulting in a multiply occupied divacancy or embryo bubble. This point will be discussed again when dealing with bubble growth.

The binding energies of implanted helium atoms trapped at vacancies in metals (eg. tungsten) have also been studies by models using computer
simulation techniques. (Wilson and Bisson (1973, 1974 and 1975). Close agreement has been found between the proposed binding energies for the various vacancy complex configurations (McCracken (1975)), and in general suggests a strong confirmation of Kornelsen's model. An extensive review of the atomistics of helium implanted in metals has also been given by Reed (1977). Further evidence for helium-vacancy complexes have been obtained by channeling experiments on the lattice location of implanted helium. Picraux and Vook (1973) found that the implanted helium atoms did not reside in interstitial sites, but were present in a helium-vacancy complex where up to three helium atoms per vacancy were observed.

Some experimental work has also been carried out on hydrogen implantation in molybdenum using the thermal desorption technique (McCracken and Erents (1973)). The behaviour of hydrogen trapping is similar to that of helium, although hydrogen exhibits diffusion coefficient for interstitial diffusion, but atoms are trapped at the damage sites. The theory of diffusion and trapping of hydrogen and helium in metals has been reviewed by Wilson (1976). The trapping of energetic hydrogen (18 keV) ions in a range of reactive metals has been studied by Hoiston and McCracken (1977), they found that the trapping behaviour was primarily dependent on the diffusion coefficient and the heat of solution of hydrogen in the metal. Close agreement was found for both ion dose and target temperature with trapping behaviour when compared with theoretical models, such as the bubble coalescence model.

4.3 **Range and damage distributions**

In the last decade the implantation of low energy (<10 keV) light
ions in solids has become increasingly important for the analysis of problems produced by radiation damage. This is particularly the case in the destruction of solar cells in outer space and for first wall problems in fusion reactions, where a knowledge of the range and damage profiles is crucial. One of the most important theories dealing with low-energy ion implantation was originally developed by Lindhard, Scharff and Schiott in 1963, which has since become known as the LSS theory. Several other theories have been developed to calculate the spatial distribution of the implanted ions. These include a transport theory developed by Brice (1970) and a moments method favoured by Winterbon (1972). Range and damage distributions have also been obtained directly by Monte Carlo calculations (Oen et al (1976)). Cascade simulations using a computational technique known as the MARLOWE program have similarly been employed (Littmark et al (1976). However the most comprehensive and widely used theory has been the LSS theory for the determination of the ranges of implanted ions in solids.

The LSS theory only applies to an amorphous target structure where the energetic projectile is considered to lose energy by two processes, namely an electronic collision and a nuclear collision. For projectile energies above 10keV electronic processes dominate, whereas for low energy ions the nuclear (elastic) collisions are important and large scattering angles occur. In the LSS theory the two energy loss processes are combined to determine the rate of energy loss with distance into the target. The theory is based on a universal nuclear stopping cross-section $S_n(E)$ calculated from a Thomas-Fermi model of the interaction between the projectile and the target atom. The electronic stopping cross-section $S_e(E)$ is assumed to be proportional to the velocity of the pro-
jectile. The energy loss per unit path length of the moving projectile is therefore given by:

\[
\frac{dE}{dx} = \left( \frac{dE}{dx} \right)_e + \left( \frac{dE}{dx} \right)_r = -N \left( S_e(E) + S_n(E) \right)
\]  

(4.5)

where \( N \) is the atomic density of the target. Thus it follows that the total path length \( R \) of the projectile slowing down from an initial energy \( E_i \) will be given by:

\[
R = \frac{1}{N} \int_0^{E_i} \frac{dE}{S_n(E) + S_e(E)}
\]  

(4.6)

In the LSS notation the energies and distances are expressed as dimensionless parameters \( \epsilon \) and \( \rho \) which are considered in approximation to be independent of each other. The values of \( \epsilon \) and \( \rho \) for a given projectile/target combination can be calculated from the following relations (Lindhard et al (1963)):

\[
\epsilon = \frac{aM_2}{z_1^2z_2^2e^2(m_1 + m_2)} E
\]  

(4.7)

and

\[
\rho = RN\pi a^2 \frac{m_1m_2}{(m_1 + m_2)^2}
\]  

(4.8)

where the symbols have the usual meaning of mass number \( m \), atomic number \( z \), electronic charge \( e \) and the Thomas-Fermi screening length \( a \), usually taken
as:

\[ a = 0.8853 \times a_0 Z^{-\frac{1}{3}} \]  \hspace{1cm} (4.9)

where \( a_0 \) refers to the Bohr radius (0.052915 nm). Using the reduced energy and distance parameters the path length of the incident projectiles then becomes from equation (4.6):

\[ \rho = \int^\infty_0 \frac{\mathrm{d} \varepsilon}{S_n(\varepsilon) + S_e(\varepsilon)} \]  \hspace{1cm} (4.10)

The electronic stopping power \( S_e(\varepsilon) \) is taken to be proportional to the projectile velocity such that (Lindhart and Schorff (1961)):

\[ S_e(\varepsilon) = k \varepsilon^\frac{1}{4} \]  \hspace{1cm} (4.11)

the constant term \( k \) depends in a complex form on the projectile-target combination, but can be calculated using the relation (Lindhart et al (1963)):

\[ k = \frac{0.0793 \frac{1}{2} \frac{1}{3} \frac{1}{6} \frac{1}{2}}{(Z_1^2 + Z_2^2)^{\frac{3}{4}} (M_1 + M_2)^{\frac{3}{2}}} \]  \hspace{1cm} (4.12)

Therefore a universal curve cannot be obtained for the electronic stopping since \( k \) is dependent on the types of colliding atoms. However, the nuclear stopping power \( S_n(\varepsilon) \) can be calculated from the differential cross-section for energy transfer. A universal nuclear stopping power derived from the Thomas-Fermi statistical model of the projectile-atom collision by numerical
solution have been tabulated by Schiott (1966) and Lindhard et al (1968). The Thomas-Fermi statistical model was chosen to simulate the interatomic potential because of a more reasonable approximation compared with the classical approach (i.e. inverse square potential). An analytical function which gives a good fit to the tabulated data for the nuclear stopping has been developed by Winterbon (1968):

\[ S_n(\varepsilon) = \frac{9}{8\varepsilon} (\log(1 - r) - \frac{r}{r}) \]  

(4.13)

where \( x = (2.618\varepsilon^3)^{1/3} \) and \( r = (1 + x^2)^{1/2} \). The combination of these expressions for \( S_n(\varepsilon) \) and \( S_e(\varepsilon) \) together with equation (4.10) allows the total path length (i.e. total range) for any projectile-target combination to be determined. An approximate value of the mean projected range can also be obtained from the relation (Schiott (1968)):

\[ \frac{R}{R_p} = 1 + \frac{M_2}{3M_1} \]  

(4.14)

where \( R \) is the total range of the energetic projectile. The approximate values of \( R \) and \( R_p \) have been calculated using a computer program, however the results presented here are those from the Manning-Mueller program which is described in the rest of this section.

The mean projected range and total range of low energy (1 to 9 keV) hydrogen and helium ions are shown in Figs. 4.2 and 4.3 for tungsten and molybdenum targets respectively. The standard deviation in the mean projected range is also included. The plots indicate that for hydrogen irradiation the mean projected range is larger in molybdenum than in tungsten, which have similar total ranges. However, the standard deviation
in mean projected range is greater tungsten than molybdenum. For helium irradiation the mean projected range is again larger in molybdenum than tungsten with similar total ranges. The standard deviation is also greater in tungsten than molybdenum and is increased at higher projectile energy (>5keV). These results however, only represent approximate values due to the assumptions in the LSS theory.

A powerful computational method has been developed by Manning and Mueller (1974), to calculate the depth distribution of damage and ion range distribution. The computer program is based on the LSS theory for amorphous materials and modified to include a Gaussian projected range distribution. A review of the program has been given by Mathews (1974), who has adapted the program to run on the Harwell computer facility. In this method the projected range distribution \( f(R_p) \) is assumed to have a Gaussian form described by the relation:

\[
f(R_p) = \frac{1}{\sqrt{2\pi}\sigma_R} \exp \left( -\frac{(R_p - \bar{R}_p)^2}{2\sigma_R^2} \right)
\]

where \( f(R_p) \) is the fraction of beam ions with projected range in the region \( dR_p \) of \( R_p \). The projected range \( \bar{R}_p \) and the root mean square deviation in the projected range \( \sigma_R \) are obtained from a modified Johnson and Gibbons (1968) program, which calculates these values according to the LSS theory.

The program also evaluates the density of displaced atoms as a function of depth \( \rho(x) \) assuming that the primary knock on atom ranges are small thus not creating further displaced atoms, then from Torrens and Robinson (1972):
Fig. 4.2 The mean projected range (and standard deviation) and total range of low-energy (1 to 9 keV) hydrogen and helium ions in tungsten, determined from LSS theory.
Fig. 4.3 The mean projected range (and standard deviation) and total range of low-energy (1 to 9 keV) hydrogen and helium ions in molybdenum, determined from LSS theory.
\[ \int_{0}^{\infty} \rho(x) \, dx = \frac{k}{E_d} \int_{0}^{\infty} S_B(x) \, dx \] 

(4.16)

where \( E_d \) is the threshold energy and \( k \) a constant (\( \approx 0.8 \)).

Using this assumption the deposited energy at \( x \), summed over all the ions can be found (Kulcinski et al (1971)). However, the use of this assumption for the deposited energy with depth leads to inaccurate energy deposition values at the surface of the target, due to a non-zero energy struggle at this point (Mathews (1974)). A further assumption has been utilised due to Johnson et al (1973), where the energy deposition profiles produced by all the ions have the same functional dependence as the profile for the ions of average range. Thus all the ions are assumed to deposit the same energy when slowing down to a minimum energy value.

The results of the Manning-Mueller computer program are illustrated in Figs. 4.4 (a-c), for 1 to 3 keV helium ions implanted in tungsten. The computer code has a displacement efficiency (k value) of 0.80 and a threshold displacement energy of 25eV. The damage profile is shown as displacements per atom (DPA) and the helium range distribution (dotted line) is depicted as an implanted concentration for a dose of \( 1 \times 10^{16} \) ions \( \text{cm}^{-2} \). The figures indicate that the maximum damage occurs at the surface, and decreases rapidly with depth from the surface. The depth of damage as shown by the damage profile increases with higher projectile energy. The peak implanted atom concentration decreases a higher projectile energy, as the peak moves deeper into the target, thus producing a broader distribution. The extent of surface damage also appears to decrease with increasing projectile energy. However, the computer program can produce inaccuracies at low projectile energies, although it also indicates the general trends of
Fig. 4.4 (a to c) The implanted helium (dotted line) and damage (full line) distributions corresponding to low-energy ion irradiation to a dose of $1 \times 10^{16}$ ions.cm$^{-2}$. The distributions have been determined using the Manning-Muehleifer computer program, with a displacement efficiency of 0.8 for helium ion energies of: (a) 1keV (b) 2 keV (c) 3 keV.
Fig. 4.4 (b) 2 keV.
Fig. 4.4 (c) 3keV.
the expected damage and range distributions under irradiation conditions. Thus computer simulation is a useful tool in irradiation studies as an aid in data interpretation of damage in the irradiated target and also the implanted atom distribution.

4.4 Conditions for blistering.

The ion irradiation of metal surfaces can result in the appearance of blisters, due to the formation of implanted gas atoms contained in bubbles within the solid. The growth of the bubbles eventually leads to the formation of surface blisters. For this to occur several conditions have been proposed. One obvious condition required for blister formation is the depth \( R \) at which the bubbles are formed, which must be larger than the thickness sputter-removed from the surface when the critical dose \( n_c \) required for blister formation has been achieved (Roth (1975)). Thus for a sputtering yield \( S \) and atomic density of the material \( N \), blistering would be expected when:

\[
S < \frac{R \cdot N}{n_c} \tag{4.17}
\]

where \( R \) corresponds to the mean projected range of the projectiles at which the bubbles are assumed to form.

The critical concentration of gas atoms to form large agglomerates can be represented by the helium concentration where the implanted atoms are not considered isolated. The implanted gas concentration can be determined from percolation theory (Wilson et al (1974)), which for bcc metals is found to be about 25 atomic per cent. Roth (1975) has shown
that for suitable values of mean projected range and sputtering yield, this condition can be applied to the helium and argon irradiation of niobium. This is shown in Fig. 4.5, where for normal ion incidence, the sputtering yield for light ions (e.g. helium) within the energy range considered is always less than \( R'N/n_c \). For the heavier argon ions, blistering occurs when the sputtering yield has decreased sufficiently at higher energies. This condition has been found in agreement with experiment (Hartley (1975)).

McCracken (1975) has also proposed that the time for formation of extensive defects must be less than the time required for erosion of the surface at which the defects are produced. For surface structure produced by irradiation to be formed the following criterion has been used (Hermanne (1973)):

\[
\frac{d_d}{V_s} < \frac{d_e}{4V_m} \tag{4.18}
\]

where \( d_d \) is the depth of damage below the surface and \( V_s \) is rate of erosion of the surface by sputtering. The distance between neighbouring defects is given by \( d_k \) and \( V_m \) is the migration velocity of the defects, where it is assumed that the average distance travelled by a defect is \( \sim d_k/4 \). Therefore the criterion for surface structure to appear due to irradiation depends on whether the defect migration velocity is less than a minimum value given by:

\[
V_{\text{min}} = \frac{4}{r_o S} \frac{J E_d}{E_o^{\frac{3}{2}} R_p} \tag{4.19}
\]

where \( J \) is the ion current density, \( E_o \) the ion energy, \( E_d \) the displacement
Fig. 4.5 A plot of sputtering yield against ion energy for helium and argon ion-bombardment of niobium. The shaded regions correspond to the implanted gas concentration, determined from percolation theory (Roth (1975)).
energy, $r_o$ the radius of the defect and the constant $K$. A comparison 
has been made between the criterion (equation 4.18 ) and experimental 
work, where surface structure was found to agree closely with prediction 
(Hermanne (1973)).

A further condition for blistering to occur has also been proposed (Roth 
1973), where the low solubility and diffusivity of the implanted ions 
within the solid will lead to blistering. Since implanted atoms with 
high diffusivities will (dependent on target temperature) rapidly diffuse 
to sinks within the solid; thus leading to a reduction in bubble and blister 
formation. This condition is fulfilled for the inert gases, but the diffu-
sion and solubility of hydrogen in bcc metals is considerably larger. For 
this reason blistering due to hydrogen irradiation occurs at very high 
critical doses $> 5 \times 10^{18} \text{ ions cm}^{-2}$ (Kaminsky and Das (1973)), and is also 
strongly dependent on the experimental ion implantation rate (flux) used 
(Verbeck et al (1973)). Low ion fluxes allow the implanted hydrogen to 
rapidly diffuse away from the implanted region, thus avoiding a high implant 
concentration necessary for blistering. In helium irradiation, the implanted 
helium has a low solubility in bcc metals, where the atoms are trapped at 
damage sites. This leads to a small effective diffusion coefficient (Blow 
(1972)), so that generally helium implantation fulfills the conditions 
necessary for blistering.

4.5 Radiation blistering.

The effect of radiation blistering has become increasingly important 
in such applications as fusion reactor technology and accelerator technology. 
Following the early work of Kaminsky (1963) and Primak (1966) in the effect
of space radiation on materials, a large number of experimental studies have been performed on radiation damage and blistering. Several extensive reviews have been written on radiation blistering by Das and Kaminsky (1975), McCracken (1974, 1979) and Roth et al (1975). Most of the blistering studies on metals and alloys have been conducted with light projectiles (e.g. H, D and He) at high energies (> 15 keV) using a range of techniques. The use of high ion energies has enabled the bubbles and blisters to be observed directly using optical and electron microscope techniques (e.g. SEM, TEM). However, by comparison, little work has been done using high resolution techniques at low-energies (e.g. FIM) and this is reviewed separately in the next chapter.

One of the most important parameters in radiation blistering is the projectile energy, which determines the depth at which the ions are implanted in the solid. Das and Kaminsky (1973, 1974) have shown that the depth profile of the implanted atoms and the energy deposited into damage are directly related to the projectile energy and can significantly affect the gas bubble and blister formation. Figure 4.6 shows the results from several studies, where the average blister size has been recorded as a function of the helium ion energy in several materials at room temperature (Das and Kaminsky (1973)). The blisters observed on the irradiated surface appear to have a Gaussian size distribution (Das and Kaminsky (1973)), where the mean blister size does not change with ion dose. The linear relationship observed in Fig. 4.6 has been qualitatively explained by Das and Kaminsky (1973, 1974) and Erents and McCracken (1975), where the gas pressure inside the blister is related to the size, height and skin thickness of the blister together with the yield strength of the material. Erents and McCracken (1973) have shown that the mean projectile range increases almost linearly with energy.
The average blister size is shown as a function of the helium ion energy after the irradiation of several materials at room temperature (Das and Kaminsky (1973) and Eents and McCracken (1975)).
Kaminsky et al (1976) have measured the blister skin thickness with increasing energy in the range 100 to 1000 keV for helium irradiation on vanadium and found a linear relationship.

Similar studies have been performed on niobium for a larger energy range (1 to 1500 keV) by several authors Roth et al (1973), Das and Kaminsky (1973, 1974), Behrisch et al (1975) and Str-Jacques (1976). In these studies the blister skin thickness has been measured and compared with the corresponding calculated projected ranges of the helium ions. Terreault et al (1978) have also studied the blister skin thickness for helium irradiation of copper at a lower energy range (1 to 25 keV). Generally a close agreement is found between the blister skin thickness and the calculated projected range values, however at the lower ion energies (< 15 keV) larger blister skin thickness measurements have been found, the difference increasing with decreasing ion energy (Roth (1973)). The blister skin thickness measured by Roth for low-energy irradiation was indirectly obtained from a Rutherford backscattering technique, whereas the other measurements were directly obtained using electron microscopy (Kaminsky et al (1973, 1974)). These results have lead to the formulation of a separate model, for low-energy irradiation (lateral stress model), where the blister formation results from differential expansion of the implanted surface layer. The blister skin thickness has been measured by Müller et al (1976) for deuterium ions implanted in nickel (200 to 400 keV), again close agreement was obtained with damage energy distribution calculated from Monte Carlo computational techniques. A similar result has been described in aluminium (Das and Kaminsky (1976)). Fenske et al (1978) and Das et al (1978) have irradiated nickel and beryllium targets with helium ions and found an empirical relationship between
the probable blister diameter and the mean blister skin thickness, although the relationship does not agree with the lateral stress model.

Helium irradiation studies have been performed on a range of materials, and helium bubbles observed in stainless steel (Walker (1970)), silver (Brebec (1964)), gold (Brown et al (1974)) and copper (Barnes and Mazey (1960)) after helium irradiation at doses from $10^{14}$ ions cm$^{-2}$ to $6 \times 10^{18}$ ions cm$^{-2}$. The helium concentrations range from about 10 ppm to several atomic percent. Evans et al (1976) were unable to distinguish any gas bubbles in helium irradiated molybdenum (36 keV) with a dose of $10^{16}$ ions cm$^{-2}$ at room temperature. However, bubbles were observed when a dose of $3 \times 10^{17}$ ions cm$^{-2}$ was attained. The bubbles were initially formed at random nucleating sites, but then developed into a bcc superlattice arrangement with a considerable degree of ordering before the critical dose for blistering was achieved. Systematic studies of the dependence of the critical dose required for blistering are limited, however, Evans (1978) has shown a general trend, depicted in Fig. 4.7. The critical dose for blistering in both molybdenum and niobium has been plotted against helium ion energy for both experimental and calculated values. A similar trend of increasing critical dose with increasing energy has been observed in nickel irradiated at low temperatures ($-150^\circ$C) with high energy (200 to 400 keV) denteron ions (Möller et al (1976)).

For doses larger than the critical blistering dose an increase in blister density occurs only when numerous small blisters cover the irradiated surface. When large blisters cover most of the irradiated area, an increase in blister skin exfoliation occurs with increasing total dose.
The critical dose required for blistering is plotted against helium ion energy for the irradiation of molybdenum and niobium targets. The shaded region corresponds to the calculated curves for different pre-threshold bubble radii, based on implanted gas concentration and bubble pressure (Evans (1976)).
This has been observed by Das and Kaminsky (1974) in high temperature (450°C), high energy (500 keV) helium irradiation of stainless steel. The blister density has also been observed to increase with increasing dose (Thomas and Bauer (1973)) in high energy irradiation of palladium. Erents and McCracken (1973) have similarly observed an increase in blister density in molybdenum when the total dose increased by a factor of two. The exfoliation of blister skins has not been observed in low-energy (< 15 keV) irradiation, Martel (1974) found blisters appearing in niobium targets irradiated with helium in the range (5 to 15 keV) for doses of $6 \times 10^{12}$ to $9 \times 10^{18}$ ions.cm$^{-2}$. Further increases in ion dose revealed that blistering did not reappear. This has been interpreted as a 'cut-off' dose, above which no blisters are observed. The 'cut-off' dose has been attributed to the dose necessary to sputter remove the material equivalent to the blister skin thickness.

This behaviour has been confirmed by Roth et al (1973) for 9 keV helium irradiation of niobium surfaces at high doses up to $7 \times 10^{20}$ ions cm$^{-2}$. Similar results have been obtained for stainless steel (type 304) irradiated with 5 keV hydrogen ions (Behrisch et al (1975)). The cut-off dose for blister disappearance has been observed to decrease with increasing target temperature for helium irradiation of niobium (St.-Jacques (1973)). Evans (1976) has suggested that blister formation at high doses of low energy (< 15 keV) helium ion irradiation may be prevented due to the increased surface roughness, which may prevent the coalescence of small bubbles. Furthermore, during the course of high dose irradiation the original blisters formed may be sputter removed leaving a rough surface. Subsequent low-energy irradiation would not produce blistering. Terrault et al (1978) have also examined copper surfaces irradiated with low helium
ions (1 to 25 keV) and found that the rougher surfaces produced by sputtering, with doses up to $2 \times 10^{18}$ ions.cm$^{-2}$, result in a reduction of blistering.

Target temperature has been considered as one of the important parameters affecting the radiation blistering process, where the average blister diameter, blister density and exfoliation of the blister skin are all dependent on temperature (Das and Kaminsky (1975)). The temperature effects on blistering has been studied by several authors, including Bauer and Thomas (1973, 1974) and Kaminsky and Das (1974) with high energy helium ions (300 to 500 keV). Similar behaviour has been observed in molybdenum irradiated with lower energy (36 keV) helium ions (Erents and McCracken (1973)). Severe exfoliation was observed at intermediate temperature ($\sim 500{^\circ}C$), but exfoliation was reduced at room temperature and at higher temperatures ($\sim 800{^\circ}C$). High irradiation temperatures ($\sim 1300{^\circ}C$) produced a surface covered with pinholes. Evans (1975) has suggested that thermal vacancies are available at high temperatures so that insufficient pressure is built up within the bubbles to deform the metal surface. Instead the bubbles grow until they intercept the surface, producing a pinhole effect. Therefore it appears that the degree of blistering and exfoliation of blister skins is maximised if the temperature is high enough so that the surface can be easily deformed, although sufficiently low so that the implanted helium release rate from the surface is low.

Gas re-emission measurements associated with blister rupture have been made for several different target-projectile combinations. Extensive studies of helium gas re-emission from niobium, molybdenum, vanadium, palladium and stainless steel during irradiation have been performed by
Bauer et al. (1972, 1973 and 1974) for different target temperatures and total doses. These experiments have revealed an abrupt change in gas re-emission occurring at the critical dose at which blistering was observed. The gas build-up prior to the sharp increase in gas release may account for the excessive re-emission rates compared with the incoming ion flux. After the gas burst the re-emission rate has a relatively low value. The gas re-emission effect appears to decrease with increasing temperature in a consistent manner associated with the changes in surface topography. Hydrogen gas re-emission after irradiation has a different behaviour from helium re-emission under similar irradiation conditions. The hydrogen re-emission rate increases to an equilibrium value after a certain dose (Bauer and Thomas (1974)). Furthermore no periodic re-emissions occur as with helium irradiation of many metals. This is consistent with observation of the lack of exfoliation of multiple blister skin layers which can occur in helium irradiation. This has been attributed to the high solubility and diffusivity of hydrogen in metals (Das and Kaminsky (1975)).

In the hydrogen implantation of metals, the critical dose for blistering is generally higher than for inert gases like helium (due to the high hydrogen permeability in metals). Evidence for this has been found by Kaminsky and Das (1973) from hydrogen and helium irradiation of niobium with 500 keV ions. Blisters were observed at helium doses < 6 \times 10^{17} \text{ ions cm}^{-2}, whereas blisters were not observed during hydrogen irradiation to the same dose at room temperature. The effect of temperature on the critical dose for blistering has been studied by Das and Kaminsky (1973) where niobium was irradiated with high energy (500 keV) helium ions at both room temperature and at 900°C. A decrease in critical blistering dose from \sim 2 \times 10^{18} \text{ ions cm}^{-2} to < 6 \times 10^{16} \text{ ions cm}^{-2} was observed. At the lower
energies (< 15 keV) the decrease in critical blistering dose is not expected to be greatly influenced by the target temperature (Das et al (1975)).

4.6 Theories of bubble and blister formation.

Following the extensive work done in the study of radiation blistering, several theories have been proposed to qualitatively describe the experimental observations. In this section the main theories are reviewed, the mechanisms of bubble formation and growth are discussed and the blistering theories examined. The formation of the bubbles has been attributed to the energy transferred by the energetic projectile to the lattice atoms, producing Frenkel pairs throughout the range of the projectile. The amount of damage, the damage profile and the range distribution are dependent on the projectile energy. The implanted atoms therefore become trapped at vacancies forming small gas filled bubbles. Thus the initial growth of the bubbles depends on the implanted gas concentration and the number of vacancies present. It would be expected that the gas bubbles growth would be subject to the temperature at which the implanted gas atoms and vacancies are mobile. However at temperatures where vacancy mobility is low, the bubble growth will be extremely difficult for the overpressurised gas bubbles to acquire sufficient vacancies to achieve equilibrium conditions.

Several mechanisms have been proposed in which the gas filled bubbles can grow in the absence of a net vacancy flux. Evans (1976) has suggested a possible mechanism, where atoms are removed from the bubble surface by displacement processes due to the energetic projectiles transferring energy
Fig. 4.8  A schematic diagram of the loop punching mechanism proposed by Evans (1978): (a) excess bubble pressure deforms surrounding atom planes, (b) shunting process allows expansion of bubble and creation of interstitial loop.
Greenwood et al (1959) have studied bubble growth in lamp filaments and proposed a different mechanism where the bubble growth was controlled by a loop punching process. A further mechanism has also been proposed by Evans (1978), where the internal gas pressure within the bubble could be sufficient to create a vacancy-interstitial pair at the bubble surface, and bubble growth occurring by the injection of an interstitial into the surrounding lattice. This mechanism is similar to that proposed by Kornelsen (1972). Evans (1978) has considered each mechanism in detail and has concluded that bubble growth is mainly controlled by the loop punching mechanism. A schematic diagram of this mechanism is shown in Fig. 4.8, where the excess bubble pressure deforms a surrounding atom plane (a), expansion of the bubble occurs by the creation of an interstitial loop (b). The further growth of the bubbles into surface blisters has been described by several theories and these are summarised briefly in the rest of the section.

The main theories concerning blister formation have been based on models to explain the experimental observations and falls into three categories. These consist of a bubble coalescence model, an atomic percolation and a stress model. The early studies of radiation blistering lead to the development of a gas driven model to explain the internal release of the trapped implanted gas atoms. The model is based on the coalescence of bubble in the formation of helium blisters in metals and was independently suggested by Das and Kaminsky (1973), Blewer and Maurin (1972), McCracken (1974) and Evans et al (1975). The basis of the model arises from the correlated evidence from high energy irradiation studies, where trapping sites produced by ion irradiation become filled with implanted gas atoms and form small (2 to 4 μm diameter) bubbles. Although the solubility and
diffusivity of helium atoms in metals is low, bubble growth occurs by the absorption of vacancies and the diffusion of helium atoms. Thus, as the total dose of implanted atoms increases, the density of the small bubbles increases to high densities at the larger doses ($>10^{17}$ ions cm$^{-2}$). Evans et al (1975) have observed high bubble densities in both the blister skin and in the regions between the blisters.

The bubble growth and blistering process in the bubble coalescence model is schematically depicted in Fig. 4.9, where the first two stages (a) and (b) correspond to the incident ions forming small bubbles about the mean projected range. The transition from stages (c) to (d) occurs very rapidly for a small increase in the implanted ion dose (Thomas et al (1975)). Evans (1975) has suggested that the coalescence of bubbles becomes rapid when the volume swelling from the gas bubbles in the implanted region reaches a critical value. Therefore the critical dose for blistering is governed by the critical volume swelling caused by the gas bubbles, where the bubble density is sufficiently high so that coalescence of small bubbles can occur. The coalescence of the bubbles then rapidly occurs leading to unstable high pressure cavity formation (e) and to plastic deformation of the surface layer. Thus the large bubble grows until the pressure on the surface exceeds the yield strength of the metal, leading to blister formation.

The gas pressure $p$ in a small bubble prior to coalescence can be determined from a knowledge of the surface energy $\gamma$ and the bubble radius $r$, using the well known expression:

$$p = \frac{2\gamma}{r} \quad (4.20)$$
Fig. A.9 A schematic diagram of the gas bubble coalescence model (see text).
Roth (1975) has developed this relation to show gas pressure dependence on bubble radius for a range of surface energies, where for $\gamma$ about $2 \times 10^{-4}$ J.cm$^{-2}$ and a 2nm bubble the gas pressure within the bubble is about $2 \times 10^3$ atmospheres. Evans (1976) has similarly calculated the number of gas atoms as a function of bubble size, shown in Fig. 4.10 where the gas atom densities have been determined from several mathematical approaches (eg. perfect gas equation, Van der Waals equation and experimental measurements (Bridgeman (1924)). The calculated helium atom densities within the bubble reveal a small variation of helium density with pressure and bubble size (Evans (1976)). However the effect of successive coalescence when the bubbles are close enough to interact at constant volume, will cause a rapid increase in the excess internal pressure, sufficient to deform the metal and create blisters. The blister size $d$ can be related to the pressure $P$ on the surface which exceeds the yield strength $Y$ of the metal from the relation (McCracken (1975)):

$$d = \frac{2RY}{P}$$

(4.21)

where R is the projected range of the projectile in the solid. For known values of $P$ and $Y$ the ratio $d/R$ can be estimated and compared with experimental measurements, McCracken (1975) has found a reasonable agreement with this ratio at higher energies ($> 15$keV).

A second model based on percolation theory and proposed by Thomas and Bauer (1973) and Wilson et al (1974); considers implanted atom-clusters to form blisters. In this model the diffusion coefficient of the implanted gas atoms is assumed to be concentration dependent. Thus the implanted
Fig. 4.10 The number of gas atoms as a function of bubble size, determined from the use of:
(a) perfect gas equation
(b) Van der Waals equation
(c) Bridgeman's data
(Evans (1976)).
atom concentration increases rapidly when the implanted atoms are in a dense region. The model considers the concentration required for the formation of an infinitely connected region of implanted atoms to occur using percolation theory. The atomic fraction of the implanted gas atoms (e.g., helium) present in the metal lattice for the interconnection of the helium atoms is dependent mainly on the type of metal lattice (e.g., bcc or fcc). Therefore, following connection, the helium atoms become mobile along the infinite chain, forming a mobile helium layer. When the implanted gas pressure exceeds the critical pressure required for surface deformation, blister formation occurs. Thus in the percolation model the critical dose for blister appearance corresponds to the ion dose at which the implanted (helium) atomic fraction reaches the onset of percolation. This occurs for an implanted atomic fraction of 0.243 for bcc metals.

The percolation model however, does not consider the yield strength of the metal, which can be strongly dependent on target temperature (Das and Kaminsky (1975)). Furthermore, no account is taken of the target microstructure which can influence the critical dose for blister formation. The effect of high concentrations of implanted gas atoms in the form of bubbles at the higher doses is not directly dealt with in this model. However, Wilson et al (1974) have determined the critical dose for blistering as a function of the diffusion coefficient of isolated atoms in high energy helium irradiation of niobium and palladium targets. A reasonable agreement was found, although radiation enhanced diffusion of the helium atoms was taken into account at higher temperatures. The model also predicts that the critical dose for blister appearance decreases with increasing dose rate, a feature observed experimentally by Verbeck and Eckstein.
The stress model of blister formation was proposed to explain the observations of blister skin thicknesses exceeding the depth of the helium peaks (helium concentration at the mean projected range) at which the bubbles are nucleated, especially at low irradiation energies (<15keV). EerNisse (1973) originally suggested that large lattice forces could lead to severe changes in the atomistic behaviour of the vacancy and interstitial diffusion as well as the implanted gas atoms. Roth (1975) and Behrisch et al (1975) have proposed that the blister formation in low-energy irradiated targets (<15keV) may be caused by stresses induced in the implanted layer. Thus leading to an increased blister skin thickness which cannot be explained in gas driven models of blistering. In the stress model, helium atoms are initially trapped in radiation induced vacancies which precipitate into small bubbles; the bubble density increasing with dosage ions. A shear stress is assumed to form with a value proportional to the implanted atom concentration gradient. The shear stress has a maximum value between the implanted layer and the bulk material. Due to volume swelling, large lateral stresses are induced which can be the driving forces for the large implanted gas bubbles. The lateral stresses are proportional to the volume swelling and have maximum values in the region of gas bubbles as illustrated in Fig. 4.11.

As the forces are parallel to the surface and balanced by the surrounding material, the implanted layer exists in an equilibrium position. However during large bubble growth, an unstable equilibrium position can occur. The resultant large forces can then rapidly deform the surface layer. Coalescence events of the large bubbles, results in excess pressure which
Fig. 4.11 A schematic diagram of the lateral stress model of blistering (Roth (1975)).
determines the spherical shape of the deformations. Consequently the blister skin thickness is approximately equal to the width of the implanted layer which may be considerably larger than the mean projected range of the low-energy projectiles. Thus the amount of implanted atoms trapped in the large gas bubbles and released during blister formation will be small compared with the total amount of implanted atoms in the blister covers. At high projectile energies (>15keV) the distribution of implanted ions is broader, and the corresponding shear stress values lower than the low projectile energies, thus exerting minimum influence on bubble and blister formation.

A simple stress model has been proposed for high energy (>20keV) helium blistering in metals (Auciello (1976)), where through the nucleation of smaller bubbles, a critical bubble radius is reached. The model is based on an earlier planar stress model for a semi-infinite plate which is assumed valid and extended to the three dimensional case. The blister skin thickness and the critical dose for blistering can be determined from the values of the mean projected range and the known blister diameters. Furthermore, the resulting values are in close experimental agreement with reported experimental values and the model can quantitively explain the fracture mechanism of blisters at high temperatures. However, although the stress model was proposed to explain the increased blister skin thickness observed under low-energy irradiation, the model also predicts an increase in helium density (and thus pressure) with increasing dose as the bubbles grow. This is not the situation in the growth of equilibrium bubbles or bubble growth by loop punching (Evans (1978)). Also in the stress model the level of stresses attained in the irradiated surface layer has no assumed upper limit, which is inconsistent with experimental observations.
Fig. 4.12 A schematic diagram of the interbubble fracture model proposed by Evans (1978):

(a) high density of overpressurised bubbles,
(b) crack formation,
(c) bubbles adjacent to the original crack become involved to widen crack and increase pressure,
(d) penny shaped crack which either extends to cause flaking or
(e) forms blister by gas driven surface deformation.
A modified gas coalescence model which includes a larger blister skin thickness has been proposed by Evans (1978), which concerns the gas driven bubble growth and the stress within the implanted region. This model has been called the interbubble fracture model, where it is proposed that as the bubbles grow and decrease the interbubble spacing, and the added internal pressure of several adjacent bubbles will eventually create a local stress sufficient to crack open the plane adjoining the bubbles. A schematic outline of the mechanism is shown in Fig. 4.12, where the high density of overpressurised bubbles (a) leads to crack formation in (b). The bubbles adjacent to the original crack become involved to increase the crack width and also produce an increase in pressure. The elongated crack thus formed (d) can therefore extend to cause flaking or can form a surface blister (e) by a gas driven surface deformation. Furthermore a theoretical approach also indicates that two processes can occur so that the bubbles can relieve the increased pressure by a loop punching mechanism below some critical bubble size and pressure. Above which interbubble fracture process can occur. Therefore extensive bubble coalescence and internal implanted gas release will then occur and lead to the formation of surface blisters or flaking.

Evidence supporting the gas driven model as opposed to the lateral stress model has been obtained by Evans and Eyre (1977) from high energy (100keV) helium irradiation of molybdenum. The wedge shaped specimen after irradiation revealed blisters formed on the unbombarded side at the thin section of the wedge, prior to blisters observed on the bombarded side. This behaviour is consistent with a gas driven model. Furthermore in the model the fracture plane where the interbubble coalescence occurs does not coincide with the helium peak concentration, but at a region of high local
stress concentration leading to the fracture mechanism, thus leading to the observation of greater blister skin thicknesses.

Among the three basic blister models proposed which have been described, the coalescence model attempts to qualitatively explain many of the experimental observations. However, no single model considers all the parameters governing blister formation and growth sufficiently to explain the various observations. Although a unified quantitative theory has not been developed, further work will be required before such a model can be deduced.
CHAPTER FIVE

Low-energy ion irradiation of tungsten and molybdenum

5.1 Introduction

In this chapter the radiation damage produced by low-energy hydrogen and helium ions in tungsten and molybdenum specimens is investigated. The FIM technique is a powerful tool for the observation of low-energy damage, especially concerning the nucleation and growth of voids leading to blister formation. Furthermore, the use of controlled field evaporation allows the precise nature and depth of damage to be determined, which is difficult or impossible to obtain with other techniques.

The low-energy irradiation of the field-ion specimens has been performed using several different ion sources. A preliminary study is conducted employing the cathode sputtering technique to establish the ion energies and doses necessary to form defect complexes. Further irradiation experiments are conducted using a mass-analysed ion source. However, the majority of the work presented in this chapter is conducted with a high current density ion source, where the ion beam is incident in a parallel direction to the emitter axis. Thus enabling the depth of damage to be determined for a given ion energy and dose.

The helium irradiation of tungsten is extensively studied in the ion energy range 0.2 to 3 keV with corresponding doses of $1 \times 10^{15}$ to $5 \times 10^{17}$ ions cm$^{-2}$. The analysis of the irradiated specimens is...
performed by controlled field evaporation, where the size of the
different defects and the depth of damage is recorded for each ion
energy and dose combination. The study enables the growth of the
defects and the corresponding depth of damage to be monitored.
Similar experiments are performed for molybdenum specimens. The
general features and trends of the irradiation damage with in-
creasing ion energy and doses are discussed in relation to the
current theories of bubble formation and blistering.

5.2 Previous FIM studies of irradiation damage.

5.2.1 Low-energy (0-1-10keV) ion bombardment.

The early work performed using the FIM to observe damage in
irradiation specimens was conducted using the cathode sputtering
technique. Müller (1960) and Dranova and Mikhailovski (1970) observed
the formation of dislocations using this technique in tungsten specimens
irradiated with (<10keV) helium ions. Müller has termed these defects
as "shallow dislocation loops", due to the low penetration of the
defects of only 10 to 20 atomic layers beneath the surface. Brandon
et al (1963) and Wald (1963) have studied the damage produced by low
energy (<100eV) neutral argon atoms in tungsten to determine the
threshold displacement energy. However, due to imaging conditions
during irradiation, large threshold energy values ~70eV were recorded.

The study of low energy (150 to 450 eV) argon irradiation of
tungsten has been performed by Gregov and Lawson (1972), where point
defects and clusters of vacancies and interstitials were observed after
400 and 450 eV argon ion irradiation. During irradiation at low temperatures (~630K), point defects only were observed at the lower ion energies (150 to 300 eV). Furthermore self diffusion occurred on annealing from 63 to 780K where vacancies and interstitial atoms were observed to diffuse to the surface during the annealing process. The amount of damage was found to be greater in the non-irradiated side compared with the irradiated side of the specimen. A further study of 400 eV argon irradiation of tungsten (Gregov and Lawson (1973)) using field evaporation sequences showed that a focused collision sequence may be responsible for vacancy formation on the non-irradiated surface. A further study of argon ion-bombardment of tungsten and rhenium has been reported by Cranston et al (1973). The damage was found to consist of vacancies and their clusters together with interstitial contrast. Dark streaks were also observed, which were attributed to bombardment induced faceting. The depth of damage in the irradiated tungsten after field evaporation was found to extend from between 1(011) and 20(011) removed planes after bombardment with argon ions with energies of 100 and 470 eV respectively.

A comparative FIM study of the effects of low-energy (<1keV) ion bombardment of tungsten has been conducted by Walls et al (1973) and (1974) using the inert gas ions (viz, He, Ne, Ar and Xe). The results of the low dose irradiation showed that extensive surface and sub-surface damage was produced, commonly in the form of vacancy and interstitial clusters together with the occurrence of dislocation contrast. The range of the heavier inert gas ions was found to be in general agreement with theoretical calculations. The use of inert ion bombardment for sputter-cleaning or ion profiling application has also been studied
with the FIM technique (Walls and Southworth (1976)). Helium radiation damage has been observed by Walls et al (1976) using the cathode sputtering technique with ion doses of up to $5 \times 10^{17}$ cm$^{-2}$. The successive removal of planes by field evaporation revealed extensive sub-surface damage, in the form of surface craters, voids, star shaped clusters and dislocation loops and complex networks of these defects. The damage was found to be dose dependent, the depth of damage increasing with ion energy, which in turn depends on the potential applied to the specimen.

Further low energy (<200eV) 'in-situ' irradiation of tungsten has been performed by Panitz (1977) using deuterium ions in an imaging field desorption mass spectrometer. The relative abundance of deuterium was determined for 80eV deuterium ions, which were implanted to depth of nearly 5nm beneath the irradiated surface. The technique also allowed the implanted surface species to be determined, which for carbon and oxygen was about 2.0 and 3.0nm respectively for 200 eV deuterium ions incident on the surface. The range profiles of low energy (300 and 475 eV) helium ions implanted in tungsten at low temperatures (60 to 90°K) have been determined experimentally by Wagner and Seidman (1978) employing an atom-probe FIM. The mean ranges of the 300 and 475eV ions which were incident in a parallel direction to the tip axis were found to be 2.0 and 5.6 nm respectively by pulsed field evaporation measurements. Isochronal annealing experiments conducted after irradiation further showed that the helium atoms were located in interstitial lattice positions which were immobile at 90°K but highly mobile at 110°K.
The study of low energy (0.5 to 9keV) inert gas (viz., He, Ne and Ar) ion irradiation of tungsten has also been performed by Tarnaki (1976) at low doses using a field emission ion source. The irradiating ions were incident on the (011) orientated emitter from the <110> direction, at right angles to the tip axis. Large scale damage in the form of vacancy clusters were not observed, furthermore, damage was evident on the un-irradiated surface. This was attributed to a channeling process. However, the observation of surface damage as a function of incident ion energy enabled the displacement energy of the surface atoms to be measured. For tungsten the value was found to be 23 ± 2eV, in good agreement with neon irradiation experiments. The total displacement energy for an internal atom was proposed in the range 30 to 37eV for a tungsten target.

5.2.2 High-energy (10-100keV) ion bombardment.

Sinha and Muller (1964) have irradiated tungsten with energetic (20keV) neutral helium atoms; the resulting damage was found to consist of vacancies and interstitials and their respective clusters. The results of the low dose irradiations indicated a threshold displacement energy of 50eV, and about half the number of internally displaced atoms were found to appear at the surface due to focusing collisions.

A quantitative FIM study of 'in-situ' irradiation of tungsten by energetic (20keV) self ions (W⁺) to doses of 1 x 10¹² ions. cm⁻² has been performed by Beavan et al (1971). This detailed study found that depleted zones were produced, revealed by a pulsed field evaporation technique. The depleted zones were presumed to be produced by single
atom collisions, with lower numbers of displaced atoms than predicted by the Kinchin-Pease model. Moreover, self interstitial atoms (SIA's) were found locally at the peripheral surfaces of the depleated zones. In a further study of self ion irradiation of tungsten, Scanlan et al (1971(a)) have also demonstrated that SIA's are immobile at low irradiation temperatures (~150K), and field effects did not produce any extensive long-range stress induced migration of the SIA's. Extensive damage has been observed in a study of tungsten irradiated with high energy (150keV) mercury (Hg\textsuperscript+) ions (Buswell (1970)). The damage was found to consist mainly of surface craters and subsurface voids resulting from collision cascades originating close to the surface of the specimen.

The FIM technique has been used to determine the damage and implant profile in iridium irradiated with energetic (10-60 keV) ions of carbon, oxygen xenon, silver and iron to high doses (~1 \times 10^{19} \text{ ions. cm}^{-2}) (O'Connor and Ralph (1971)(a)and (b)). The irradiation occurred in a direction parallel to the tip axis and the spatial distribution of the damage was studied by successive field evaporation. Hudson and Ralph (1972) have irradiated iridium with energetic (20-100keV) heavy inert gas ions (viz. Ne, Ar and Xe) with doses in the range 10^{11} to 10^{16} ions cm\textsuperscript{-2} to simulate the damage produced by fast neutron irradiation. Following irradiation at 78\textsuperscript{0}K, single vacancies, dispersed and compact clusters together with dislocation loops were observed. The size of the dislocation loops and compact vacancy clusters were found to be independent of ion dose and energy above ion energies of 60keV.
5.2.3 Neutron irradiation

The migration of $<110>$ split interstitials and vacancy motion have been observed in tungsten irradiated by fast neutrons by Attardo et al (1967). The use of annealing experiments to study the recovery processes of neutron irradiated tungsten has been performed by Bowkett and Ralph (1969). Further work by Jennotte and Galligan (1970) in the kinetics of vacancy removal in neutron irradiated tungsten have shown that vacancies are mobile in the temperature range 700 to 900°C. Rao and Thomas (1967) have also studied the radiation damage produced by fast neutrons in molybdenum. They found that large numbers of vacancies were not produced although interstitial and impurity contrast was observed. A FIM study of voids in molybdenum produced by fast neutron irradiation at high total doses ($\sim 10^{22} \text{ nc m}^{-2}$) has been conducted by Brenner and Seidman (1975). The controlled field evaporation sequences of the irradiated surface revealed numerous small voids ($<8\text{nm in diameter}$). Moreover, complex atomic displacements were observed surrounding the voids when the voids intersected the surface. The formation of small microvoids in a neutron irradiated iron-copper alloy has been observed by Brenner (1978), where the size of the micro-voids ($<1\text{nm}$) was determined by controlled field evaporation.

5.3 Specimen preparation

In this study two preparative techniques have been used to produce field-ion specimens from commercial grade tungsten and molybdenum wires. The two techniques employed involve the electropolishing of a
wire to a fine point and are described in detail:

(a) The floating-zone technique.

The floating-zone or thin-layer technique is used for tungsten where a small quantity (about 5-10mL) of etchant is placed above an inert solution contained in a Petri dish. A small portion of the tungsten wire (0.25 mm diameter) held in a copper specimen holder is then positioned vertically through the electrolyte layer into the inert solution which is usually carbon tetrachloride. The electrolyte used for tungsten is oxidised Gevaert G150 photographic developer. A counter electrode is also immersed in the electrolyte, usually in the form of a hemicylindrical copper plate. An AC voltage (~10V) is then applied between the electrodes and the current monitored as the wire is electrolytically dissolved in the electrolyte layer. During etching the wire forms a 'neck' and the current is reduced, etching is then continued until the bottom piece of the wire drops away, leaving a finely taper wire with a pointed tip. Etching is allowed to continue for a further 10 seconds to remove any fragments and deformed material from the tip. The sharpened tip is removed from the electrolyte, washed in distilled water and rinsed in acetone. The specimen is then ready for insertion into the FIM for examination and characterisation prior to any experimental work.

(b) The single-layer technique.

In this electropolishing technique, normally used for molybdenum specimen preparation, the wire is mounted in a copper specimen holder and directly immersed into the etchant. This is usually
achieved by a vertically positioned glass 'U' tube, where the wire is placed in one arm and a stainless steel counter electrode in the other arm. The electrolytes used are usually a 10% aqueous solution of sodium hydroxide or alternatively 10% solution of hydrochloric acid may be used. Polishing is achieved by the application of a 10V AC voltage across the electrodes. A microscope is used (x 50 magnification) to monitor the progress of the polishing of the tip by back illumination. The etching rate may be accurately controlled by the variation of the voltage applied to the electrodes, although considerable expertise is required with periodic observation of the polished tips to gain consistency of specimen preparation. The electropolishing of the wire usually results in a finely tapered wire with a sharp tip, at which the polishing process is stopped. The specimen is subsequently removed from the electrolyte and washed with distilled water, rinsed with acetone and imaged in the FIM. Further polishing may be necessary to produce suitable molybdenum specimens for irradiation studies.

5.4 Preliminary experimental work

In the preliminary experimental work, two separate irradiation techniques were used in a series of experiments to establish the effect of ion energy and irradiation dosage on the analysis of the emitter surfaces. The two irradiation techniques consisted of field induced ion bombardment (cathode sputtering) and direct ion-bombardment by a mass-analysed ion source. The combined use of the two techniques in the FIM provide different forms of information regarding the analysis of the irradiated emitter surface. In the following section the
Experimental work of the two techniques is discussed and the results provide the basis of the main experimental irradiation studies reported in the subsequent part of the chapter.

5.4.1 Field induced ion bombardment

In a series of experiments the specimens were prepared as previously discussed and placed within the FIM where 'in-situ' ion irradiation was performed at 78°C using either hydrogen or helium gases. The experimental procedure was followed so that a clean well characterised emitter surface was produced by suitable field evaporation. The imaging gases were then quickly removed from the vacuum chamber until the base pressure of the system was attained (about $5 \times 10^{-10}$ Torr) while the imaging voltage was still present on the emitter. The polarity of the electric field was then reversed and a suitable negative potential in the range 1 to 5 kv was selected. The irradiating gas species was then allowed into the chamber via a UHV leak valve to a preselected operating pressure in the range $1 \times 10^{-4}$ Torr to $1 \times 10^{-6}$ Torr. This procedure tends to reduce any contamination effects which may occur in the vacuum chamber due to any residual contaminant gases. The ion dosage may be directly controlled by the pressure of the irradiating gas species together with the ion current incident on the specimen as a function of ion bombardment time, typically between 1 to 10 mins. The current flowing through the specimen was monitored using an isolated Keithley 602 picoameter in series between the specimen and the high voltage supply. Following ion-bombardment for the required duration the irradiating gas was removed and the applied potential on the specimen
was returned to a positive polarity. The specimen was immediately reimaged on introduction of the imaging gases and the emitter surface analysed for irradiation damage, with subsequent depth analysis by field evaporation. The total ion dosage was approximately determined using equation 2.8 as previously discussed in section 2.6.

In a previous cathode sputtering study of helium irradiation of tungsten conducted by Walls et al (1976) at low ion energies (<3 keV) and with doses in the range $10^{13}$ to $5 \times 10^{17}$ ions.cm$^{-2}$. Minimal surface damage was observed at the lower doses. The image contrast on the emitter surface following low energy He irradiation consisted mainly of small numbers of vacancies and interstitials 'bright spot' contrast. The surface vacancy formation was attributed to sputtering effects which existed close to the original irradiated surface. The vacancies disappeared after the removal of several apex planes during field evaporation. At higher doses ($> 5 \times 10^{16}$ ions.cm$^{-2}$) however, considerable surface damage was observed in the form of voids and star shaped clusters of vacancies. The voids having diameters in the range 0.5 to 3.5 nm. Dislocation loop contrast was found after successive field evaporation with continued vacancy and vacancy cluster contrast at depths up to 40(011) layers ($\sim$ 9nm) below the surface. Increased damage of similar contrast was also observed at higher doses ($> 5 \times 10^{17}$ ions.cm$^{-2}$) and at higher applied potentials (5 kV) the irradiation damage extended to a depth of 70(011) planes (about 16 nm) below the irradiated surface.

Several experiments were performed using high doses ($> 1 \times 10^{17}$ ions.cm$^{-2}$) at low helium ion energies of 1keV and revealed extensive surface damage on the emitter, especially at doses $> 5 \times 10^{17}$ ions.cm$^{-2}$,
where catastrophic tip failure occurred. Thus the high dose limit of $5 \times 10^{17}$ ions cm$^{-2}$ was adopted to reduce specimen failure and enabling useful information to be gained from these experiments.

A further series of experiments were conducted using hydrogen irradiation of a tungsten specimen at low applied voltage corresponding to an ion energy of 1keV. The hydrogen ion dosages were approximately $1 \times 10^{13}$, $2 \times 10^{16}$ and $5 \times 10^{17}$ ions cm$^{-2}$ respectively. The damage at the low doses was negligible and consisted mainly of surface vacancy contrast with little void formation. The extent of the vacancy and interstitial contrast increasing with dose and the depth of damage increasing from approximately 2.5 to 4.4 nm respectively. However at higher doses ($5 \times 10^{17}$ ions cm$^{-2}$) several large voids were observed. The irradiation damage is illustrated in Fig. 5.1 where Fig. 5.1(a) shows the characterised surface prior to ion bombardment. The subsequent micrographs show the position of a large void after successive field evaporation. Fig. 5.1(b) was recorded after 5(011) apex planes (1.1nm) were removed from surface, a large void is evident near the (011) plane manifest as a dark contrast area. The void was located close to the surface of the irradiated emitter and a second void can also be distinguished close to the (101) plane.

Fig. 5.1(c) depicts the surface after 7(011) planes were removed (1.5nm) where surface atoms at the base of the void produce a bright spot contrast in the image. One noticeable feature in this micrograph, is the slight distortion of the atoms at the periphery of the void where a slight inward relaxation of the surface atoms position is observed. This is manifest by the translation of the planar rings (011) for example intersecting the void at the surface. This effect has been observed
Fig. 5.1 The damage produced in an (011) orientated tungsten specimen by bombardment with $\sim 5 \times 10^{17}$ hydrogen ions cm$^{-2}$, using a field emission potential of -2kV. The circles indicate the voids present in the tungsten emitter.

a) Reference surface prior to bombardment.
b) The damage at a depth of 5(011) layers (1.1nm).
c) The damage after 7(011) layers removed (1.5nm).
d) The damage after 19(011) layers removed (4.2nm).
e) The damage after 38(011) layers removed (8.5nm).
f) The damage after 73(011) layers removed (16.1nm).
by Seidman et al (1975) and has been attributed to an enhanced atom relaxation due to the interaction of the void with the emitter surface.

Following continued field evaporation the void decreases in size as shown in Fig. 5.1(d) after 19(011) planes have been removed (4.2nm). The central void has nearly been removed from the emitter surface after a total field evaporation of 38(011) planes (8.5nm) from the emitter as shown in Fig. 5.1(e). Several bright streak and interstitial contrast is evident, particularly from the void located near the (101) planar region. The bright streaks may be due to field effects from protruding atoms, which are affected by the pressure of the large void. Fig. 5.1(f) depicts the emitter surface after 73(011) planes (16nm) have been removed. A relatively well characterised emitter surface is evident although the void close to the (101) planar region is still present.

The central void persisted for the removal of 38(011) planes and corresponds to a void size $\sim 8.5$nm. The second large void persisted for the total number of field evaporated planes, however, due to the angular orientation of the void, the actual void size is $\sim 9$nm. The voids were initially observed close to the surface if the irradiated emitter. This compares favourably with the total range of 1keV hydrogen ions in tungsten which from LSS theory was found to be $8.6$nm. However the mean projected range is $5.0$nm which suggests that voids exist at depths greater than the mean projected range. At the higher doses, catastrophic tip failure occurred.

Some preliminary work was done on hydrogen irradiation of molybdenum specimens at low ion energies (500eV) and at low doses
<10^{14} \text{ions.cm}^{-2}. \text{Minimal irradiation damage in the form of small numbers of vacancies and vacancy clusters were observed with a lack of planar defects visible in the image. The void sizes were small, less than 0.8nm, although this was difficult to determine exactly due to the image contrast from molybdenum at 77^0\text{K}.}

The preliminary study has shown that the field induced ion bombardment technique can yield qualitative data regarding the irradiation of tungsten and molybdenum specimens with hydrogen or helium ions. However the technique does suffer from some disadvantages, in that the ion-bombardment produces a large spread in the number and energy of the ions incident over the entire specimen surface. Furthermore it is difficult to obtain exact ion dose measurements due to the nature of the ionisation process and the actual area of ion collection to determine accurate ion dose measurement. However, the preliminary study using this technique has shown that the range of ion doses at low energies (<3keV) which are of interest for irradiation studies using the FIM lies between 1 \times 10^{15} to 5 \times 10^{17} \text{ions.cm}^{-2} for hydrogen and helium irradiation.

5.4.2. \textit{Mass-analysed ion bombardment.}

A preliminary irradiation study was conducted using a specially designed mass-analysed ion source (see section 2.4). The ion source is capable of producing low-energy(<3\text{keV}) helium ions for 'in-situ' irradiation experiments in the FIM. The mass-analysed source was used to ensure that the correct ion species were bombarded on the specimen surface by the use of the Wien filter.
The experimental arrangement for the 'in-situ' irradiation is similar to that shown in Fig. 2.14, where the ion beam is incident at an oblique angle onto the specimen. The ion source was operated in a dynamic gas mode with constant leak rate to achieve a gas pressure of about $4 \times 10^{-4}$ Torr. The ion source was operated under optimum conditions and Wien filter voltages (see section 2.4) with helium ion energies of 1 and 2 keV respectively. Electrostatic charging effects due to electrically isolated components in the FIM (screen, channel-plate etc.), were removed by earthing the appropriate feedthroughs. The ion currents measured by a Keithley 602 picoammeter connected between the specimen and earth were low, typically about 0.5 μA. The total dose was calculated from previous work (section 2.4) for the known source operating conditions and ion beam characteristics and is dependent on the total irradiation time. To achieve high doses of $1 \times 10^{16}$ ions.cm$^{-2}$, irradiation times of up to several hours are required (~3.5 hours) due to the low ion current densities incident on the specimen.

In this preliminary study, a series of irradiation experiments were conducted on tungsten with doses in the range $2 \times 10^{13}$ to $1.2 \times 10^{16}$ ions.cm$^{-2}$ at an ion energy of 2keV. The previously characterised tungsten specimens were irradiated for a known duration under well defined dynamic irradiation conditions. The irradiated surfaces were then immediately imaged in a He-Ne gas mixture and the resulting damage observed.

At the lower doses the emitter surface showed little surface damage and was quickly removed on the field evaporation removal of
several apex planes. The damage consisted mainly of surface vacancies and bright spot contrast. The damage region was localised to the incident ions on the surface, which due to the oblique angle occurred at the periphery of the image, thus making a proper analysis difficult to assess. Increasing the ion dose to $5 \times 10^{15}$ and to $1.2 \times 10^{16}$ ions $\text{cm}^{-2}$ respectively resulted in greater surface damage again in the form of surface vacancies and bright spot contrast. The damage region of the emitter surface was removed by slow evaporation within 10(011) planes of the original irradiated surface. This corresponds to a depth of less than 2nm. Furthermore only one side of the emitter surface was exposed to the incident ion beam, thus reducing the depth profiling ability of the FIM technique. At these doses small voids ($<0.5\text{nm}$) were distinguished, together with some planar defects in the form of split planar regions, although these were difficult to distinguish due to the location of the damage around the peripheral side of the image. The helium irradiation of an annealed tungsten specimen ($1000^\circ\text{C}$ for 10 mins. at $< 1 \times 10^{-9}$ Torr) by 2keV ions was also performed with similar results, as previously described, with little evidence of total damage to the emitter surface for a total duration of 3.5 hours corresponding to a dose of $1 \times 10^{16}$ ions $\text{cm}^{-2}$.

The analysis of the damage revealed that small surface vacancies and their clusters were formed although rapidly removed during a controlled field evaporation sequence. Moreover, the location of the damage at the periphery of the image tends to complicate the size and depth of the voids below the surface. Thus forming a complex picture of the number of voids and the depth distribution within the emitter surface due to oblique irradiation. Further high dose experiments ($\geq 5 \times 10^{16}$ ions).
cm$^{-2}$) were not attempted due to the unduly large duration of the ion bombardment required for these doses (> 4 hours). Prolonged exposure to the high helium pressures may cause surface contamination of the emitter surface, by residual gases during ion-bombardment. However, the advantages offered of a precise knowledge of the ion species and energy during irradiation may be particularly suited to sputtering studies of well defined regions of the emitter surface for low dose conditions using inert gas ion-bombardment.

5.5 Helium irradiation of tungsten.

In the following study the results of the irradiation damage produced by low-energy (< 3keV) helium ions which are incident at a parallel direction to the specimen axis is reported. In a series of experiments the two factors governing the defect growth and bubble formation are investigated. The main factor is the total ion dosage which determines the growth of the bubbles, whereas the energy of the incident projectile is related to the extent of the surface damage. The study of helium irradiation has been separated into three areas of interest dealing with low and high doses (> $1 \times 10^{16}$ ions.cm$^{-2}$) in ion the energy range 1 to 3keV. The final section deals with low energy (< .5keV) helium irradiation, where the helium ion energy transferred energy due to a collision event is comparable or less than the threshold displacement energy in tungsten.

5.5.1 Experimental procedure.

Tungsten field-ion specimens of [011] orientation were prepared
using an electro-polishing technique from commercial grade (99.9% purity tungsten wire < 0.25mm diameter). The specimens were placed in the FIM where a clean and well characterised emitter surface was produced by field evaporation and the final imaging voltage recorded. The characterised specimens were then transferred to an ion pumped ultra-high vacuum system for helium irradiation. In this system the specimens were orientated so that the ion beam was parallel to the wire axis by means of a specially mounted jig located on a precision XYZ manipulated specimen holder. Precise ion beam current density measurements could be performed with a movable Faraday cup (with secondary electron suppression). Helium irradiation was performed at ambient temperature with a 3keV Varian monoenergetic ion source which has been previously described (2,5). Ion bombardment was carried out under static conditions at a helium gas pressure of $5 \times 10^{-6}$ Torr. The specimens were irradiated at a given ion energy ranging from 0.2 to 3keV for a known duration and the total helium ion dose determined for each specimen. After each irradiation the specimen was returned to the FIM, where the emitter surface was reimaged at 70°C and the irradiated specimen surface was examined. A carefully controlled field evaporation sequence was then performed, where the irradiated specimen surface was photographically recorded for each central (011) plane removed.

A series of control experiments was also performed to determine the effects of atmospheric and residual gas contamination on the field-ion specimens. Several previously characterised specimens were exposed to atmospheric conditions for different times up to several days in a covered container. Re-examination of the emitter surfaces revealed no deleterious effects, where the emitter surfaces were predominately covered
with bright spot contrast due to surface contamination. In all instances a characterised emitter surface was produced after the field evaporation removal of several (< 2) apex (011) planes. A similar observation was recorded for the effects of residual gas contamination experiments, where the imaging field was removed under gas imaging conditions for periods of up to one hour.

The results for an irradiated specimen were analysed from a sequential series of enlarged photographic prints of the emitter surface. The presence of each void over the imaging region of the emitter surface was recorded for the persistence of each void with respect to the number of field evaporated apex (011) planes (void sizing technique). Thus the use of field evaporation allows the sub-surface of the irradiated specimens to be investigated.

5.5.2 Helium irradiation at low doses.

5.5.2.1 1keV helium irradiation.

A range of systematic experiments have been performed to investigate the effects of increasing helium ion doses up to $1 \times 10^{16}$ ions.cm$^{-2}$ in the ion energy range 1 to 3 keV. The results for a given ion energy and dose are presented as a series of field evaporation sequences of the irradiated specimens. The sequence of micrographs from a low dose irradiation experiment are shown in Fig. 5.2 for an ion energy of 1keV and a dose of $1 \times 10^{15}$ ions.cm$^{-2}$.

The original characterised surface after irradiation is shown in Fig. 5.2(a) where isolated vacancy and 'bright spot' contrast
Fig. 5.2  Series of micrographs of 1keV helium irradiated tungsten to a total dose of $1 \times 10^{15}$ ion.cm$^{-2}$. Several small voids are circled.

a) The irradiated surfaces

b) The damage at a depth of 10(011) layers (2.2nm).

c) The damage at a depth of 19(011) layers (4.2nm).

d) The damage at a depth of 28(011) layers (6.2nm).

e) The damage at a depth of 38(011) layers (8.5nm).

f) The damage at a depth of 68(011) layers (15.2nm).
is evident together with small vacancy clusters. Slow field evaporation of the surface layers revealed small numbers of vacancy clusters occurring close to the emitter surface (< 1.5 nm). Fig. 5.2(b) reveals the surface after 10(011) planes have been removed, corresponding to a depth of 2.2 nm. The point defect contrast on the emitter surface at this depth is reduced although small vacancy clusters may be distinguished on the emitter surface. Field evaporation of a further 9(011) planes to a total of 19(011) planes corresponding to a depth of 4.2 nm shows a reduction in the number of point defects as illustrated in Fig. 5.2(c). Several small vacancy clusters in the form of small voids can be distinguished in Fig. 5.2(d) after the removal of 28(011) planes to a depth of 6.2 nm. However, continued field evaporation after approximately 30(011) planes reveals minimal damage on the emitter surface. Figs. 5.2(e) and (f) depict the irradiated emitter surface after 38 and 68(011) planes have been removed, corresponding to depths of 8.5 and 15.2 nm respectively. The micrographs reveal that the irradiated specimen has little point defect damage and the emitter has a relatively well characterised surface. Analysis of the field evaporation sequence shows that the damage was predominantly in the form of isolated defects and vacancy clusters forming small voids with sizes ranging from 0.3 to 0.9 nm and a mean void size of 0.54 nm (standard deviation of 0.35 nm). The voids were found to persist to a depth of approximately 32(011) planes or 7 nm beneath the original irradiated surface.

The effect of increasing the helium ion dose for a specimen irradiated with 1 keV helium ions to a dose of \(5 \times 10^{15}\) ions cm\(^{-2}\) is shown in Fig. 5.3. A total of 80(011) planes (\(\sim 18 \text{ nm}\)) were field
Fig. 5.3
void size and depth distributions after 1 keV helium irradiation in tungsten to a ion dose of $5 \times 10^{15}$ ions cm$^{-2}$.

He-W
1 keV
$5 \times 10^{15}$ ions cm$^{-2}$
evaporated under controlled conditions and the void sizing technique was used to determine the void sizes and the depth below the irradiated surface for which the void persisted. The analysis reveals the void sizes range from 0.2 to 1.2 nm with a mean void size of 0.7 nm (standard deviation of 0.4 nm). The depth distribution of the voids indicates that the large number of voids are formed close to the surface and extend to a depth of about 8 nm below the irradiated surface.

A detailed analysis of the voids produced by 1 keV helium irradiation to a dose of $1 \times 10^{16}$ ions cm$^{-2}$ is shown in Fig. 5.4 for a series of micrographs representing the emitter surface at different depths below the original irradiated surface of the specimen. Fig. 5.4(a) shows the surface of the irradiated specimen after 1(011) plane has been removed at an imaging voltage of 9.8 kV. A large number of bright spot contrast is evident with several topographical features present, which were not observed on the characterised emitter surface. In one feature, contrast corresponding to a dislocation can be distinguished emerging on the (112) plane. A planar defect on the (121) plane is also evident and has a split plane configuration. Isolated surface vacancies and vacancy clusters are also present on the emitter surface. Fig. 5.4(b) shows the emitter surface after $2\frac{1}{2}$ (011) planes (≈ 0.6 nm) have been removed. A slight reduction in the 'bright spot' contrast corresponding to interstitial atoms can be distinguished together with numerous vacancy clusters occurring on several regions of the emitter surface.

In Fig. 5.4(c) the emergence of a grain boundary may be distinguished on the emitter surface in the (001) planar region. Numerous small voids
Fig. 5.4 The tungsten specimen irradiated with 1keV helium ions to a total dose of $1 \times 10^{16}$ ions.cm$^{-2}$. The circles and arrows indicate the presence of void and dislocation contrast respectively.

a) The irradiated surface after nearly 1(011) layer removed ($\approx 0.2$nm).

b) The damage after 2$\frac{1}{2}$(011) layers removed, (0.6nm).

c) The damage after 4$\frac{1}{2}$(011) layers removed (1.0nm).

d) The damage after 5$\frac{1}{2}$(011) layers removed (1.2nm).

e) The damage after 6$\frac{1}{2}$(011) layers removed (1.5nm).

f) The damage after 8$\frac{1}{2}$(011) layers removed (1.9nm).

g) The damage after 10(011) layers removed (2.2nm).

h) The damage after 12(011) layers removed (2.7nm).

i) The damage after 22(011) layers removed (4.9nm).

j) The damage after 30(011) layers removed (6.7nm).

k) The damage after 43(011) layers removed (9.6nm).

l) The damage after 70(011) layers removed (15.6nm).
can be observed and several planar defects are also present on the emitter surface at a depth of 1.0nm. Fig. 5.4(d) shows the surface after the removal of $5\frac{1}{2}(011)$ planes at a depth of 1.2nm. Extensive damage in the form of small voids and planar defects are present on the emitter surface. In Fig. 5.4(e) the surface is shown at a depth of 1.5nm where several voids are evident. In the (112) planar region a dislocation intersects the surface forming complex image contrast. A small void (~ 0.5nm) can be distinguished, several 'bright spot' interstitial contrast are also present at the periphery of the void. Fig. 5.4(f) shows the surface of the irradiated specimen $8\frac{1}{2} (011)$ planes have been removed (1.9nm). Several small voids are present and the planar defects are still evident. A reduction in interstitial 'bright spot' contrast may also be distinguish compared with the previous micrograph, although 'bright spots' are still evident particularly in the (011) planar rings. Fig. 5.4(g) reveals the surface of the specimen at a depth of 2.2nm at an imaging voltage of 9.9kV. A spiral contrast indicating the emergence of a dislocation on the (011) plane is evident. The complex planar defect on the (112) plane is still present. Fig. 5.4(h) shows the surface after 12(011) planes have been removed (2.7nm) where the spiral contrast is still evident on the (011) plane. Several small vacancy clusters may also be distinguished, particularly in the proximity of the (112) planar region. In Fig. 5.4(i) the spiral contrast on the (011) plane has been removed after the field evaporation of 22(011) planes (4.9nm) from the original irradiated surface. However, a complex planar defect present on the (112) plane is still present. Fig. 5.4(h) shows the surface after 12(011) planes have been removed (2.7nm) where the spiral contrast is still evident on the (011) plane. Several small vacancy clusters may also be distinguished, particularly
in the proximity of the (112) planar region. In Fig. 5.4(i) the spiral contrast on the (011) plane has been removed after the field evaporation of 22(011) planes (4.9nm) from the original irradiated surface. However, complex planar defects are present on the (112) and (121) planar regions. Several small voids (vacancy clusters) may also be distinguished in planar regions close to the planar defects. Small voids are still evident however after 30(011) planes have been removed (6.7nm). In this Fig. (5.4(j)) a complex planar defect is still present on the (112) planar region due to the intersection of a dislocation with the emitter surface. Fig. 5.4(k) shows the irradiated specimen at a depth of 9.6nm, where the surface shows a well defined crystalline surface topography, although a complex contrast is still evident in the (112) planar region. In this instance several voids may be clearly distinguished close to the (112) planar region. The final micrograph in Fig. 5.4(t) reveals the emitter surface after a total of 70(011) planes have been removed by field evaporation corresponding to a depth of 16 nm. A well defined surface topography is evident. The grain boundary which was originally observed in the (001) planar region, now intersects the emitter surface through the (101) planar region. The different orientation of the grain boundary being due to the field evaporation removal of the surface layers of the irradiated specimen.

The results of the void sizing analysis of the 1keV helium irradiation to a dose of $1 \times 10^{16}$ ions.cm$^{-2}$ are shown in Fig. 5.5. The void size distribution reveal that the voids have sizes ranging from 0.2 to 1.9nm with a mean void size of 0.8nm (standard deviation of 0.43nm). A total number of 37 voids were found in a total number
Fig. 5.3
void size and depth distributions after 1 keV helium irradiation in tungsten to a total dose of $1 \times 10^{16}$ ions cm$^{-2}$.

He-W
1 keV
$1 \times 10^{16}$ ions cm$^{-2}$
of 70(011) planes removed by controlled field evaporation. The void depth distribution shows that a large number of voids were formed close to the surface of the irradiated specimen consisting of small vacancy clusters. However, a second group of voids is also present, which forms a series of voids about 5nm below the irradiated surface. Several voids were also distinguished at depths of up to 13nm beneath the surface.

5.5.2.2 2keV helium irradiation

The maximum transferred energy for a collision event in a tungsten target for 1 keV helium ions is approximately 83 eV (assuming a 'head-on' inelastic collision). The transferred energy increases with incident ion energy to approximately 166 and 250 eV for 2 and 3 keV helium ion irradiation respectively. Fig. 5.6 depicts a small evaporation sequence through the surface layers of a specimen irradiated with 2 keV helium ions to a total dose of $5 \times 10^{15}$ ions.cm$^{-2}$. Fig. 5.6(a) shows the irradiated surface of the previously characterised emitter. The channel plate dark spot may be clearly distinguished in micrograph by the continuous non-imaging area. Extensive surface damage may be distinguished in the form of numerous isolated vacancies and vacancy clusters. Fig. 5.6(b) reveals the surface after 2(011) planes have been removed (0.4nm), where large numbers of small voids (< 0.5nm) are present. Considerable 'bright spot' contrast may also be distinguished indicating that interstitials are present on the emitter close to the irradiated surface. A slight reduction in the extent of the damage present in the emitter surface is shown in Fig. 5.6(c) after the removal of 4(011) planes to a depth
Fig. 5.6 The surface damage in 2keV helium irradiated tungsten after a dose of $5 \times 10^{15}$ ions.cm$^{-2}$. The contrast due to small voids are indicated by the circles.

a) The irradiated surface
b) The damage at a depth of 2(011) layers (0.4nm).
c) The damage at a depth of 4(011) layers (0.9nm).
d) The damage at a depth of 6(011) layers (1.3nm).
e) The damage at a depth of 9(011) layers (2.0nm).
f) The damage at a depth of 11(011) layers (2.5nm).
of 0.9nm. Several voids may be distinguished close to the (010) and (121) planar regions. The following micrograph in Fig. 5.6(d) shows the surface after the removal of 6(011) planes to a depth of 1.3nm below the irradiated surface. The irregularly shaped void close to the (010) planar region is still evident and several other smaller voids are present. A small number of interstitial bright spots are also evident. Fig. 5.6(e) shows the surface of the irradiated specimen after 9(011) planes (2nm) have been removed from the specimen. A slight reduction in the number of bright spots is evident although isolated vacancies and vacancy clusters are still apparent. The final micrograph in this sequence is shown in Fig. 5.6(f) where a total of 11(011) planes have been removed corresponding to a depth of 2.5nm. Several small voids and vacancies are evident and an irregularly shaped void close to the (010) planar region is also present.

The void size and depth distributions for a 2keV helium irradiated specimen to a dose of $5 \times 10^{15}$ ions.cm$^{-2}$ is depicted in Fig. 5.7. The analysis was conducted over approximately 70(011) planes removed from the specimen in which a total of 41 voids were recorded. The void size distribution shows that voids were formed in the size range 0.2 to 0.8nm with a mean void size of 0.36nm (standard deviation of 0.2nm). The void depth distribution shows that a considerable number of voids exist close to the surface of the irradiated specimen. The voids are formed in small numbers extending up to 13nm beneath the original irradiated surface. The total range and mean projected range of 2keV helium ions in tungsten has been previously determined from LSS theory to be 14 and 5.8nm respectively. The void
He–W
2 keV
5 × 10^{15} ions cm^{-2}

Fig. 5.7: Void size and depth distributions after 2 keV helium irradiation in tungsten to a total dose of 5 × 10^{15} ions cm^{-2}.
depth distribution however, shows that voids are produced at depths greater than the mean projected range nearly extend to the total range of 2keV helium implanted in tungsten.

Helium irradiation at this energy was also conducted at a dose of $1 \times 10^{16}$ ions.cm$^{-2}$, where considerable surface damage was observed after the removal of 1(011) plane. The damage consisted of large numbers of bright spot contrast with small voids in the form of vacancy clusters interspersed over the emitter surface. Spiral contrast was observed on the (011) plane of the emitter surface at a depth of 2.6nm which persisted for several (011) planes. Several planar defects were observed emerging from the (112) and (121) planar regions which persisted for the removal of 7(011) planes (1.5nm). Numerous voids were distinguished in the emitter surface during the field evaporation in which approximately 70(011) planes were removed. Planar defects were observed in several planar regions due to the emergence of dislocation loops on the emitter surface to a depth of 68(011) planes (15.2nm) beneath the irradiated surface.

The analysis of the controlled field evaporation sequence for a 2 keV helium irradiated specimen to a dose of $1 \times 10^{16}$ ions.cm$^{-2}$ is shown in Fig. 5.8. In all, a total of 74 voids were recorded in approximately 70(011) planes removed during the depth profile analysis. The void size distribution shows that the voids range in size from 0.2 to 1.6nm with a mean void size of 0.6nm (standard deviation of 0.3nm). Large numbers of voids occur at low sizes which exist close to the surface. This is illustrated in the void depth distribution where numerous small voids are present close to the surface. However, in-
In tungsten to a total dose of 1 x 10^16 ions/cm^2.

**Fig. 5.8** Voids size and depth distributions after 2 keV helium irradiation.

No. of Voids

<table>
<thead>
<tr>
<th>No. of Voids</th>
<th>0</th>
<th>2</th>
<th>4</th>
<th>6</th>
<th>8</th>
<th>10</th>
<th>12</th>
</tr>
</thead>
<tbody>
<tr>
<td>Depth below surface (nm)</td>
<td>0</td>
<td>2</td>
<td>4</td>
<td>6</td>
<td>8</td>
<td>10</td>
<td>12</td>
</tr>
</tbody>
</table>

He-W

2keV

1 x 10^16 ions/cm^2
creasing numbers of voids exist close to the mean projected range of the 2keV implanted helium (about 6nm) below the irradiated surface. The voids extend to the total depth of the helium ions in the tungsten specimen.

5.5.2.3 3keV helium irradiation

The results of the 3keV helium irradiation of the tungsten specimen are shown in a sequence of micrographs in Fig. 5.9 for a total ion dose of $1 \times 10^{15}$ ions.cm$^{-2}$. The surface of the characterised specimen following irradiation is shown in Fig. 5.9(a) after the removal of 1(011) plane at an imaging voltage of 6.9kV. Numerous 'bright spot' contrast is evident with interspersed vacancy clusters on the emitter surface. The second micrograph in Fig. 5.9(b) illustrates the irradiated specimen after 3(011) planes have been removed (0.7nm). Small numbers of bright spots are present on the surface, predominantly due to metastable zone decoration atoms. Isolated vacancies and vacancy clusters forming small voids may be distinguished on the emitter surface. Contrast due to the emergence of a dislocation may be observed on the (Z11) planar region which persisted for several (011) planes removed, about 0.5nm. Fig. 5.9(c) shows the emitter surface after the removal of 7(011) planes corresponding to a depth of 1.5nm beneath the irradiated surface. Extensive surface damage is present in the form of bright spot contrast with numerous small irregularly shaped voids. In contrast the micrograph shown in Fig. 5.9(d) reveals minimal damage manifest on the emitter surface after a total of 25(011) planes have been removed. At this depth (5.6nm) isolated vacancies are evident with a limited number of small voids. Fig. 5.9(e) shows
Fig. 5.9  A series of micrographs of tungsten irradiated with 3keV helium ions to a dose of $1 \times 10^{15}$ ions.cm$^{-2}$. The contrast due to voids and dislocations are illustrated by circles and arrows respectively.

a) The irradiated surface after 1(011) layer removed.
b) The damage at a depth of 3(011) layers (0.7nm).
c) The damage at a depth of 7(011) layers (1.5nm).
d) The damage at a depth of 25(011) layers (5.6nm).
e) The damage at a depth of 31(011) layers (6.9nm).
f) The damage at a depth of 53(011) layers (11.8nm).
the emitter surface after 31(011) planes have been removed which corresponds to a depth of 6.9nm below the irradiated surface. Minimal surface damage is evident on the emitter surface, apart from 'bright spot' contrast which is present in the (011) planar rings in non-zone decoration atom positions. A similar observation is illustrated in Fig. 5.9(f) after the removal of 53(011) planes (11.8nm) from the original irradiated specimen surface. Analysis of the void size and depth distribution for the field evaporation sequence described above, showed that the voids ranged in size from 0.2 to 1.2nm with a mean void size of 0.44nm (standard deviation of 0.29nm). A total number of 21 voids were determined during the analysis, in which 31 (011) planes were removed corresponding to a depth of 6.9nm. However, due to the angular corrections performed incorporated in the void sizing technique, the voids only persisted to a depth of about 5nm below the irradiated surface. Further field evaporation did not reveal the presence of further voids, although isolated vacancy clusters may exist, the emitter surface remained in a well characterised surface topography.

In a similar experiment 3 keV helium irradiation was performed to a total dose of $5 \times 10^{15}$ ions.cm$^{-2}$. Extensive surface damage was observed on the irradiated specimen after 1(011) plane was removed by field evaporation. Small voids ($< 0.5$nm) were distinguished on the emitter surface together with bright spot interstitial contrast. Several planar defects were evident on (112) type planar regions indicating the emergence of dislocations on the emitter surface. The planar defects persisted for several (011) planes removed ($\sim 1$mm), close to the irradiated surface of the specimen ($< 3$mm). One dislocation emerging on
the (011) plane persisted for a total of 11(011) planes removed, the dislocation extended from the irradiated surface. An in-depth void analysis of the irradiated specimen is depicted in Fig. 5.10. The void size distribution shows that the voids range in size from 0.2 to 1.8 nm with a mean void size of 0.6 nm (standard deviation of 0.35 nm). A total number of 36 voids were recorded in 60(011) planes removed under a controlled field evaporation sequence. The void depth distribution illustrates the formation of a large number of voids present in the irradiated specimen existing close to the irradiated surface. The voids extend in small numbers to a total depth of nearly 13 nm below the irradiated surface.

The effect of an increasing higher dose of $1 \times 10^{16}$ ions/cm$^2$ at 3 keV helium irradiation is shown in Fig. 5.11, where a detailed field evaporation sequence of the irradiated specimen is illustrated. Fig. 5.11(a) reveals the irradiated surface after 1(011) plane has been removed. Bright spot interstitial contrast can be distinguished with numerous interspersed surface voids and vacancies occurring over the imaging region of the specimen. The second micrograph in Fig. 5.11(b) shows the surface after 3(011) planes have been removed (0.7 nm). A spiral dislocation on the (011) plane reveals the emergence of a dislocation on the emitter surface. A further planar defect is also present on the (121) planar region. The interstitial contrast is reduced and a grain boundary can be clearly distinguished passing through the (112) planar region. Fig. 5.11(c) shows the surface of the specimen after 6(011) planes have been removed (1.34 nm), where several irregularly shaped voids may be discerned. Spiral contrast is also evident in the (011) planar rings and the (112) and (121) planar regions.
He-W
3 keV
$5 \times 10^{15}$ ions.cm$^{-2}$

Fig. 5.10
Void size and depth distributions after 3 keV helium irradiation to a total of $5 \times 10^{15}$ ions.cm$^{-2}$. 

VOID SIZE (nm)

DEPTH BELOW SURFACE (nm)

No. of voids

No. of voids
Fig. 5.11 The effect of 3keV helium irradiation in tungsten after a dose of $1 \times 10^{16}$ ions.cm$^{-2}$. The presence of void and dislocation contrast are indicated by circles and arrows in the field evaporation sequence. The series of micrographs illustrates the specimen after:

a) The irradiated surface
b) After 3(011) layers removed (0.7nm).
c) 6(011) layers removed (1.3nm).
d) 9(011) layers removed (2nm).
e) 13(011) layers removed (2.9nm).
f) 17(011) layers removed (3.8nm).
g) 38(011) layers removed (8.5nm).
h) 40(011) layers removed (9nm).
i) 46(011) layers removed (10.3nm).
j) 54(011) layers removed (12nm).
k) 69(011) layers removed (15.4nm).
l) 81(011) layers removed (18.1nm).
Fig. 5.11(d) reveals the irradiated specimen after 9(011) planes have been removed (2nm). Dislocations emerging from the (011) and (112) planes are still evident together with numerous small voids. Continued field evaporation of a total of 13(011) planes (2.9nm) again reveals the presence of the spiral contrast on the (011) plane in Fig. 5.11(e). A small vacancy cluster on the (121) plane ring is evident with a 'bright spot' interstitial contrast present on the same atomic row of the (121) planes. Fig. 5.11(f) shows the irradiated specimen after 17(011) planes have been removed (3.8nm) where the spiral contrast is still present on the (011) planar region. The small vacancy cluster on the (121) plane is present on the emitter surface and a dislocation may be distinguished on the (112) plane. In Fig. 5.11(g) several interstitial 'bright spot' contrast can be observed close to small voids in different planar regions (i.e. (112) type planes). The surface occurs after 38(011) planes have been removed (8.5nm) from the irradiated surface. The spiral contrast on the (011) plane is no longer evident, however, the dislocation contrast is still present, manifest as an included half planar ring. This is illustrated in Fig. 5.11(h) where a half planar ring now forms part of the spiral contrast due to the dislocation intersecting the emitter surface after 40(011) planes (9nm) have been removed. Fig. 5.11(i) shows the surface after 46(011) planes have been removed (10.3nm), where the spiral contrast typical of a perfect dislocation is present on the emitter surface.

Several small voids in the form of vacancy clusters may be distinguished and a complex planar defect intersects the emitter surface near the (121) planar region. Similar features are present in the next micrograph shown in Fig. 5.11(j) after 54(011) planes have been
removed (12.1nm). A relatively characterised surface is evident in Fig. 5.11(k) after 69(011) planes were removed, corresponding to a depth of 15.4nm below the irradiated surface. The spiral contrast is still present on the emitter surface together with small irregularly shaped voids and small numbers of interstitial contrast. The final micrograph of the sequence occurs at a depth of 18.1nm, after the removal of 81(011) planes from the irradiated surface. The grain boundary has shifted in orientation (due to the removal of the surface layers of the specimen) and now forms an ill defined irregular boundary between the two crystallites. However small voids in the form of vacancy clusters may be distinguished. The 'bright spot' interstitial contrast exists mainly as metastable zone decoration sites typical of unirradiated tungsten emitter surfaces.

The analysis of the voids formed during 3 keV helium irradiation to a dose of $1 \times 10^{16}$ ions cm$^{-2}$ is shown in Fig. 5.12, where the void size distribution ranges from 0.2 to 2.2nm with a mean void size of 0.73nm (standard deviation of 0.43nm). A total number of 51 voids were recorded in a field evaporation sequence of approximately 100(011) planes. The depth distribution of the voids in the irradiated specimen shows that numerous small voids are present close to the irradiated surface, however the voids extend from the irradiated surface to a depth of approximately 21nm. Thus voids are formed at greater depths than the mean projected range (ie. 8nm) and extend to the total depth of the implanted helium ions which is approximately 18nm. Several dislocations were distinguished during the analysis, where those present on the (112) type plane persisted for up to 1.1nm at varying depths during the field evaporation of the irradiated specimen. The spiral contrast on the
He-W
3keV
$1 \times 10^{16}$ ions.cm$^{-2}$

Fig. 5.12
Void size and depth distributions after 3 keV helium irradiation.

In tungsten to a total dose of $1 \times 10^{16}$ ions.cm$^{-2}$. 
(011) planar region however, persisted for approximately 70 (011) planes removed and extended from close to the irradiated surface (< 0.5nm) to a depth of 15.6nm below the surface.

5.5.3 Helium irradiation at high doses.

5.5.3.1 1keV helium irradiation

In this section a series of helium irradiation of tungsten specimens were performed with ion energies in the range 1 to 3keV at high ion doses (> $1 \times 10^{16}$ ions.cm$^{-2}$). A detailed analysis of a field evaporation sequence of micrographs of 1keV helium irradiation to a dose of $5 \times 10^{16}$ ions.cm$^{-2}$ is illustrated in Fig. 5.13. The characterised surface prior to helium irradiation is shown in Fig. 5.13(a), the second micrograph in Fig. 5.13(b) shows the irradiated emitter surface at an imaging voltage of 4.4kV. Extensive surface damage is evident on the emitter surface after 1(011) plane has been removed, with several large voids present on the emitter surface. Smaller voids are evident interspersed amongst the larger voids and 'bright spot' interstitial contrast are also present. The severe surface distortion due to the large voids intersecting the emitter surface may still be distinguished in Fig. 5.13(c) after 3(011) planes have been removed (0.7nm). Several voids may be discerned in different planar regions on the emitter surface together with 'bright spot' contrast which appears at the periphery of the voids.

Fig. 5.13(d) shows the surface at a depth of 9(011) planes (2nm) where some of the surface voids have been removed during the field
Fig. 5.13 The damage following 1keV helium irradiation in tungsten after a total dose of $5 \times 10^{16}$ ions.cm$^{-2}$. The series of micrographs illustrates the damaged specimen, where void and dislocation contrast is illustrated by the circles and arrows respectively.

a) The characterised surface, prior to irradiation
b) the irradiated surface after 1(011) layer removed
c) after 3(011) layers (0.7nm)
d) 9(011) layers (2nm)
e) 13(011) layers (2.9nm)
f) 17(011) layers (3.8nm)
g) 19(011) layers (4.2nm)
h) 24(011) layers (5.4nm)
i) 30(011) layers (6.7nm)
j) 34(011) layers (7.6nm)
k) 40(011) layers (8.9nm)
l) 49(011) layers (11nm)
m) 64(011) layers (14.3nm)
n) 70(011) layers (15.7nm).
evaporation sequence. The central (011) plane has nearly returned to a characterised surface after the considerable distortion on the (011) planar rings due to the presence of several voids in the (011) planar region. The distortion of the planar rings close to the voids may be attributed to the complex contrast pattern produced by the intersection of the void and the emitter surface under imaging stresses. Fig. 5.13(e) reveals the irradiated specimen after 13(011) planes have been removed corresponding to a depth of 2.9nm. Extensive surface damage is still present on the emitter topography, where several small voids may be observed interspersed with the larger surface voids. Severe planar distortion is still evident particularly in the (112) plane, which is due to a closely allied void.

Fig. 5.13(f) depicts the surface after 17(011) planes have been removed (3.8nm) from the irradiated surface. A dislocation may be discerned emerging on the (112) plane and the (011) planar rings are distorted by voids. The distortion in the emitter surface is also apparent in the (112) planar region where small voids and interstitial contrast is evident. Fig. 5.13(g) shows the surface after 19(011) planes (4.2nm) have been removed. Several large voids are manifest in the emitter surface, one of which exhibits a 'star shaped' image contrast which has previously been attributed to an ion optical effect caused by the enhanced field at the sharp edges of the void intersecting the emitter surface (Godfrey et al (1976)). A complex planar defect may be distinguished on the (112) plane, due to the intersection of a dislocation and the emitter surface. Several small voids are also present in the same planar region where considerable lattice damage is present. The (112) planar region is severely
distorted, however, field evaporation of a total of 24(011) planes (5.4) reveals the planar defect to be extensive with voids occurring near to the defect as illustrated in Fig. 5.13(h). Numerous voids may be distinguished over the emitter surface, some which exhibit 'star shaped' image contrast. The (011) and (101) planar regions both exhibit planar defects, a spiral contrast on the (011) plane and a split plane defect on the (101) corresponding to the emergence of a dislocation with the emitter surface.

A complex dislocation structure is evident in the central (011) planar rings illustrated in Fig. 5.13(i) after 30(011) planes have been removed (6.7nm). The defect structure in the (101) planar rings is also evident in this micrograph together with a dislocation emerging on the (112) planar region. Many voids are present on the emitter surface which are smaller (less persistent) than the original initial elongated voids present at the surface. The complex dislocation present in the (011) planar rings is still manifest in Fig. 5.13(j) and several planar defects are also present on the emitter surface after a total of 34(011) planes (7.6nm) have been removed from the irradiated surface. Fig. 5.13(k) reveals the surface after 40(011) planes (8.9nm) have been removed, where several planar defects and voids are present on the emitter surface. Extensive surface damage is manifest in the emitter, due to emergent dislocations and the intersection of voids with the emitter surface forming a complex image contrast. Planar defects may be distinguished in the (001), (112) and (110) planar regions, and several voids are particularly evident in the (111) planar region. Fig. 5.13(l) shows the surface after 49(011) planes (11nm) have been removed from the irradiated surface. Spiral contrast, typical of a
perfect dislocation is evident in the central (011) plane, a further planar defect is also present in the (121) plane. Interstitial image contrast is present in the (001) planar region, a feature also present in the previous micrograph. No planar defect is evident in the (101) plane although spiral contrast may be distinguished in the (010) plane. The spiral contrast is still present in the (011) plane in Fig. 5.13(m) after 64(011) planes (14.3nm) have been removed where planar defects are manifest in the (101), (121) and (112) planes.

Several voids may also be distinguished especially near the (001) planar region of the emitter surface. The final micrograph shown from the field evaporation sequence is illustrated in Fig. 5.13(n) after a total of 70(011) planes (15.7nm) have been removed from the irradiated surface. Several planar defects and small voids are evident although the dislocation contrast previously present in the (011) planar regions is no longer evident. The field evaporation sequence was continued until 86(011) planes had been removed from the irradiated emitter surface corresponding to a depth of 19.2nm below the original surface. At this depth minimal surface damage was discerned and a relatively well characterised emitter topography was observed.

The void size and depth distributions have been determined for the previously described irradiated specimen, the results of which are depicted in Fig. 5.14. The analysis was conducted over a total number of 86(011) planes removed (19.2nm) under controlled field evaporation conditions in which 74 voids were recorded. The void sizes range from 0.2 to 5.2nm with an average void size of 1.4nm. The void size distribution is similar to that observed in low dose irradiation in
Fig. 5.14 Void size and depth distributions after 1keV helium irradiation of tungsten to a total dose of $5 \times 10^{16}$ ions.cm$^{-2}$. 
that large numbers of small voids are produced. The void depth
distribution reveals the presence of larger voids occurring close
to the irradiated surface. The large numbers of voids, exceeding
even the numbers of voids produced at or close to the surface are
present at greater depths beneath the irradiated surface. One such
high concentration of voids occurs at approximately 4nm below the
irradiated surface. Less numerous voids are also present at greater
depths, extending to a total depth of approximately 19nm. The mean
projected range and total range of 1keV helium implanted in tungsten
has been previously found from LSS theory to be 3.7nm and 9.2nm res-
spectively. The void depth distribution shows that voids are produced
at depths much greater than (nearly a factor of 2) that predicted by
theory. Furthermore numerous dislocations have been observed to ex-
tend from the irradiated surface to sub-surface of approximately 70(011)
planes corresponding to a depth of 16nm.

5.5.3.2 2keV helium irradiation

An irradiation experiment at the higher dose of $5 \times 10^{16}$
ions.cm$^{-2}$ has also been performed at an helium ion energy of 2keV on a
tungsten emitter. The results shown in a sequence of micrographs in
Fig. 5.15. At this irradiation energy the maximum transferred energy is
186eV. The effects of the higher dose on the tungsten emitter are
remarkable compared with the low dose irradiation. The previously
characterised and subsequently irradiated emitter surface is shown in
Fig. 5.15(a) at an imaging voltage of 5.1kV after 1(011) plane has been
removed. The specimen surface is severely damaged and extensive planar
defect occurs in the (011) plane. A large void is present on the
Fig. 5.15 The damage following 2keV helium irradiation of tungsten to a dose of $5 \times 10^{16}$ ions.cm$^{-2}$. Void and dislocation contrast are illustrated by the circles and arrows.

a) Irradiation surface after 1(011) layer removed (0.2nm)  
b) The damage after 4(011) layers removed (0.9nm)  
c) The damage after 8(011) layers removed (1.8nm)  
d) The damage after 14(011) layers removed (3.1nm)  
e) The damage after 29(011) layers removed (6.5nm)  
f) The damage after 80(011) layers removed (17.9nm)
emitter surface close to the (011) plane, the void having a size of approximately 3.5nm. A second large void (~3nm) may also be distinguished on the emitter surface. Considerable emitter surface distortion is still present in Fig. 5.15(b) after the removal of 4(011) planes (0.9nm), where the two large voids are clearly evident. The distortion of the (011) planar rings at the edge of the voids are particularly evident in the two micrographs, due to the complex interaction of the voids with the emitter surface under imaging conditions.

The specimen unfortunately suffered a partial collapse of the emitter surface following a slow field evaporation sequence, where a large number of (011) planes were suddenly removed under normal imaging conditions. (imaging voltage 5.9kV in a He/Ne gas mixture at a gas pressure of 5 x 10^{-5} Torr). The most likely cause is that of induced field stress which is unavoidably present during the imaging process. Thus stresses would be produced greater than the ultimate strength of the irradiated specimen, due to the presence of large voids close to the emitter surface. The specimen cannot therefore be used for quantitative depth analysis but serves to illustrate the effects of high irradiation doses at 2keV helium ion energy.

Fig. 5.15(c) reveals the surface after 8(011) planes were removed from the collapsed emitter surface. The two large voids are still evident indicating that the voids may have an elongated structure due to the number of (011) planes which may have been removed, which is implied by the increased radius of the emitter surface. A small partial plane of atoms present in the (011) planar rings is evident, close to
the void in the (011) planar rings. The (112) plane also appears to have a planar defect present with associated bright spot contrast, a feature which was manifest in the previous micrograph. Fig. 5.15(d) shows the surface after 14(011) planes have been removed where some surface damage contrast is present on the (011) plane. The (112) planar defect is clearly visible and several bright spot contrast may be distinguished on the emitter surface. The large void initially present on the irradiated surface has been nearly completely removed by continued field evaporation, and the periphery of the void has bright spot contrast. Smaller voids are also present on the emitter surface together with some vacancy contrast. Several dislocations may be observed emerging from the (011), (112) and (112) planar regions on the emitter surface in Fig. 5.15(e) after 29(011) planes have been removed from the collapsed emitter surface. However the emitter surface has regained a more characterised surface topography compared with the previous surface damage. The final micrograph in the sequence shows the emitter surface after 80(011) planes have been removed from the collapsed emitter surface in Fig. 5.15(f). In this micrograph a relatively well characterised emitter topography is evident with minimal damage occurring on the emitter surface.

This sequence of micrographs recorded at higher doses (viz. $5 \times 10^{16}$ ions.cm$^{-2}$) at 2keV helium ion energy illustrates the difficulties involved with high dose irradiation especially at higher transferred ion/atom collision energies, where considerable radiation damage is produced in the specimen and which may produce catastrophic specimen failure. This analysis of each type of defect is not only difficult but complex due to the higher defect and void densities.
However, limited analysis of the voids present in the irradiated specimen reveals that voids are produced in sizes ranging from small vacancies and their clusters to large surface voids. The surface voids persist for a large number of removed surface planes and thus appear as elongated void structures extending into the sub-surface of the specimen. Furthermore the higher dose irradiations produce numerous and extensive sub-surface dislocations which may persist for depths approaching the total range of the implanted helium ions in the specimen.

Several irradiation experiments using high helium doses (> $1 \times 10^{17}$ ions.cm$^{-2}$) were attempted at ion energies less than 3keV, however, catastrophic damage occurred at the emitter surface. The high dose irradiations resulted in specimen tip failure during field evaporation of the irradiated specimen surface. An example is shown in Fig. 5.16 for a tungsten emitter irradiated with 2keV helium ions to a total dose of $1 \times 10^{17}$ ions.cm$^{-2}$. Extensive surface damage is evident in Fig. 5.16(a) where the irradiated surface is considerably distorted at an imaging voltage of 6.1kV. A large elongated non-imaging region is evident, which passes through the (011) planar rings. Bright spot contrast is present on the emitter surface with several small vacancy clusters. The careful field evaporation removal of 4(011) planes (0.9nm) reveals the dark line feature in better detail, shown in Fig. 5.16(b). Considerable planar mismatch is evident in the (011) planar rings, which indicates that the defect is planar in nature. Extensive surface damage and severe distortion of the emitter topography is apparent and several voids are evident on the surface particularly in the vicinity of the (031) planar region. Further
Fig. 5.16  The damage tungsten specimen following 2keV helium irradiation to a dose of $1 \times 10^{17}$ ions cm$^{-2}$.

a) The irradiated surface of the specimen (note the extensive surface damage).

b) The damage after 4(011) layers removed.
field evaporation resulted in catastrophic tip failure, which occurred
due to the large imaging stresses present on the unstable emitter
topography. The instability resulting from the presence of high
defect concentrations in the form of dislocation and voids close
to the irradiated emitter surface. Similar topographical features
were observed in a tungsten specimen irradiated with 1keV helium ions
to a total dose of $1 \times 10^{17}$ ions$\cdot$cm$^{-2}$. Considerable surface damage
and planar distortion was observed during the field evaporation removal
of 10(011) planes at an imaging voltage of 8.0kV. Further field
evaporation resulted in tip failure.

5.5.3.3 3keV helium irradiation

The effect of 3keV helium irradiation on a tungsten specimen
was conducted at a dose of $1 \times 10^{17}$ ions$\cdot$cm$^{-2}$ where extensive surface
damage was observed on the irradiated surface. Dislocations were
manifest on the characterised surface after irradiation (particularly
the (011) planar region) with the presence of large voids ($\sim$ 2 to 3nm)
interspersed with numerous smaller voids with sizes ranging from 0.5
to 1.0nm. The dislocation contrast on the (011) plane persisted for
approximately 50(011) planes removed by field evaporation indicating
an extensive surface defect extending to $\sim$ 10nm beneath the irradiated
surface. Further dislocation contrast could be distinguished through-
out the field evaporated sequence up to 110(011) planes removed corres-
ponding to a total depth of 25mm below the irradiated surface. The dislo-
cation contrast was manifest on the (112) type, (011) and (001) planar
regions. Severe image distortion was observed with numerous inter-
stitial 'bright spot' contrast present on the irradiated surface and
sub-surface regions.

The void size and depth distributions are shown in Fig. 5.17 for 3keV helium irradiation at a dose of $1 \times 10^{17}$ ions.cm$^{-2}$. The void size distribution reveals that large numbers of small voids are produced in the size range 0.3 to 0.7nm. However the voids range in size from 0.2 to 3.2 nm, with an average void size of 0.7nm. A total number of 167 voids were recorded during the field evaporation sequence in which 135(011) planes (30nm) were removed from the irradiated surface. The void depth distribution shows that the voids extend up to 20nm below the irradiated surface. Numerous small voids are interspersed amongst the larger voids present on the irradiated surface, although a larger number of voids occur at a depth of approximately 4.0nm beneath irradiated surface. Further numbers of voids are formed at depths of up to 13nm beneath the surface, after which only smaller numbers of voids were found and where minimal void damage was observed at depths greater than 20nm below the irradiated surface. The mean projected range and the total range of 3keV helium in tungsten has been determined from LSS theory to be 8.0 and 18.0nm respectively. Thus it appears that the voids are formed along the total path length of the implanted helium ions with higher void concentrations occurring near the mean projected range of the implanted helium ions.

The result of 3keV helium irradiation to a total dose of $5 \times 10^{17}$ ions.cm$^{-2}$ is illustrated in Fig. 5.18(a), which shows the irradiated specimen surface at an imaging voltage of 6.8kV. Considerable surface damage is evident, where the (011) plane is only partially resolved. Several regions of the emitter surface produce an intense
Fig. 5.17 Void size and depth distributions after 3 keV helium irradiation of tungsten to a total dose of $1 \times 10^{17}$ ions.cm$^{-2}$. 
Fig. 5.18 The result of 3keV helium irradiation of tungsten after a dose of $5 \times 10^{17}$ ions.cm$^{-2}$.

a) The irradiated surface

b) The partially collapsed irradiated emitter surface, where severe damage is evident.
image contrast due to protruding surface atoms and surface field enhancement; the large non-imaging areas of the emitter surface corresponding to large surface voids. The irradiated emitter surface became unstable during the field evaporation of a single (011) plane, resulting in a partial emitter failure. This implies that extensive surface and sub-surface damage may have been present, thus producing an unstable emitter structure under field evaporation conditions. The partially collapsed emitter surface is shown in Fig. 5.18(b) where extensive surface damage is evident with a large planar defect manifest on the (011) plane. The further use of field evaporation resulted in a catastrophic failure of the emitter after the removal of approximately half of a (011) plane.

Several dark streaks or line contrast is particularly evident in the micrograph, where the streaks pass through planar regions of the emitter surface. The occurrence of large dark streak features in field-ion micrographs is rare, although Müller (1960) has observed smaller dark line features which have been attributed to slip. Page and Ralph (1970) have analysed slip in an irradiation specimen, and found that each side of the slip line would have a bright/dark contrast due to the shielding of the unslipped portion of the crystal to the electric field. A similar feature is also apparent in Fig. 5.20. The extensively damaged surface prevents a rigorous analysis of the slip planes which exist for tungsten (e.g., (011) <111>, (112) <111> and (123) <111>), however, the effect of blister formation at the higher doses, does not preclude the possibility of a collapsed unstable surface due to the presence of a large void or voids close to the emitter surface. The effect of the high electric field stress could therefore produce de-
formation of the emitter topography in which slipped planes may occur.

5.6 Low-energy helium irradiation

The study of low energy helium irradiation at energies less than 1keV was performed to investigate the effects of the damage where the transferred energy is comparable to or less than the threshold displacement energy, \( E_d \), necessary to produce a Frenkel pair in the tungsten specimen. Helium ion doses were chosen \( (> 5 \times 10^{16} \text{ ions cm}^{-2}) \) so that any damage would be directly observed using the FIM technique. Previous work at low energies and doses using the cathode sputtering irradiation method has shown that minimal surface and sub-surface damage was observed at doses below those chosen for the irradiation carried out in this study.

Low-energy helium (< 1keV) irradiation has been previously used as a diagnostic probe in several techniques such as thermal desorption (Kornelsen (1972)) and ion scattering spectroscopy (Honig et al (1973)) where helium atoms provide information regarding the defect damage substructure and surface topography respectively. This is especially true in ISS, where analysis of the energy spectrum of the reflected helium atoms allows the identification of surface atoms. Thus the observation of low-energy helium irradiation using field-ion microscopy can provide further information concerning irradiation damage produced at higher doses \( (> 5 \times 10^{16} \text{ ions cm}^{-2}) \) which may have deleterious effects in the use of these techniques. Furthermore, the effects of large doses of low-energy helium irradiation may produce damage due to the presence of a high concentration of implanted gas atoms situated close to the surface of the
irradiated specimen, where the transferred energy is insufficient to produce Frenkel defects. Assuming a head-on collision, helium atoms in tungsten may only transfer a maximum of 8.3% of the total ion energy, which for 500 and 200 eV helium ions corresponds to 41.5 and 16.6 eV respectively.

The result of low-energy helium irradiation at an ion energy of 500 eV on a previously characterised emitter are shown in a sequence of micrographs in Fig. 5.19, after a total dose of $5 \times 10^{16}$ ions.cm$^{-2}$. The surface shows some irradiation damage in the form of small vacancy clusters together with some bright spot image contrast in Fig. 5.19(a). The surface after the removal of 3(011) planes (0.7nm) is depicted in Fig. 5.19(b), where several interstitial atoms are present on (101) and (110) planes. A small vacancy cluster exists near the (112) plane, which is still present in Fig. 5.19(c) after the removal of 8(011) planes (1.8nm).

A planar defect is evident in the (∑21) plane, a feature which is persistent in Fig. 5.19(d) after 12(011) planes have been removed, a depth of 2.8nm below the irradiated surface. The (∑21) planar region has an apparent complex arrangement of surface atoms corresponding to the intersection of a planar defect (such as a dislocation) with the emitter surface. The defect persisted for a total of 8(011) planes corresponding to a layer of approximately 1.5nm in thickness. Several voids were also observed during the field evaporation sequence with sizes ~ 0.5nm within the removed layer.
Fig. 5.19  The result of low-energy (500eV) helium irradiation in tungsten after a dose of $5 \times 10^{16}$ ions.cm$^{-2}$. Void and dislocation contrast are indicated by the circles and arrows.

a)  The irradiated surface.
b)  The damage at a depth of 3(011) layers (0.7nm)
c)  The damage at a depth of 8(011) layers (1.8nm)
d)  The damage at a depth of 12(011) layers (2.7nm)
e)  The damage at a depth of 28(011) layers (4.4nm)
f)  The damage at a depth of 31(011) layers (6.2nm)
Fig. 5.19(e) shows the surface after the removal of 28(011) planes (4.4nm), where several vacancy clusters are evident together with some interstitial atoms contrast in the vicinity of the (110) type planar regions. A dark streak is also evident close to the (131) planar region. Fig. 5.19(f) depicts the surface after a total of 31(011) planes (6.2nm) have been removed, at this depth below the irradiated surface, minimal damage is present on the emitter surface which reveals an almost characterised topography.

The void size and depth distribution for this sequence of micrographs is illustrated in Fig. 5.20 for the 500eV helium ion irradiation. The void sizes range from 0.2 to 1.4nm with a mean void size of 0.45nm (standard deviation of 0.37nm). The analysis was conducted over a total of 36(011) planes (8.1nm) removed by evaporation in which 29 voids were recorded. The void depth distribution reveals a relatively large number of voids located close to the irradiated surface. However, the voids extend to a total depth of 5.4nm.

The effect of increasing the helium ion dose at this energy (500eV) is illustrated in Fig. 5.21 for a total dose of $1 \times 10^{17}$ ions.cm$^{-2}$. Fig. 5.21(a) shows the irradiated surface (at an imaging voltage of 5.3kV) after the first (011) plane was removed by careful field evaporation. Extensive surface damage is evident with severe surface distortion, where several large surface voids are present which extend to 6(011) planes to a depth of 1.3nm beneath the surface. Numerous smaller voids may be distinguished interspersed amongst the large voids, the small voids having sizes in the range 0.4 to 0.9nm.
Fig. 5.20 Void size and depth distributions after 0.5 keV helium irradiation of tungsten to a total dose of $5 \times 10^{16}$ ions.cm$^{-2}$. 
Fig. 5.21  Low-energy (500eV) helium irradiation of tungsten after a dose of $1 \times 10^{17}$ ions.cm$^{-2}$.

a) Irradiated surface after the removal of 1(011) layer
b) Stabilised emitter topography following partial specimen collapse
c) The damage after the further removal of 15(011) layers (3.4nm)
d) The damage after the removal of 37(011) layers (8.2nm).
Unfortunately the emitter surface became unstable at this point in the sequence with uncontrolled removal of an unknown number of (011) apex planes. Fig. 5.21(b) shows the emitter surface of the unstable emitter at an imaging voltage of 6.9kV. Further extensive damage is apparent, although this may arise from artefacts due to the unstable emitter surface collapsing to a stable surface configuration under the imaging field stresses. However, a large void is present, and a line of disordered atoms is evident in the (011) planar ring system. Several smaller voids are also apparent on the emitter surface.

Fig. 5.21(c) depicts the surface after the removal of a further 15(011) planes (3.4nm), where extensive surface damage is evident in the form of irregularly shaped voids (< 0.8nm). One particular void has an elongated structure and is located close to the (121) planar region. Dislocation contrast is also present on the (011) plane, the dislocation persisted for a total of 4(011) (0.9nm) planes removed from the emitter surface. Fig. 5.21(d) reveals the surface after a total of 37(011) planes have been removed from the re-stabilised emitter surface corresponding to a depth of 8.2nm. The emitter topography, however, has a well defined crystallinity with minimal surface damage, although interstitial contrast is evident and a planar defect is present on the (112) planar region.

This sequence of micrographs for the low energy (viz 500eV) helium irradiation of the tungsten specimens, although not complete, illustrates that at sufficiently high doses (~ 1 x 10^{17} ions.cm^{-2}) extensive surface and sub-surface damage occurs.
A further irradiation experiment was carried out with 200eV helium ions at a dose of $1 \times 10^{17}$ ions.cm$^{-2}$. The results are shown in Fig. 5.22, for a sequence of micrographs where the emitter surface after irradiation is depicted in Fig. 5.22(a). The surface showed considerable bright spot contrast prior to field evaporation due to the presence of surface contamination when imaged in a He-Ne gas mixture at 11.1kV. However, the removal of a single (011) plane, shown in Fig. 5.22(b), reveals further interstitial contrast and numerous small vacancy clusters. A planar defect is evident on the (031) plane. The voids present on the surface range in size from 0.2 to 1.0nm. The removal of a further (011) plane (0.44nm) illustrated in Fig. 5.22(c) shows the removal of the defect on the (031) plane indicating the presence of a surface defect. However, several interstitial atoms may be discerned on the (011) planar ring system and a planar defect is present on the (112) plane.

Fig. 5.22(d) reveals the surface after 4(011) planes have been removed corresponding to a depth of 0.9nm. Several voids are apparent and a dislocation is manifest on the (112) planar region. Voids are also present after $5\frac{1}{2}$(011) planes (1.2nm) have been removed shown in Fig. 5.22(c). A further planar defect is evident in the (121) planar region which is also manifest in Fig. 5.22(f) after the removal of 8(011) planes (1.8nm). Interstitial contrast can be distinguished on the (011) and (112) planar rings, particularly near the small voids occurring in the (112) planar region. The bright spot interstitial contrast is still evident although less numerous in Fig. 5.22 (g and h) after the removal of 12 and 16(011) planes corresponding to a depth of 2.7 and 3.3nm respectively.
Fig. 5.22  Irradiation with 200eV helium to a dose of $1 \times 10^{17}$ ions cm$^{-2}$ in tungsten. The presence of void and dislocation contrast are indicated by the circles and arrows in the field evaporation sequence. The sequence of micrographs correspond to:

a) The irradiated surface  
b) The damage after 1(011) layer removed (0.2nm)  
c) The damage after 2(011) layers removed (0.4nm)  
d) The damage after 4(011) layers removed (0.9nm)  
e) The damage after 5½(011) layers removed (1.2nm)  
f) The damage after 8(011) layers removed (1.8nm)  
g) The damage after 12(011) layers removed (2.7nm)  
h) The damage after 16(011) layers removed (3.3nm)  
i) The damage after 19(011) layers removed (4.2nm)  
j) The damage after 26(011) layers removed (5.8nm)  
k) The damage after 32(011) layers removed (7.1nm)
Several planar defects may be observed in the (112) and (121) planar regions shown in Fig. 5.22(i). Several small voids are also evident close to the (121) planar region. Fig. 5.22(j) shows the emitter surface after the removal of 26(011) planes at a depth of 5.8nm. The extensive planar defect on the (121) plane; a small void may be discerned close to the (010) plane. The planar defect on the (121) plane has been finally removed after 32(011) planes (7.1nm) have been field evaporated from the irradiated surface, depicted in Fig. 5.22(k), where a small void is apparent on the (121) plane. The final micrograph of the sequence, shown in Fig. 5.22(l), reveals the irradiated emitter after 40(011) planes have been removed corresponding to a depth of 9.0nm. The emitter has an apparently well characterised surface topography, however, a planar defect is present on the (121) plane. Apart from this defect the surface has a minimal amount of damage.

The application of the void sizing technique allows the analysis of the void size and depth distribution to be performed. The results are shown in Fig. 5.23 for the first 30(011) planes (6.7nm) removed by controlled field evaporation where a total number of 42 voids were detected. The void sizes were found to range in sizes from 0.2 to 1.6nm with a mean void size of 0.85nm (standard deviation of 0.78nm). The void depth distribution is also shown and consists of two groups of voids. The first group occurring close to the surface, the second is centred about a depth of 2.0nm. The voids appear to extend to about 3.8nm beneath the irradiated surface within the depth analysed.
He-W
200 eV
$1 \times 10^{17}$ ions.cm$^{-2}$

Fig. 5.23 Void size and depth distributions after 0.2 keV helium irradiation of tungsten to a total dose of $1 \times 10^{17}$ ions.cm$^{-2}$. 
The void depth distributions of the implanted atoms agrees favourably with a previous low energy helium irradiation study. Wagner and Seidman (1978), using the Field Ion Atom-Probe technique have reported mean ranges of $4.0 \pm 0.4$ and $5.6 \pm 0.6$nm respectively for $300\text{eV}$ and $475\text{eV}$ helium ions implanted in tungsten at low doses ($< 1 \times 10^{16} \text{ ions.cm}^{-2}$). This is in reasonable agreement with the present irradiation experiments in which voids have been found to extend to total depths of $3.8$ and $5.4$nm for $200$ and $500\text{eV}$ helium ions respectively.

5.7 Discussion

The study of low-energy helium irradiation of tungsten using the FIM technique has revealed new information regarding the extent and nature of the damage resulting in the surface and sub-surface region of the target. Generally the incident projectiles penetrate the surface and collide with the target atoms; either coming to rest within the lattice, or leaving the target with reduced energy. Thus the radiation damage produced by these energetic projectiles may be brought about by several different ways as previously described in section 4.2. However, the basic types of damage in the irradiated tungsten specimens due to helium irradiation at low-energy may be catagorised as:-

a) The incident projectiles produce Frenkel pairs by collisions with lattice atoms producing both vacancies and interstitials.

b) Some of the projectiles may become embedded in the target forming interstitial atoms, which may perturb the surrounding lattice atoms to form a complex defect.
c) The trapping of the implanted projectiles at damage sites, such as vacancies or impurities, leading to the formation of gas-filled voids or bubbles.

d) The size and extent of the bubbles beneath the irradiated surface will be dependent on the ion dose and projectile energy.

e) Surface atoms of the target may be removed by the sputtering process.

Generally the damage due to low energy (<3keV) helium irradiation in tungsten has been found to consist of small voids in the form of vacancies and their clusters. The shape of the voids of small voids in the form of vacancies and their clusters. The shape of the voids of small sizes is that of an irregular cluster, the void shape becoming symmetrical at larger void sizes; large voids (>2nm) appear as elongated structures, either in the planar orientation in which the void is formed, or more generally in the plane of field evaporation removal. Comparison of the FIM observations for the irradiations performed show that the voids are nucleated by displacement events created by projectile/lattice atom collisions.Interstitial contrast has been observed in all the irradiation experiments conducted, the number of interstitials appears to be dependent on the total ion dose and projectile energy. However, extensive complex defects, such as dislocations have been observed only under certain irradiation conditions in large numbers. Isolated planar defects have been observed in low dose irradiations in all the ion energies used, however the number and extent (in terms of defect persistence during field evaporation) of the planar defects increases with higher dosage, and to a smaller amount with ion energy. Complex planar
defects have been observed at relatively low doses at the higher ion energies, but are usually observed in greater numbers for a high dose and projectile energy combination. Therefore, the frequency of occurrence of the planar defect increases with larger doses and high projectile energy. This may be attributed to two effects, namely, at high doses the implanted concentration is sufficient to produce numerous vacancies and interstitials, the latter may agglomerate to form a complex defect. Secondly, at higher ion energies the transferred energy during a collision event is greater, thus producing greater numbers of vacancies and interstitials which may form complex defects.

The depth distribution of the voids reveals several interesting features which have been observed during the field evaporation sequences. The void depth distribution has two distinct clusters or groups of voids situated: a) close to the surface and b) within the subsurface of the irradiated specimen generally close to mean projected range of the voids. Taking the 1keV helium irradiation at a dose of $1 \times 10^{16}$ ions.cm$^{-2}$ as an example; the two groupings of the voids may be clearly distinguished. The first occurs close to the surface, extending to a depth of nearly 2nm; the second occurs about a mean depth of approximately 4.5nm. The mean projected range of helium in tungsten at this dose has been calculated to be 3.7nm. The group of voids occurring close to the surface may be attributed to sputtering damage, where small clusters of vacancies may be produced due to displaced atoms on the surface and to a limited extent beneath the surface. The sputtering yields of low energy helium ions on tungsten are known to be low (McCracken and Stott (1979)). The group
of voids occurring close to the mean projected range would be expected following the assumption of a Gaussian range distribution, at which the maximum implanted helium would occur.

The two groups of voids are observed throughout the irradiation experiments, however, the effect becomes more pronounced at higher doses ($> 1 \times 10^{16}$ ions/cm$^2$). At the higher doses the numbers of voids observed at greater depths are larger, and several groups of voids may be discerned. More importantly however, the increased number of voids at depths greater than the mean projected range becomes more evident. Numerous voids of comparable magnitude to those occurring at the surface and the mean projected range, have been observed at depths greater than the calculated total range of the incident projectiles (see for example the 1keV helium irradiation at a dose of $5 \times 10^{16}$ ions/cm$^2$).

The two primary measurements in the irradiation study have been the void or bubble size and depth distribution of the voids as a function of ion dose and projectile energy. The results for the void sizes and depth distribution are shown in tabulated form in Table 5.1, for a range of ion doses and energies. One general trend observed is the increase in void size with ion dose for a given energy, in the helium energy range 0.2 to 3keV. In particular, the void size for 1keV helium irradiation shows a marked increase (nearly a factor of three) when the ion dose rises from $1 \times 10^{15}$ to $5 \times 10^{16}$ ions/cm$^2$. The growth of the bubbles or void size together with the number of voids with increasing ion dose observed in the study agrees with the predictions of several of the bubble and blistering theories. In particular, the gas coalescence
Table 5.1

Summary of data for helium irradiation of tungsten

<table>
<thead>
<tr>
<th>Helium ion Energy keV</th>
<th>Total ion Dose ions. cm⁻²</th>
<th>Mean void Size * nm</th>
<th>Observed Depth nm</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.2</td>
<td>1 x 10¹⁷</td>
<td>0.85 (0.78)</td>
<td>2.8</td>
</tr>
<tr>
<td>0.5</td>
<td>5 x 10¹⁶</td>
<td>0.43 (0.37)</td>
<td>5.4</td>
</tr>
<tr>
<td></td>
<td></td>
<td>2.93 (2.36)</td>
<td>~ 6</td>
</tr>
<tr>
<td>1.0</td>
<td>1 x 10¹⁵</td>
<td>0.54 (0.35)</td>
<td>8.2</td>
</tr>
<tr>
<td></td>
<td></td>
<td>0.70 (0.40)</td>
<td>~ 10</td>
</tr>
<tr>
<td>1.0</td>
<td>1 x 10¹⁶</td>
<td>0.80 (0.43)</td>
<td>13.2</td>
</tr>
<tr>
<td></td>
<td>5 x 10¹⁶</td>
<td>1.43 (1.08)</td>
<td>19.2</td>
</tr>
<tr>
<td>2.0</td>
<td>5 x 10¹⁵</td>
<td>0.36 (0.19)</td>
<td>13.2</td>
</tr>
<tr>
<td></td>
<td>1 x 10¹⁶</td>
<td>0.60 (0.30)</td>
<td>14.8</td>
</tr>
<tr>
<td>3.0</td>
<td>1 x 10¹⁵</td>
<td>0.44 (0.29)</td>
<td>&gt;5 nm</td>
</tr>
<tr>
<td>3.0</td>
<td>5 x 10¹⁵</td>
<td>0.64 (0.35)</td>
<td>12.6</td>
</tr>
<tr>
<td>3.0</td>
<td>1 x 10¹⁶</td>
<td>0.73 (0.43)</td>
<td>21.2</td>
</tr>
<tr>
<td>3.0</td>
<td>1 x 10¹⁷</td>
<td>0.60 (0.59)</td>
<td>19.2</td>
</tr>
</tbody>
</table>

* The numbers in parentheses indicate the standard deviation of the mean values.
(McCracken (1975)) and stress (Roth (1975)) models predict the growth of the bubbles with increasing ion dose due to the accumulation of implanted gas atoms at the trapping sites. However, in present study the further increase in ion dosage above $5 \times 10^{16}$ ions cm$^{-2}$ even at these low energies results in catastrophic specimen failure.

A similar trend is observed in the depth at which the voids extend from the irradiated surface, within the specimen volume examined by field evaporation. The effect of increasing the total ion dose produces a greater depth at which the voids occur (see for example 1keV helium irradiation) at a given ion energy. This observation is consistent with the theoretical predictions that increasing dose produces a higher concentration of implanted gas atoms (helium) along the total path length of the energetic ions. The increased implanted atom concentration, then permits the formation of voids, at the radiation induced trapping sites. Therefore, the number and extent of the voids within the sub-surface of the specimen is essentially dependent on the ion dose. Furthermore, increasing the ion dose enables the voids to form at depths corresponding to the total range of the ions. The energy of the incident projectiles thus defines the total depth to which the projectiles can become implanted and controls the ultimate depth at which voids may be formed.

The catastrophic failure of the specimen following high doses of helium irradiation occurs at about $1 \times 10^{17}$ ions cm$^{-2}$ at 1keV.

* Part of the work presented in this chapter has been published in J. Nucl. Mat. (1978) 76 & 77, 251.
despite repeated attempts to obtain FIM irradiation observations under these conditions. The reason for the failure is not clear, although some evidence for slip has been observed. This implies that the combination of the high electric field induced stresses and the existence of large voids close to the surface (with associated large deformation stresses), is thought to play an important role in specimen failure. The large voids (bubbles) may be produced by the rapid coalescence of the smaller voids, (Evans (1975)). The effect is reduced slightly at higher projectile energies (eg. 3keV), where failure occurs above doses of $1 \times 10^{17}$ ions. cm$^{-2}$. This may be due to the presence of large voids at greater depths from the specimen surface. However, the formation of blisters (due to the rapid agglomeration of the bubbles) on the surface may also produce unstable specimens and could lead to catastrophic failure, especially for blisters having dimensions comparable to the specimen tip radius.

The helium irradiation at low energy (200eV) has also shown void formation, although the doses at which the voids occur are high ($< 5 \times 10^{16}$ ions.cm$^{-2}$). The results show that planar defects (eg. dislocation) are also present in the sub-surface of the specimen. This is an important observation since the maximum transferred energy for such irradiation is $\sim 16$eV, while the threshold displacement energy for tungsten is usually taken as $\sim 25$eV. Thus, since the damage (in the form of voids) cannot have nucleated due to displacement events, a different mechanism may be responsible for void formation. One such mechanism is that of mutated trap formation, which has been predicted to occur in tungsten at high helium concentrations using computer modelling techniques (Van Veen et al (1977)). Wagner and Seidman (1978) have studied low-energy ($< 500$eV)
helium irradiation in tungsten and found that the helium atoms became trapped at interstices of the lattice and not at SIA's or vacancies. Therefore the accumulation of sufficient numbers of helium atoms at one trapping site may create a vacancy by the mutated trap mechanism, thus nucleating a void or bubble. The growth of the voids would then be controlled by the total dose of the implanted helium.

5.8 Helium irradiation of molybdenum

A series of irradiation experiments have been conducted in molybdenum specimens as a comparison with the study of irradiation damage in tungsten. Molybdenum is a useful technological material which has been considered as a first wall material in fusion devices, and has been used as limiters within current Tokomak designs (McC racken and Stott (1979)). Irradiation studies using molybdenum have been extensively reported (see for example McC racken (1975)), especially at higher hydrogen and helium ion energies (> 10keV). The work reported in this section deals with low energy helium irradiation (<1keV) and with ion doses in the range $1 \times 10^{15}$ to $5 \times 10^{17}$ ions.cm$^{-2}$.

5.8.1 Experimental procedure

The molybdenum specimens were prepared from commercially available wire (diameter 0.25mm) using the etching technique previously described in section 5.3(b). The molybdenum specimens were subsequently imaged in the FIM using Ne-He gas mixtures at pressures of up to $1 \times 10^{14}$ Torr. For larger radius specimens (R > 50nm) neon was used solely as
the imaging gas, the reduced electric field allowing the surface
topographical features to be more fully distinguished than with
helium imaging. The characterised emitter specimens were then
transferred to another UHV vacuum chamber and irradiated with helium
ions of known energy and ion dose in an identical manner to that
previously described in section 5.6.1. In a control experiment,
where non-irradiated specimens were examined, no deleterious surface
effects were discerned, and the bright spot contrast due to surface
contamination was found to be removed after the field evaporation
of one or two apex planes. The analysis of the irradiated molybdenum
specimens was conducted in an identical manner to that previously
described for the study in tungsten, using the void sizing tech-
nique, whereby successive micrographs were analysed for surface
damage under controlled field evaporation of the (011) apex plane
of the molybdenum specimens. Molybdenum has a bcc structure and
a similar emitter crystallography to tungsten; the central apex
plane of the specimen has an [011] orientation with an interplanar
spacing of 0.222nm.

5.8.2 Results

A series of irradiation experiments were conducted at a
helium ion energy of 1keV with ion doses ranging from $5 \times 10^{12}$ to
$1 \times 10^{15}$ ions.cm$^{-2}$. At this ion energy (1keV) the maximum trans-
ferred energy for a 'head-on' collision is 153eV, and sufficient to
produce Frenkel pair defects in molybdenum; which was a threshold
displacement energy $E_d = 40$eV. The sequence of micrographs shown
in Fig. 5.24 was recorded for a helium ion dose of $1 \times 10^{15}$ ions.cm$^{-2}$.
at 1keV. The previously characterised surface after irradiation is shown in Fig. 5.24(a), imaged in a Ne-He gas mixture (1 x 10^{-5} Torr Ne and 1 x 10^{-6} Torr He) at an imaging voltage of 6.0kV. The irradiated surface has numerous bright spot contrast due to adsorbed gases. However, small vacancy clusters may be distinguished on the emitter surface. Fig. 5.24(b) depicts the irradiated surface after 4(011) planes have been removed (0.9nm) where further small voids are evident together with some interstitial contrast.

The small vacancy complexes (voids) are also evident after continued field evaporation of the irradiated surface layers, shown in Fig. 5.24(c) after 10(011) planes were, removed (2.2nm). A small void can be distinguished in Fig. 5.24(d) in the (112) planar region after a total of 13(011) planes have been removed (2.9nm). Several small bubbles are also present in this particular planar region, shown in Fig. 5.24(e) after further field evaporation to a depth of 3.5nm. The final micrograph is shown in Fig. 5.24(f) after a total of 20(011) planes have been removed, corresponding to a depth of 4.4nm below the original irradiated surface. Small vacancy clusters are still evident, although the emitter topography has nearly returned to a characterised surface with minimal surface damage.

The void size and depth distribution for this irradiation experiment are illustrated in Fig. 5.25. The analysis was performed over 25(011) planes in which 37 voids were recorded. The void size distribution shows that voids range in size from 0.2 to 0.7nm with a mean void size of 0.4nm. The narrow void size distribution shows a maximum in the void having a size between 0.3 to 0.4nm. The void
Fig. 5.24 Irradiation of molybdenum with 1 keV helium ions after a dose of $1 \times 10^{15}$ ions cm$^{-2}$. Several small voids are indicated by the circles. The sequence of micrographs depict the irradiated specimen as:

a) The irradiated surface
b) The damage at a depth of 4(011) layers (6.9nm)
c) The damage at a depth of 10(011) layers (2.2nm)
d) The damage at a depth of 13(011) layers (2.9nm)
e) The damage at a depth of 16(011) layers (3.5nm)
f) The damage at a depth of 20(011) layers (4.4nm)
He-Mo
1 keV
$1 \times 10^{15}$ ions cm$^{-2}$

Fig. 5.25
Void size and depth distributions after 1 keV helium irradiation of molybdenum to a total dose of $1 \times 10^{15}$ ions cm$^{-2}$.
depth distribution indicates that the voids predominantly occur close to the irradiated surface ($< 2\text{nm}$ in depth) and gradually decrease in number, although larger voids extend to a total depth of approximately $4\text{nm}$ below the irradiated surface.

The helium irradiation of molybdenum at $1\text{keV}$ ion energy and a dose of $5 \times 10^{15}$ ions.$\text{cm}^{-2}$ was also conducted. The void size and depth distribution are shown in Fig. 5.26, where the analysis was performed over $36(011)$ planes removed from the irradiated surface. A total number of 60 voids were recorded within the analysis depth. The void sizes were found to range from $0.2$ to $1.5\text{nm}$ in a broad distribution, however, large numbers of voids were recorded with sizes between $0.3$ and $0.4\text{nm}$ and a mean void size of $0.47\text{nm}$. An increase in the number of voids with sizes greater than $0.5\text{nm}$ is observed compared with the irradiation dose of $1 \times 10^{15}$ ions.$\text{cm}^{-2}$. The depth distribution indicates that numerous voids are formed close to the irradiated surface similar to that observed in the lower dose irradiation. However, at this dose ($5 \times 10^{15}$ ions.$\text{cm}^{-2}$) the number of voids in the sub-surface region ($> 2\text{nm}$) has increased both in number and void size. The voids were found to extend to a depth of nearly $8.0\text{nm}$ beneath the irradiated surface. This agrees favourably with the numerical calculations based on LSS theory, where $1\text{keV}$ helium ions are found to have a total range of $8.6\text{nm}$ with a mean projected range of $5.0\text{mm}$.

Several experiments have been conducted at higher doses ($> 1 \times 10^{16}$ ions.$\text{cm}^{-2}$) at an ion energy of $1\text{keV}$. At a dose of $1 \times 10^{16}$ ions.$\text{cm}^{-2}$ the irradiated surface was found to have extensive surface damage, in the form of large numbers of voids interspersed with inter-
Fig. 5.26 Void size and depth distributions after 1 keV helium irradiation of molybdenum to a total dose of $5 \times 10^{15}$ ions cm$^{-2}$. 
stitial contrast indicating numerous displacement atoms. Spiral contrast indicating a dislocation was observed on the central (011) plane and further dislocation contrast was discerned on the (112) type planar regions. The dislocation contrast persisted from the surface to a total of 15(011) removed planes, corresponding to a persistence depth of 3.3nm. Numerous voids were distinguished on the emitter surface, the voids ranging in size from 0.3 to 6.0nm. Unfortunately the specimen suffered catastrophic failure after approximately 20(011) planes had been removed. This corresponds to a total depth of nearly 4.4nm beneath the irradiated surface. The cause of the specimen failure may be due to the high defect density present in the irradiated specimen, since extensive damage was observed in the sub-surface of the specimen, which together with the high electric field stresses may contribute towards the specimen failure. Further experiments at higher doses were conducted with neon as the sole imaging gas to reduce any field stress effects.

The result of 1keV helium irradiation at a dose of $5 \times 10^{16}$ ions.cm$^{-2}$ is illustrated in Fig. 5.27, the initial surface after irradiation however, was unstable under normal neon imaging conditions. The irradiated surface was extensively damaged in the form of voids and planar defects. However, the surface topography changed under imaging conditions, resulting in a surface atomic re-arrangement to form a new stable emitter topography. Therefore Fig. 5.27(a) depicts the original irradiated surface after atomic re-arrangement into a stable end form. The total number of (011) planes removed during the instability is not known, however a large elongated void ($\sim 7$nm) is present on the stable emitter surface, which was not apparent on the
Fig. 5.27  The damage following 1keV helium irradiation of molybdenum to a dose of $5 \times 10^{16}$ ions. Void and dislocation contrast are indicated by the circles and arrows in the field evaporation sequence.

a) The irradiated and partially collapsed surface
b) The damage after 27(011) layers removed (6nm)
c) The damage after 50(011) layers removed (11.1nm)
d) The damage after 60(011) layers removed (13.3nm)
e) The damage after 82(011) layers removed (18.2nm)
f) The damage after 109(011) layers removed (24.2nm)
irradiated surface. The occurrence of the large void may not be due solely to radiation damage. The possibility also exists that the damaged surface may result from the collapsing of the emitter surface.

Further controlled field evaporation of the emitter reveals further surface topography as the surface atoms surrounding the void are removed in successive layers. Fig. 5.27(b) shows the surface after 27(011) planes (6.0nm) have been removed from the stable surface. Severe planar distortion is evident on the (011) planar rings, the distortion may correspond to the emergence of a planar defect (dislocation) in the (011) plane. Several voids may also be distinguished on the emitter surface. Fig. 5.27(c) reveals the emitter surface after 50(011) planes (11.1nm) have been removed from the stable surface after irradiation. Numerous small voids are evident particularly in the (112) and (031) planar regions. Bright spot interstitial contrast may also be distinguished on the emitter surface.

Fig. 5.27(d) shows the surface after 60(011) planes (13.3nm) have been removed from the stable surface. Numerous small voids are evident, particularly in the (010) planar region, and small vacancy clusters are also present on the (112) plane with some interstitial contrast. The continued field evaporation is shown in Fig. 5.27(e) after 82(011) planes (18.2nm) have been removed from the stable surface. Extensive numbers of small voids are evident together with interstitial contrast. The final micrograph illustrated in Fig. 5.27(f) shows the surface after 109(011) planes (~23nm) have been removed, considerable surface damage is still evident. The surface topography is distorted and a series of small voids may be distinguished between the (010) and
(112) planar regions which were previously unresolved. The surface topography retained damage in the form of voids and planar defects for a further 15(011) planes, a total of 124(011) planes (27.5nm) at which point the surface regained a characterised appearance with minimal damage evident on the emitter surface.

The analysis of the void size and depth distribution was not performed due to the initial collapse of the emitter surface and the formation of a stable emitter topography. However the analysis of the remaining voids indicates that the voids present in the stable emitter surface persisted for a total number of 82(011) planes (18.2nm). The large void present on the stable emitter surface is elongated in shape with an approximate persistence of ~7nm. The extensive dislocation observed on the (011) plane persisted from close to the re-stabilised surface for nearly 27(011) planes removed (~6nm). The voids observed during the field evaporation sequence had sizes ranging from 0.3 to 7.0 nm, the majority of the voids having sizes between ~2.0 to 5.0 nm.

The effect of increasing the helium dose to a value of $1 \times 10^{17}$ ions.cm$^{-2}$ is illustrated in a series of micrographs in Fig. 5.28 after irradiation with 1keV helium ions. The original irradiated surface again suffered a rapid collapse, where an unknown number of (011) apex planes were removed during the imaging of the irradiated emitter surface and which prevents the formation of a depth scale. However, Fig. 5.30 reveals the extent of the damage after 32(011) planes (7.1nm) were removed from the re-stabilised irradiated emitter. The series of six micrographs were recorded after the sequential removal of one (011)
Fig. 5.28 The damage following 1keV helium irradiation of molybdenum to a dose of $1 \times 10^{17}$ ions cm$^{-2}$. Void and dislocation contrast are indicated by the circles and arrows in the field evaporation sequence.

a) The damage at a depth of 32(011) layers removed following the partial collapse of the irradiation surface.

b) - f) illustrates the extensive sub-surface damage after 1(011) layer has been removed in each following sequence.
planes and are shown in Figs. 5.30(a) to (f). The micrographs illustrate the emergence of a complex planar defect (dislocation) occurring on the central (011) plane, with several other defects evident on the (112) type planes. A large void (> 3.0nm) is manifest in the (031) planar region and numerous small voids with sizes ranging from 0.3 to 1.2nm are present on the emitter surface. Several small voids are particularly evident close to the defect present on the (011) plane (see Fig. 5.30(c) and (d)). The distortion of the voids (< 0.5nm) is evident, due to the intersection of the void and the emitter surface.

A further feature of the sequence of micrographs is the bright spot contrast, especially in the (011) planar ring system. Several bright spots appear as clusters, particularly in the (021) planar region, the effect decreasing with continued field evaporation and also present are reduced numbers of bright spot clusters on different regions of the emitter surface. The field evaporation of the re-stabilised emitter surface continued to a total number of 81(011) planes corresponding to a depth of 18.0nm below the collapsed irradiated surface. At this depth the surface damage present on the emitter surface was minimal with a well characterised emitter topography. A small number of voids were evident in the form of isolated vacancy clusters, although no other defects could be distinguished at this depth. The field evaporation study of this irradiated specimen following the stabilised emitter surface, showed that extensive sub-surface damage was present consisting of large voids, dislocations and bright spot contrast. The planar defects were extensive persisting in some cases for up to 20(011) planes, corresponding to a layer of nearly 4.5nm in
A further irradiation experiment was conducted with 1keV helium ions to a total dose of $5 \times 10^{17}$ ions.cm$^{-2}$. The molybdenum specimen was imaged using solely neon gas at an imaging voltage of 7.0kV, revealing severe surface damage consisting of large voids (> 1.0nm) with extensive distortion of the surface atoms forming bright spot clusters on the emitter topography. The emitter surface became unstable during imaging and collapsed revealing a large elongated void (> 5nm) in a severely damaged emitter surface. Several planar defects were distinguished on the (112) type planes. The continuation of further field evaporation resulted in the total collapse of the emitter surface leading to catastrophic failure of the specimen.

Several preliminary experiments have also been conducted in molybdenum at lower helium ion energies (500eV) to investigate the effects of high doses (> $5 \times 10^{16}$ ions.cm$^{-2}$) under low energy conditions, where the displacement energy is reduced to about 76eV. The results of a 500eV helium ion irradiation to a dose of $5 \times 10^{16}$ ions.cm$^{-2}$ is shown in a series of micrographs in Fig. 5.29. The first micrograph (5.29(a)) reveals the previously characterised surface following irradiation, where extensive surface damage is evident. Numerous small voids are interspersed between the larger voids and displaced surface atoms. The subsurface damage is illustrated in Fig. 5.29(b) after 9 central (011) planes have been removed (2.10nm) by field evaporation, spiral contrast is evident on the central (011) plane. Extensive surface damage is present on the emitter surface, manifest as voids (~ 0.5nm) and planar defects ((121) planar region).
Fig. 5.29  Low-energy (500eV) helium irradiation of molybdenum to a dose of $5 \times 10^{16}$ ions.cm$^{-2}$. The presence of void and dislocation contrast are indicated by the circles and arrows in the field evaporation sequence.

a) The irradiated surface (note the severe damage).
b) The damage at a depth of 9(011) layers (2nm)
c) The damage at a depth of 19(011) layers (4.2nm)
d) The damage at a depth of 24(011) layers (5.3nm)
e) The damage at a depth of 34(011) layers (7.5nm)
f) The damage at a depth of 49(011) layers (10.9nm).
In Fig. 5.29(c) the dislocation contrast on the (011) plane is still evident after 19(011) planes (4.2nm) have been removed. Several other planar defects are also present on the emitter surface together with some interstitial contrast. Fig. 5.29(d) reveals the emitter surface after 24(011) planes (5.3nm) have been removed, the dislocation contrast on the (011) plane may still be distinguished. Numerous small voids are evident over the emitter surface with associated image distortion due to the slight electric field enhanced emission from displaced surface atoms. The planar defect which was present on the (011) plane is not evident in Fig. 5.29(c) after 34(011) planes (7.5nm) have been removed. However, numerous small voids (< 1nm) are present on the emitter surface interspersed amongst a small number of large voids. The extensive damage located about the (112) planar region is reduced (compared with previous micrographs) although interstitial contrast is still evident in this planar region. The final micrograph depicted in Fig. 5.3(f) occurs after 49(011) planes have been removed corresponding to a total depth of 10.8nm beneath the initial irradiation surface. Minimal surface damage is evident in the form of planar or point defects.

The results of the void sizing analysis for this irradiation experiment are shown in Fig. 5.30 for an ion energy of 500eV and a total dose of 5 x 10^{16} ions.cm^{-2}. The void size distribution shows that the voids range from 0.3 to 3.5nm with a mean void size of 0.81nm (a standard deviation of 0.61nm). The void depth distribution reveals extensive surface and sub-surface damage in the molybdenum specimen. Relatively large numbers of voids occur close to the surface of the emitter and numerous voids are also present at greater depths (~ 4.0nm) beneath the
He–Mo
500eV
$5 \times 10^{16}$ ions cm$^{-2}$

Fig. 5.30
Void size and depth distributions after 0.5 keV helium irradiation of molybdenum to a total dose of $5 \times 10^{16}$ ions cm$^{-2}$. 
irradiated surface. The voids were found to exist to a total depth of 10.2nm beneath the surface. A large planar defect due to a dislocation emerging on the (011) plane in the irradiated specimen was found to persist for a total depth of 6.5nm close to the irradiated surface. Smaller planar defects had a total persistence of approximately 2nm in the sub-surface region of the specimen. Further irradiation experiments at 500eV ion energies with higher doses (> 1 x 10^{17} ions.cm^{-2}) resulted in catastrophic failures of the specimens during initial imaging of the irradiated surfaces. No information regarding the formation of voids occurring close to the surface could be deduced from these experiments due to the total failure of the molybdenum specimen.

5.8.3 Discussion

The helium irradiation of molybdenum at low energies (0.5 and 1keV) has been studied in the dose range 1 x 10^{15} to 5 x 10^{17} ions.cm^{-2}. The radiation damage is similar to that observed for tungsten, consisting of small voids in the form of vacancy clusters, interstitials and complex planar defects. The study of the helium irradiation in molybdenum indicates the growth of the voids with increasing ion dose as shown in Table 5.2. A concomitant increase in apparent interstitial contrast is observed and in the number and extent of planar defects such as dislocations. The number of interstitial atoms formed by helium irradiation appears to increase with total ion dose and energy. However, no quantitative measurements have been conducted during depth analysis due to the ambiguous nature of these defects at the surface of the FIM specimen. Similarly the planar
Table 5.2

Summary of data for helium irradiation of molybdenum

<table>
<thead>
<tr>
<th>Helium Ion Energy keV</th>
<th>Total Ion Dose ions.cm⁻²</th>
<th>Mean Void Size + nm</th>
<th>Observed Depth nm</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.5</td>
<td>5 x 10¹⁶</td>
<td>0.81(0.61)</td>
<td>10.2</td>
</tr>
<tr>
<td>1.0</td>
<td>1 x 10¹⁵</td>
<td>0.4(0.12)</td>
<td>4.2</td>
</tr>
<tr>
<td>1.0</td>
<td>5 x 10¹⁵</td>
<td>0.41(0.23)</td>
<td>6.8</td>
</tr>
<tr>
<td>1.0</td>
<td>1 x 10¹⁶</td>
<td>~ 0.3-0.6</td>
<td>4.4 *</td>
</tr>
<tr>
<td>1.0</td>
<td>5 x 10¹⁶</td>
<td>~ 0.3-0.7</td>
<td>27.5 *</td>
</tr>
<tr>
<td>1.0</td>
<td>1 x 10¹⁷</td>
<td>-</td>
<td>&gt; 18.0</td>
</tr>
</tbody>
</table>

* The numbers in parentheses indicate the standard deviation of the mean values.

* Partially collapsed surface, where an unknown number of (011) layers removed.
defects rise in number, depth and extent (persistence) with increasing ion dose beneath the irradiated surface.

The void size distribution in helium irradiated molybdenum is similar to that of tungsten, where numerous vacancies and their clusters (< 0.5 nm) are produced at lower doses. Furthermore, the same general features in the depth distributions of the voids are observed; in that sputtering damage occurs on the molybdenum surface, with the formation of numerous voids close to the mean projected range. At the lower doses, close agreement is achieved with LSS calculated mean projected range and total ranges. However, at the higher doses, specimen failure has been frequently observed and no observations of the irradiation damage in molybdenum has been obtained at doses greater than $1 \times 10^{17}$ ions cm$^{-2}$.

The specimen failure has been attributed to the same mechanism as that of high dose helium irradiation in tungsten, where large voids (> 5 nm) occur close to the irradiated surface. The formation of the large sub-surface voids may be due to the rapid coalescence of the smaller voids. These results suggest that molybdenum may have a similar radiation damage behaviour as tungsten, in that bubble growth and especially blister formation may be initiated at the same total ion doses. However, the exact depth at which the large voids are situated has not been determined due to the removal of an unknown number of apex planes during specimen failure.

Extensive damage has been observed at the lower doses ($\sim 1 \times 10^{16}$ ions cm$^{-2}$), and evidence has been recorded showing that the damage extends
\( \sim 20 \text{nm} \) beneath the irradiated surface at higher doses (\( \geq 5 \times 10^{16} \text{ ions.cm}^{-2} \)). The depth of damage appears to be greater than that predicted by LSS theory for the total range of helium in molybdenum at these low energies and high doses. A similar feature of the damage produced by 500eV helium ions is also observed at a dose of 5 \( \times 10^{16} \text{ ions.cm}^{-2} \). Furthermore, the maximum transferred energy of 156eV for 1keV helium ions is greater than that for tungsten under similar conditions, thus channeling or focussed collision sequences may produce higher implanted helium penetrating into the molybdenum specimen.
Conclusions and Suggestions for Future Work.

6.1 Field-ion microscopy

A comprehensive review of the basic principles of field-ion microscopy, including imaging theory, field evaporation and resolution of the technique has been provided. The efficacy of the technique for the investigation of low-energy radiation damage has been outlined, especially related to the nucleation and growth of damage defects, where the influence of field induced stresses and choice of imaging gases has been described.

In this thesis the interpretation of field-ion microscopy has been reviewed, where the basic crystallography and topography of the emitter surface have been examined. The projection geometry of the FIM has been reviewed and the theoretical treatment of the projection geometry has yielded several projection relationships. The introduction of the moiré analogue has enabled further information to be deduced regarding the shape of projected image ring shapes from an idealised emitter topography.

The application of the moiré theory to the treatment of irregularly shaped planar facets in field-ion micrographs has been investigated. The use of non-spherical zone plates representing these facets has been analysed and several new relationships have been developed. The detailed analysis of the moiré patterns formed by the overlapping zone plates has
given rise to generalised equations which characterise the patterns in terms of shape, location, orientation and phase. Several important features have been deduced concerning the location and orientation of the patterns, which have useful applications in the analysis of damaged emitter surfaces. Further work is necessary to correlate the theoretical model with actual field-ion micrograph data regarding the properties of the non-spherical planar facets.

The theoretical treatment of field evaporation has been discussed in detail, where the relative removal rates of different planar regions of the emitter surface have been deduced. The idealised model has shown that the planar removal rate has a cosine dependence on the number of apex planes removed. A similar result has been derived for a non-spherical idealised emitter surface. The theoretical model and the correlated experimental measurements form the basis of a semi-quantitative size analysis technique, which is an essential requisite for radiation damage studies. The controlled field evaporation technique therefore provides a powerful method for the analysis of low-energy radiation damage. In addition, the technique has several applications in metallurgical studies concerned with the analysis of microstructural phases. Further experimental work, however, is required to extend the technique on a range of materials, including fcc metals. It is likely that the next stage of development of the technique will lie in the analysis of the evolution of the emitter topography during field evaporation.

6.2 Low-energy ion beams

The production of low-energy ion beams by several types of ion
sources has been investigated; the sources include, a simple ion source (AGl), a mass-analysed ion source, a commercial ion source and the cathode sputtering technique. In each case the ion beam characteristics have been examined and the emphasis placed on each type of ion source to produce ion beams suited to the experimental requirements. The major part of the irradiation experiments conducted in this study has been performed using the commercial ion source, where ion bombardment occurs in a direction parallel to the emitter axis in a reproducible manner under controlled experimental conditions.

A novel type of ion source has been described, wherein a modified simple ion source (AGl) has been electrically isolated to facilitate 'in-situ' helium ion-bombardment using the imaging gas within the FIM. This source may enable the direct observation of damage, especially in sputtering and surface deformation (blistering) studies. Further work incorporating realtime photographic techniques, such as successive time dependent 35mm photography and/or closed circuit TV may be employed to directly monitor the surface topography of the specimen under irradiation.

6.3 **Low-energy radiation damage.**

The FIM study has revealed new information concerning the nucleation and growth of voids leading to blister formation following low-energy ion irradiation of tungsten and molybdenum. This is the first detailed study, on the atomic scale, of the defects produced by low-energy light ion irradiation leading to blister formation, especially at these low-energies (<5keV). This has been achieved by the combined
use of the FIM and a semi-quantitative analysis technique, where the
precise nature and extent of the damage has been established. A
preliminary study of the damage caused by low-energy hydrogen and
helium in tungsten and molybdenum respectively has been conducted
using a cathode sputtering technique and a mass-analysed source to
establish the doses necessary for the formation of defect complexes
within the specimen. However, the major part of this thesis concerns
the low-energy damage, where the irradiation has occurred in a direc-
tion parallel to the emitter axis. Thus, the depth of damage has been
determined for helium in tungsten and molybdenum respectively, for
ion energies of 0.2 to 3 keV and doses in the range $1 \times 10^{15}$ to $5 \times 10^{17}$
ions.cm$^{-2}$.

The study of the helium irradiation of tungsten has shown that
the damage consists of point defects (interstitials and vacancies),
vacancy clusters (voids) and more extensive defects such as dislocations.
The small voids occur as irregular clusters, the apparent void shape
becoming symmetrical at larger void sizes and finally become elongated
(usually along the emitter axis). The interstitial contrast appears
qualitatively to be related to increasing ion dose and energy, furthermore, the observed interstitial contrast appears to vary with depth be-
neath the irradiated surface. Isolated dislocations have been observed
after low dose irradiation at a range of ion energies; however, the
frequency of occurrence and extent (in terms of persistence) of the
dislocation contrast increases with ion dose. Complex planar defect
contrast has been found to occur at the higher ion dose/energy combinations.
The size distribution of the radiation induced voids has been shown to be dependent on the ion dose, where the mean void size increases with helium ion dose. The depth of the damage has also been deduced from the controlled field evaporation technique, where it has been shown that the depth of damage (in terms of void formation) is dependent on the total ion dose. For a given ion energy, the depth of damage increases with dose until the observed damage extends from the surface to the total range of the implanted ions. The distribution of damage with depth beneath the irradiated surface has been shown to exhibit several features indicating two forms of damage. The first form consists of surface damage, where groups or clusters of smaller voids (vacancy clusters) are situated on or close to the surface. This has been attributed to sputtering by the incident helium ions. Secondly, the damage present within the sub-surface of the irradiated specimen has a higher concentration of voids situated close to the theoretical mean projected range of the implanted ions. Furthermore, the effect becomes more pronounced at higher doses. Specimen failure was found to occur at the higher doses and has been attributed to the combination of field induced stresses and the presence of large sub-surface voids with associated deformation forces.

The observed damage produced by 200eV helium ions in tungsten has shown that void formation (vacancy clusters) occurs at high doses. Due to the low transferred collision energies, less than the threshold displacement energy, the presence of helium in interstitial sites leading to a mutated trap mechanism is thought to play an important role in low-energy void nucleation. Further low-energy experiments are necessary to elucidate the actual mechanism leading to void nucleation.
The study of low-energy (0.5 and 1.0keV) helium irradiation of molybdenum has shown that the damage is analogous to that in tungsten. The damage consists of vacancies and their clusters, interstitials and dislocations. Similarly the nature and extent of the damage (and void sizes) increases with ion dose. Moreover, the depth distribution of damage reveals the occurrence of surface (sputtering) damage and void grouping close to the theoretical mean projected range. However, although the depth of damage at low doses agrees with theoretical range calculations, the observed depths of damage are greater at the higher doses.

In conclusion, the FIM study has shown that the low-energy damage produced by implanted helium ions in tungsten and molybdenum appears to follow the nucleation and growth of voids as predicted by the gas bubble coalescence model for doses $< 1 \times 10^{17}$ ions.cm$^{-2}$. However, no unambiguous evidence has been obtained regarding the rapid coalescence or agglomeration of voids, which is predicted to occur at the onset of surface blister formation. The damage produced by helium irradiation at doses $> 1 \times 10^{17}$ ions.cm$^{-2}$ usually results in catastrophic failure. A parallel TEM study, including FIM specimens, is therefore required to establish the occurrence of any void coalescence; especially at the higher doses. Such work would go some way towards elucidating the precise blistering mechanism. Future work using the already developed FIM technique should also extend the range of materials used in the study of low-energy damage; one such example would be stainless steel. In this study the low-energy damage has been investigated under room temperature and liquid nitrogen (cathode sputtering) irradiation conditions, however, the damage produced under different elevated temperatures should also be considered, particularly on void growth and its effect on blister formation.
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