Direct current generation due to harmonic mixing: from bulk semiconductors to semiconductor superlattices

Direct current generation due to harmonic mixing: From bulk semiconductors to semiconductor superlattices

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We discuss an effect of dc current and dc voltage (stopping bias) generation in a semiconductor superlattice subjected by an ac electric field and its phase-shifted n-th harmonic. In the low field limit, we find a simple dependence of dc voltage on a strength, frequency, and relative phase of mixing harmonics for an arbitrary even value of n.

We show that the generated dc voltage has a maximum when a frequency of ac field is of the order of a scattering constant of electrons in a superlattice. This means that for typical semiconductor superlattices at room temperature operating in the THz frequency domain the effect is really observable. Indeed, such conditions are most used in the modern experiments on THz harmonics generation in superlattices.

We also made a comparison of a recent paper describing an effect of a directed current generation in a semiconductor superlattice subjected by ac field and its second harmonic ($n = 2$) [K. Seeger, Appl. Phys. Lett. **76**, 82 (2000)] with our earlier findings describing the same effect [K. Alekseev *et al.*, Europhys. Lett. **47**, 595 (1999); cond-mat/9903092]. In particular, here we found that the maximum value of dc current is associated with the THz domain that also follows from Seeger's calculations. This maximum value has already been highlighted in our previous work.

For the mixing of an ac field and its n-th harmonic with $n \geq 4$, we found that additionally to the phase-shift controlling of the dc current, there is a frequency control. This frequency controlling of the dc current direction is absent in the case of $n = 2$. The found effect is that, both the dc current suppression and the dc current reversals exist for some particular values of ac field frequency. For typical semiconductor superlattices such an interesting behavior of the dc current should be observable also in the THz domain.

Finally, we briefly review the history of the problem of the dc current generation at mixing of harmonics associated with arbitrary coherent electromagnetic waves in semiconductors and semiconductor microstructures.

I. INTRODUCTION

It is natural that a coherent mixing of the waves with commensurate frequencies (like, for example, having different harmonics) in a nonlinear medium can result in a product which has a zero frequency or a static (dc) electromagnetic field. If such a nonlinear interference phenomenon will happen in semiconductors or semiconductor devices, like a semiconductor superlattice (SSL), then the static electric field may result into a dc current or a dc voltage generation. Such dc current (DC), which may be created in stationary or nonstationary regimes, depends on amplitudes, frequencies, and relative phases of mixing waves.

It was Rosenblat [1] who first at the end of 40th suggested that the dc component of electric or magnetic field may arise in pure ac-driven electrical circuits with nonlinear and symmetric characteristics. A similar idea has been proposed by Skov and Pearlstein in the middle of 60th [2]. Soon after these theoretical predictions, the DC due to mixing of ac electric field and its second harmonic has been observed for the warm electrons in germanium by Pozhela and Karlin [3], and, independently, by Schneider and Seeger [4].

Our paper is related to the same issue of the DC generation and consists of two parts. The first part has been stimulated by recent paper [5] and contains original results on the DC generation due to harmonic mixing in semiconductor superlattices, as well as a comparison of the Seeger results [5] with the results obtained earlier in our previous paper [6]. The second part is devoted to a short review of papers devoted to the harmonic mixing induced DC in bulk

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semiconductors and SSLs. Here, a special attention is paid to a consideration of different mechanisms of nonlinearity in semiconductors leading to their non-ohmic behavior.

II. THE DIRECT CURRENT DUE TO A MIXING OF ARBITRARY HARMONICS IN A SEMICONDUCTOR SUPERLATTICE

An appearance of DC in SSL [7] under simultaneous action of ac electric field and its phase-shifted second harmonic has been recently shown by Seeger in the theoretical article [5]. He attributed this DC effect to an "Esaki-Tsu non-ohmicity of SSL for the case where it Bloch oscillates" and pointed out that a DC direction depends on the phase-shift [5]. In the present paper we describe a DC arising in SSL due to mixing of ac field and its phase-shifted n-th harmonic, where n is arbitrary even number, like, $n = 2, 4, 6, 8, 10$. Our main findings are following.

First, we show that for a weak field strength, the DC is maximal only when the mixing arises for waves having the THz frequencies. Thus, the effect is relevant to a strong current interest to THz-field harmonics generation in SSLs [8,9].

Second, we show that for a mixing of ac field and its n-th harmonic with $n \geq 4$ in typical SSLs there appear reversals of direction of the DC and the DC may vanish for some particular frequencies, which values also belong to the THz frequency domain. This novel mechanism of the frequency-control is supplementary to the control of magnitude and direction of DC using a variation of phase shift. Moreover, the controlling of the phase shift is a difficult task due to a very short dephasing time. On the other hand the frequency control is easy to handle. The conditions for DC vanishing are different from the well-known conditions for a dynamic localization in ac-driven superlattices [10–12]. The effect found is important because of a strong interest to high-order harmonic generation in SSLs [13,14].

Consider a mixing of the first and the $\phi-$phase-shifted n-th harmonics of the ac electric field

$$E(t) = E_1 \cos(\omega t) + E_n \cos(n\omega t + \phi). \tag{1}$$

in a single miniband SSL having the tight-binding energy-quasimomentum dispersion relation

$$\varepsilon(k) = \frac{\Delta}{2}\left[1 - \cos(ka)\right], \tag{2}$$

where k is an electron wave vector along the axis of SSL with the spatial period a and with the miniband width Δ. The current induced in SSL is proportional to the electron velocity $v = \hbar^{-1}\partial\varepsilon/\partial k$. Using a standard semiclassical theory of the wave mixing in SSLs [15–17], we obtain the following expression for the DC

$$j_{\rm dc} = j_0 \sum_{\mu_1,\mu_2=-\infty}^{+\infty} \sum_{\nu=-\infty}^{+\infty} \frac{(\mu_1 + n\mu_2)x\cos(\nu\phi) + \sin(\nu\phi)}{1 + (\mu_1 + n\mu_2)^2 x^2} J_{\mu_1}(\xi_1) J_{\mu_2}(\xi_n) J_{\mu_1 - n\nu}(\xi_1) J_{\mu_2+\nu}(\xi_n), \tag{3}$$

where $x = \omega\tau$, $j_0 = \frac{\hbar\sigma}{e\tau a}$ with σ being a static, ohmic conductivity along the SSL's axis, τ is a characteristic scattering time, $J_\mu(\xi)$ is the Bessel function, and $\xi_1 = (eaE_1)/(\hbar\omega)$, $\xi_n = (eaE_n)/(n\hbar\omega)$. The Eq. (3) is a direct generalization of the corresponding expression for DC derived in Ref. [6] for the case $n = 2$ and is an exact result obtained from the Boltzmann equation with a single constant relaxation time. This equation is also consistent with a corresponding Eq. (6) of the Seeger's paper [5], where, however, a summation over the phase variable has been neglected. In the limit of a weak field $\xi_1, \xi_n \ll 1$, and using $J_l(\xi) \approx (\xi/2)^l (1/l!)$, from Eq. (3) we obtain

$$\xi_{dc} = -f_n(x)\xi_1^n \xi_n \cos\phi + O(\xi_1^l \xi_n^k), \quad l + k = n + 3. \tag{4}$$

Here, following the Ref. [18] we have introduced the stopping bias as $E_{dc} = j_{dc}/\sigma$ and the scaled dc voltage as $\xi_{dc} = (eaE_{dc})/(\hbar\omega)$. The nontrivial prefactor $f_n(x)$ is nonvanishing for any even value of n and has the following forms

$$f_2(x) = \frac{3}{2}\,\frac{x^2}{1 + 5\,x^2 + 4\,x^4}, \tag{5}$$

$$f_4(x) = \frac{5}{4}\,\frac{x^4\left(-1 + 5\,x^2\right)}{1 + 30\,x^2 + 273\,x^4 + 820\,x^6 + 576\,x^8}, \tag{6}$$

$$f_6(x) = \frac{21}{32}\,\frac{x^6\left(1+84\,x^4-35\,x^2\right)}{1+91\,x^2+3003\,x^4+44473\,x^6+296296\,x^8+773136\,x^{10}+518400\,x^{12}}, \tag{7}$$

$$f_8(x) = \frac{9}{32}\,\frac{1}{\Psi_8(x)}\left(126\,x^2+3044\,x^6-1-1869\,x^4\right)x^8, \tag{8}$$

$$\Psi_8(x) = 1+204\,x^2+16422\,x^4+669188\,x^6+14739153\,x^8+$$
$$173721912\,x^{10}+1017067024\,x^{12}+2483133696\,x^{14}+1625702400\,x^{16}, \tag{9}$$

To derive these formulae for $f_n(x)$, we have performed symbolic computations using MAPLE with a summation in all indices in Eq. (3) from -12 up to 12. We should note that, as follows from Eq. (4), the dc voltage depends only on the first degree of the n-th harmonic's field strength E_n, as well as on the phase difference between the first and the n-th harmonic via cosine term. An interesting physical meaning has the nontrivial prefactor $f_n(x)$.

We start the analysis of the obtained expression with the case of $n = 2$ [Eq. (5)]. The function $f_2(x)$ is always positive and has a maximum at the value $x_{max} \approx 0.71$ with $f_2(x_{max}) \approx 0.17$ (see Fig. 1a). Note that an expression analogous to Eqs. (4),(5) has been obtain in our earlier work [6]; the same form can be also derived from Eq. (7) of the Ref. [5].

For $n \geq 4$ the functions $f_n(x)$ demonstrate more complex behavior. So, the function $f_4(x)$ may be both positive and negative(see Fig. 1b), has a zero value at $x_0 = 5^{-1/2} \approx 0.45$, has a maximum at $x_{max} \approx 1.07$ [$f_4(x_{max}) \approx 2.96 \times 10^{-3}$] and a minimum at $x_{min} \approx 0.29$ [$f_4(x_{min}) \approx -8.58 \times 10^{-4}$]. In the limit $x \to 0$ this function has a negative derivative.

The function $f_6(x)$ (see, Fig. 1c) has two zeros at the value $x_0^{(1)} \approx 0.62$ and the value $x_0^{(2)} \approx 0.18$. Its global maximum and global minimum are located at the value $x_{max}^{(1)} \approx 1.27$ [$f_6(x_{max}^{(1)}) \approx 2.27 \times 10^{-5}$] and at the value $x_{min}^{(1)} \approx 0.4$ [$f_6(x_{min}^{(1)}) \approx -1.18 \times 10^{-5}$], respectively. The second small maximum, which is almost invisible on the scale of Fig. 1c, exists at the value $x_{max}^{(2)} \approx 0.14$ with the function value $f_6(x_{max}^{(2)}) \approx 4 \times 10^{-7}$. In the limit $x \to 0$ this function has a positive derivative.

The function $f_8(x)$ (see, Fig. 1d) is vanishing in three points, i.e. it has three zeros at the value $x_0^{(1)} \approx 0.73$, the value $x_0^{(2)} \approx 0.26$ and at the value $x_0^{(3)} \approx 0.1$. Its global maximum and minimum are located at the value $x_{max}^{(1)} \approx 1.4$ [$f_8(x_{max}^{(1)}) \approx 9.58 \times 10^{-8}$] and at the value $x_{min}^{(1)} \approx 0.48$ [$f_8(x_{min}^{(1)}) \approx -6.5 \times 10^{-8}$], respectively. The other smaller maxima and minima exist at the points: $x_{max}^{(2)} \approx 0.2$ with $f_8(x_{max}^{(2)}) \approx 6.6 \times 10^{-9}$ and $x_{min}^{(2)} \approx 0.08$ with $f_8(x_{min}^{(2)}) \approx -4 \times 10^{-11}$. In the limit $x \to 0$ this function has a negative derivative.

All functions, $f_n(x)$, with $n \geq 4$ have maximums and minimums for $x \simeq 1$ with $\max|f_n(x)| \simeq 10^{-n+1}$ and show asymptotically universal behavior: $\propto x^n$ for $x \ll 1$ and $\propto x^{-2}$ for $x \gg 1$. For typical SSLs at room temperature the scattering time τ is of the order of 10^{-13} sec [19,9], therefore, the condition $x = \omega\tau \approx 1$ corresponds to a frequency of about several THz. Thus, the most effective generation of DC arises in the THz domain of frequencies. This is an important conclusion missed in the Ref. [5]. The condition, $\omega\tau \approx 1$, at which a maximal DC arises, is just typical for experiments on the THz-field harmonic generation [8,9].

The fact that the functions $f_n(x)$ with $n \geq 4$ can change a sign means that the multiple DC reversals are possible not only by a change of the phase difference ϕ, but also *by a variation of the ac frequency itself at the fixed phase difference*. The multiple DC reversals in ac-driven SSL is a new effect; earlier such a behavior of the DC has been described only in the ratchet systems, i.e., for a particle moving in a periodic *asymmetric* potential in an external ac field [20,21].

We also should note that the effect of the harmonic-mixing-induced DC vanishing for a particular value of x is different from the well-known dynamic localization of electrons in ac-driven SSL [10–12][1]. Indeed, we have a suppression of the dc current component in the limit of a weak field $\xi_{1,n} \ll 1$ and for $\omega\tau \simeq 1$, while there in the dynamic localization arises a whole current suppression for a scaled field strength greater or of the order of unity and for $\omega\tau \gg 1$ [10–12,22].

[1] Recently, the theory of a dynamical localization in tight-binding lattices was generalized to the case of a two-frequency driven field (see, Ref. [22]). In a comparison with the case of a single-frequency field [11,12], the localization in the two-color field is more robust and takes place for a more wide range of the field amplitude and the frequencies [22]. However, the effect is also observable only for $\omega\tau \gg 1$.

III. HISTORICAL PREVIEW

In different semiconductors the different types of nonlinearity (or their combinations) can result in the DC at harmonic mixing. The most universal nonlinearity is caused by a nonparobolicity of conduction band. This type of nonlinearity is mainly typical for the semiconductors and semiconductor microstructures with the relatively wide bands and narrow gaps, as well as for an opposite case of the narrow band and the wide gap materials [16]. The later class includes SSLs. Another main type of nonlinearity is the carrier's heating mechanism, which is mainly responsible for the DC at wave mixing the semiconductors with wide band and wide gap [3,4]. Which type of nonlinearity would be dominant for the effects associated with the wave mixing in many semiconductors with competing nonlinearities, strongly depends on the temperature and the frequencies of applied ac fields [23]. In the bulk semiconductors the effect of the DC generation has been mainly considered for the mixing of microwaves, while for the SSLs the main interest is paid to the THz frequency domain (submillimeter wavelengths). In this part of the work we briefly review both situations.

A. Wave mixing in bulk semiconductors

The DC at the mixing of microwaves having commensurate frequencies in such semiconductors as n-Ge and n-Si has been intensively studied both experimentally and theoretically by Vilnus [3,24–28] and Wien [4,29,30] groups. The experiments were performed at 77 K or at room temperatures and the majority of them dealt with the mixing of the fist and the second harmonics of coherent microwaves, although the effect of mixing of even higher harmonics (up to 6th) has been also considered in Ref. [25]. The main nonlinear mechanism responsible for a generation of the DC at wave mixing in these semiconductors was found to be the electron's heating by the field. It has been shown that a simple model based on balance equations and incorporating a parabolic band and the dependence of the carrier's relaxation time on the field (or on their energy), can well describe the effect in Ge and Si [29]. It was also suggested that the effect is suitable for a determination of different carrier's relaxation times [29,26,27].

Independently from this research the DC generation in semiconductors associated with a mixing of a coherent electromagnetic wave with its second harmonic has been also described in theoretical papers [31–33] and named as "coherent photovoltaic effect". The description based on a quantum kinetic equation for the model of free electrons interacting with acoustical phonons was used in the paper [31], while the paper [32] dealt with an approach of the classical Boltzmann equation with collisional integrals incorporating a field absorption by free classical electrons or by impurity-band transitions. Finally, the paper [33] suggested that a mechanism of the DC generation at the wave mixing can be associated with quantum corrections to a nonlinear rf-conductivity. We should stress that in all enumerated papers the band has been assumed to be parabolic and the nonlinearity was caused by different forms of the dependence of the carrier's relaxation time on the electron energy or on the field strength.

However, the experiments of Patel *et al.* [34] have clearly demonstrated that an optical waves mixing in several III-V type semiconductors with narrow gaps, such as n-doped InAs, InSb and GaAs, can not be explained without taking into account the miniband nonparabolicity as a source of nonlinearity within the material[2] [35–37]. In these narrow-gap semiconductors the conduction band has a strong nonparabolicity due to the strong interaction with valence band. It is conventionally described by a Kane two-band or a four band model, which has a strong nonlinear dispersion law [38,39].

Later on, several experiments on the mixing of mm-waves in the same materials at liquid helium and 77K temperatures have been performed [41–45]. Their analysis indicate that for microwaves both nonlinearity mechanisms, due to the non-quadratic dispersion law in the conduction band and to changes in the collision time because of electron heating, are important [42,23,46]. The estimates presented in Refs. [23,46] for such semiconductors as n-InSb and n-GaAs demonstrate that for the high frequencies ($\gtrsim 10^{13}$ s^{-1}) and moderate temperatures, the so-called "dynamical nonlinearity" (band nonparabolicity) prevails, in a contrast to a case of low frequencies and low temperatures, where the heating nonlinearity mainly predominates.

Finally, an experiment on the third harmonic generation of a submillimetere radiation ($\lambda_\omega = 0.9$ mm, $\lambda_{3\omega} = 0.3$ mm) in the n-type indium antimonide has been described in the Ref. [47]. There the main mechanism of a nonlinearity

[2] Also note that nonparobolicity of electron dispersion law as possible mechanism for the strong third-order optical mixing has earlier been pointed out by Lax *et al.* [40].

driving the wave mixing was identified as being related to a nonparobolicity of the conduction band. However, to the best of our knowledge, the possibility of the DC generation due to a harmonic mixing of THz fields in such narrow gap bulk semiconductors was recently discussed only in our paper [6].

B. Wave mixing in semiconductor superlattices

Another class of semiconductors, in which the band nonparabolicity should be important, is associated with materials having narrow bands and wide gaps, such as SiC and ZnS. Recently a new class of the systems with narrow bands appear: Arrays of quantum dots or quantum wells. One of the most popular and most studied system is a semiconductor superlattice (SSL) consisting of a series parallel layers with quantum wells. In general the SSL [7] may be viewed as an artificial crystal having narrow (mini-)bands and relatively wide gaps. Nowadays these semiconductor microstructures attracts much attention [48].

Already in the first paper on SSLs, Esaki and Tsu suggested that the miniband nonparabolicity is a main source of nonlinearity, which can be used for an observation of Bloch oscillations and a negative differential conductivity [7]. For low or moderate field strengths, the nonlinear transport properties of a single miniband SSL could be well described in the framework of a semiclassical Boltzmann equation with a constant relaxation time [7,49]. The theoretical predictions obtained within such an approximation are in a good agreement with the measurements of voltage-current characteristics in SSLs for a temperature above 40 K [50,51]. On the other hand, there is an important theoretical result by Suris and Shchamkhalova [52], which states that a collisional integral in the quantum kinetic equation is practically independent on the field in the low field limit. This and other theoretical results [53,54] give a support for an application of a constant relaxation time approximation for low field and high temperatures.

In 1971 Esaki and Tsu [15] and Romanov [16] suggested to use the SSL as a new, artificial, nonlinear material for electromagnetic wave mixing and new harmonics generation. They showed that strong SSL's miniband nonparabolicity could be source of nonlinearity for harmonics generation in analogy with the case of bulk semiconductors studied in Refs. [34,35]. The theory of wave mixing in SSLs, based on a solution of the Boltzmann equation with a constant relaxation time for single miniband, has been developed in works [55,56,17,57] (see also [49]). Our result on the DC generation due to mixing of the first and the second harmonic [6], as well as corresponding results by Seeger [5], could be immediately derived using the results of previous paper by Orlov and Romanov [17].

Earlier the DC due to the mixing of ac field and its second harmonic in the tight-binding lattice has been described by Goychuk and Hänggi [58] using a theory of quantum ratchets. The so-called ratchet system consists of a particle moving in a periodic *asymmetric* potential and under the action of an external time-dependent force with a zero average [59,60]. The force can include a noise or can be pure deterministic and spatially uniform ("rocking ratchets") [61]. Both types of ratchets demonstrate a rectification effect. For a recent realization of the ratchet system in an antidot array of SSL driven by a far-infrared field, see, Ref. [62]. Another mechanism for the DC generation due to a mixing of first and second harmonics in a quantum tight-binding lattice has been considered in the Ref. [22]. Following [22] the DC can be generated if the intersite coupling constants of the lattice are linearly dependent on a lattice site number.

Among other recent theoretical results related to the DC arising at the harmonic mixing in superlattices, we would like to mention a consideration of SSL driven by a monochromatic ac field [63–67]. There, in single miniband of SSL a self-consistent field is generated by electron motion [68–70]. Within this model the transport properties of SSL are effectively controlled by a mixing of an external, pure harmonic driving field and the self-consistent field [6,63].

For an ac field strength which is lower than a some critical value, a Fourier spectrum of the self-consistent field contains only odd harmonics and, therefore, the DC can not be generated. However, for an external ac field, which is strong enough, the self-consistent field is generated and its Fourier spectrum contains even harmonics as well. The mixing of the even and the odd harmonics results in the DC [6]. For a relatively weak scattering of electrons with impurities and phonons the self-generated current is chaotic [64,65,67], while for a strong scattering it is quantized [63,64,66]. For further development of the wave-mixing theory in a pure ac-driven SSL see recent work [71].

Another interesting theoretical direction is an investigation of symmetries and the symmetry breaking for a particle moving in a spatially periodic potential under an action of a time-periodic force [72–75]. In particular, the direct current may arise at the harmonic-mixing even if an underlying motion of the particle in a cosine-like spatial potential is chaotic (dynamical chaos) [72,74]. However, in semiconductor microstructures, as far as we aware, this interesting theoretical suggestion has not been realized, yet.

IV. CONCLUSION

In summary, the results of papers [6,5], together with a consideration based on the theory of quantum ratchets [58], demonstrate that in semiconductor superlattices the effect of a dc current (voltage) generation due to a mixing waves associated with different commensurate harmonics is now well established theoretically. The source of the non-ohmicity in current-voltage characteristics of the strongly-coupled SSLs may be associated with many factors. This may primarily be related to a nonparabolic band energy-momentum relation. This type of nonlinearity exists in any semiconductor or in any semiconductor device and does not depend on the fact if it is subjected by an intense high-frequency electric field or not.

We believe that this paper attracts attention of wide scientific community to these old and at the same time novel effects, will stimulate new experiments and a construction of new devices associated with a rectification and a detection of the THz radiation. The experimental conditions for an observation of the dc current effect are practically identical to those fulfilled in a recent experiment on a generation of harmonics of the THz radiation in a semiconductor superlattice [9].

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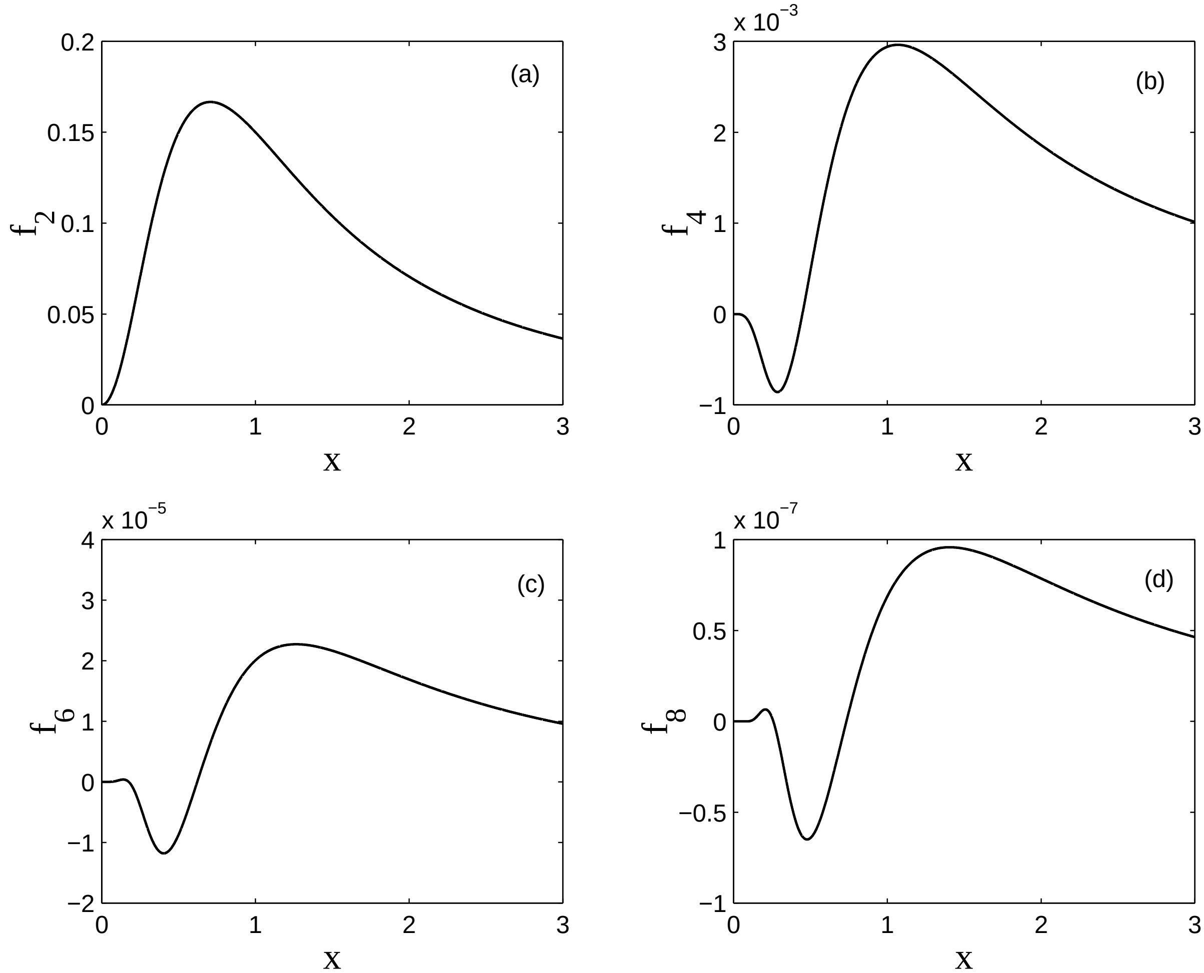

FIG. 1. The function $f_n(x)$ for $n = 2$ (a), $n = 4$ (b), $n = 6$ (c), and $n = 8$ (d).