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Green’s and spectral functions of the small Fröhlich polaron

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According to recent Quantum Monte Carlo simulations the small polaron theory is practically exact in a wide range of the long-range (Fröhlich) electron-phonon coupling and adiabatic ratio. We apply the Lang-Firsov transformation to convert the strong-coupling term in the Hamiltonian into the form of an effective hopping integral and derive the single-particle Green’s function describing propagation of the small Fröhlich polaron. One and two dimensional spectral functions are studied by expanding the Green’s function perturbatively. Numerical calculations of the spectral functions are produced. Remarkably, the coherent spectral weight (Z) and effective mass (Z’) renormalisation exponents are found to be different with Z’ >> Z, which can explain a small coherent spectral weight and a relatively moderate mass enhancement in oxides.

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1. Introduction

The problem of a fermion on a lattice coupled with the bosonic field of lattice vibrations has an exact solution in terms of the coherent (Glauber) states in the extreme strong-coupling limit, λ = ∞ for any type of electron-phonon interaction conserving the on-site occupation numbers of fermions. For the intermediate coupling the 1/λ perturbation diagrammatic technique has been developed both for a single- and multi-polaron systems. The expansion parameter actually is 1/2 z λ/∞, so the analytical perturbation theory might have a wider region of applicability than one can expect from a naive variational estimate (z is the lattice coordination number). However, it is not clear how fast the expansion converges. The exact numerical diagonalisation of vibrating clusters, variational calculations, dynamical mean-field approach in infinite dimensions, and Quantum-Monte-Carlo simulations have revealed that the ground state energy (the polaron binding energy E_p) is not very sensitive to the parameters. On the contrary, the effective mass, the bandwidth and the shape of polaron density of states strongly depend on polaron size and adiabatic ratio in case of a short-range (Holstein) interaction. In particular, numerical diagonalisation of the two-site-one-electron Holstein model in the adiabatic ω_0/t < 1 as well as in the nonadiabatic ω_0/t > 1 regimes shows that perturbation theory is almost exact in the nonadiabatic regime for all values of the coupling constant. However, there is no agreement in the adiabatic region, where the first order perturbation expression overestimates the polaron mass by a few orders of magnitude in the intermediate coupling regime. Here ω_0 is the characteristic phonon frequency and t is the nearest-neighbour hopping integral so that the dimensionless coupling constant is λ = E_p/(zt). A much lower effective mass of the adiabatic small polaron in the intermediate coupling regime compared with that estimated by first order perturbation theory is a result of poor convergence of the perturbation expansion owing to the appearance of the familiar double-well potential in the adiabatic limit. The tunnelling probability is extremely sensitive to the shape of this potential.

It has also been understood[16] that the range of applicability of the analytical theory[15] strongly depends on the radius of interaction. While the analytical approach is applicable only if ω_0 > t for short-range interaction, the theory appears almost exact in a substantially wider region of the parameters for a long-range (Fröhlich) interaction. The exact effective mass of both small and large Fröhlich polarons, calculated with the continuous-time path-integral Quantum Monte Carlo (QMC) algorithm, m*(λ) is well fitted by a single exponent[17]. As an example, e^{0.73λ} for ω_0 = t and e^{1.44λ} for ω_0 = 0.5t describe m*(λ) in a one-dimensional lattice. The numerical exponents are remarkably close to those obtained from the Lang-Firsov transformation, e^{0.78λ} and e^{1.56λ}, respectively. Hence, in the case of the Fröhlich interaction the transformation is perfectly accurate already in the first order of 1/λ expansion even in the adiabatic regime, ω_0/t ≤ 1 for any coupling strength.

In this paper we use this result to calculate the single-particle Green’s function of a fermion on a lattice coupled with the bosonic field via the long-range Fröhlich interaction.

2. Green’s functions of the small Fröhlich polaron

The classical approach to the small polaron problem is based on the canonical displacement (Lang-Firsov) transformation of the electron-phonon Hamiltonian, allowing for the summation of all diagrams including the vertex corrections,

\[ H = \sum_{i,j} t(m - n) c_i^d c_j + \sum_q \omega_q \hat{n}_i [u_i(q)d_q + H.c.] + \sum_q \omega_q (d_q^d d_q + 1/2) \tag{1} \]

with the bare hopping integral t(m) and the matrix element of the electron-phonon interaction

\[ u_i(q) = \frac{1}{\sqrt{2N}} \gamma(q) e^{iqm}. \tag{2} \]

Here i = (m, s), j = (n, s'), include site m and spin s, \( \hat{n}_i = c_i^d c_i \), and c_i, d_q are electron (hole) and phonon operators, respectively.
The canonical transformation $e^S$ diagonalising the Hamiltonian in the $\lambda = \infty$ limit is

$$\tilde{H} = e^S H e^{-S},$$

where

$$S = \sum_{\mathbf{q},i} \hat{n}_i [u_i(\mathbf{q}) d_{\mathbf{q}} - H.c.].$$

The electron operator transforms as

$$\tilde{c}_i = c_i e^{\gamma(\mathbf{q})} \exp \left( -\sum_{\mathbf{q}} u_i(\mathbf{q}) d_{\mathbf{q}} - H.c. \right)$$

and the phonon one as:

$$\tilde{d}_\mathbf{q} = d_{\mathbf{q}} + \sum_i \hat{n}_i u_i^*(\mathbf{q})$$

It follows from Eq.(6) that the Lang-Firsov transformation shifts ions to new equilibrium positions. In a more general sense it changes the boson vacuum. As a result,

$$\tilde{H} = \sum_{i,j} \tilde{\sigma}_{ij} c_i^\dagger c_j - E_p \sum_i \hat{n}_i$$

$$+ \sum_{\mathbf{q}} \omega_{\mathbf{q}} (d_\mathbf{q}^\dagger d_{\mathbf{q}} + 1/2) + \frac{1}{2} \sum_{i\neq j} v_{ij} \hat{n}_i \hat{n}_j,$$

where

$$\tilde{\sigma}_{ij} = t(\mathbf{m} - \mathbf{n}) \delta_{s,s'} \exp \left( \sum_{\mathbf{q}} [u_i(\mathbf{q}) - u_j(\mathbf{q})] d_{\mathbf{q}} - H.c. \right)$$

is a renormalised hopping integral depending on the phonon variables,

$$E_p = \frac{1}{2N} \sum_{\mathbf{q}} |\gamma(\mathbf{q})|^2 \omega_{\mathbf{q}}$$

the polaron level shift (binding energy), and

$$v_{ij} = -\frac{1}{N} \sum_{\mathbf{q}} |\gamma(\mathbf{q})|^2 \omega_{\mathbf{q}} \cos[\mathbf{q} \cdot (\mathbf{m} - \mathbf{n})]$$

the attractive interaction of polarons owing to lattice deformation.

We consider a single-particle problem when the last term in Eq.(7) is absent. The QMC result allows us to average the first hopping term with respect to phonons no matter what the value of the Fröhlich interaction is. Then we can apply the canonical transformation together with the averaged Hamiltonian to derive the single-particle Green’s function (GF). The interaction is completely integrated out from the averaged Hamiltonian.

$$\tilde{H} = H_p + H_{ph}$$

where the 'free' polaron part is given by

$$H_p = \sum_{\mathbf{k}} \xi(\mathbf{k}) c_{\mathbf{k}}^\dagger c_{\mathbf{k}},$$

(from now we omit the spin index), and the free phonon part is

$$H_{ph} = \sum_{\mathbf{q}} \omega_{\mathbf{q}} (d_{\mathbf{q}}^\dagger d_{\mathbf{q}} + 1/2).$$

Here $\xi(\mathbf{k}) = Z'E(\mathbf{k}) - \mu$ is the renormalised polaron-band dispersion with the chemical potential $\mu$, which includes polaron binding energy $(-E_p)^{\frac{1}{2}}$. $E(\mathbf{k}) = \sum_m \xi(\mathbf{m}) \exp(-i\mathbf{k} \cdot \mathbf{m})$ is the bare dispersion in a rigid lattice, and the mass-renormalisation exponent is

$$Z' = \frac{\sum_m \xi(\mathbf{m}) \exp(-i\mathbf{k} \cdot \mathbf{m})}{\sum_m \xi(\mathbf{m}) \exp(-i\mathbf{k} \cdot \mathbf{m})}$$

with

$$g^2(\mathbf{m}) = \sum_{\mathbf{q}} |\gamma(\mathbf{q})|^2 [1 - \cos(\mathbf{q} \cdot \mathbf{a})].$$

Quite generally one finds $Z' = \exp(-\gamma E_p/\omega)$, where the numerical coefficient

$$\gamma = \sum_{\mathbf{q}} |\gamma(\mathbf{q})|^2 [1 - \cos(\mathbf{q} \cdot \mathbf{a})]/\sum_{\mathbf{q}} |\gamma(\mathbf{q})|^2,$$

might be as small as 0.24 and even smaller in the cuprates with nearest neighbour oxygen-oxygen distance less than the lattice constant, $\gamma \approx 0.24$.

Applying the Lang-Firsov canonical transformation the Fourier component of the retarded GF,

$$G_R(\mathbf{k}, \omega) = -i \sum_m \int_0^\infty \langle 0 | c_m(t) c_m^\dagger(0) | 0 \rangle dt$$

is expressed as a convolution of the Fourier components of the coherent retarded polaron GF, $G_p(\mathbf{m}, \omega)$, and the multiphonon correlation function $\sigma(\mathbf{n}, t)$:

$$G_R(\mathbf{k}, \omega) = \frac{1}{2\pi} \sum_{\mathbf{m}} e^{-i\mathbf{k} \cdot \mathbf{n}} \int_{-\infty}^\infty d\omega' G_p(\mathbf{m}, \omega') \sigma(\mathbf{n}, \omega - \omega').$$

Here

$$G_p(\mathbf{m}, \omega) = -i \int_0^\infty dt e^{i\omega t} \langle 0 | e^{iH_p t} c_0 e^{-iH_p t} c_m^\dagger(0) \rangle,$$

and

$$\sigma(\mathbf{m}, \omega) = \int_0^\infty dt e^{i\omega t} \langle 0 | e^{iH_{ph} t} X_0 e^{-iH_{ph} t} X_m^\dagger(0) \rangle.$$
3. Spectral functions of the small Fröhlich polaron

The Green’s function of a polaronic carrier, Eq.(22) comprises two different contributions. The first ($l = 0$) coherent $k$-dependent term arises from the polaron band tunneling,

$$G_R^{(0)}(k, \omega) = \frac{Z}{\omega - \xi(k) + i\delta}. \quad (26)$$

The spectral weight of the coherent part is strongly (exponentially) suppressed as $Z = \exp(-E_p/\omega_0)$ while the effective mass might only be slightly enhanced, $\xi_k = Z'E_k - \mu$, because $Z \ll Z' < 1$ in the case of the Fröhlich interaction.

The second incoherent phonon-assisted contribution with $l \geq 1$ describes the excitations accompanied by emission of phonons. We believe that this term is responsible for the background in optical conductivity and in photoemission spectra of cuprates and manganites. We notice that its spectral density spreads over a wide energy range of about twice the polaron level shift $E_p$. On the contrary the coherent term shows an angular dependence in the energy window of the order of the polaron bandwidth, $2Z'zt$. Interestingly, there is some $k$ dependence of the incoherent background as well, if the matrix element of electron-phonon interaction depends on $q$ (see also Ref.[2]). To illustrate this point we calculate the single-phonon contribution ($l = 1$) to the spectral function

$$A(k, \omega) \equiv -(1/\pi)\Im G_R(k, \omega) = \sum_{l=0}^{\infty} A_l(k, \omega). \quad (27)$$

The coherent part ($l = 0$) of the spectral function is a $\delta$-function in agreement with the well-established fact (see Ref.[2] and references therein) that small polarons exist in the Bloch states at zero temperature no matter which values the parameters of the system take. The single-phonon contribution to the incoherent background is given by

$$A^{(1)}(k, \omega) \sim \int_{BZ} dqq^{-2}\delta(\omega - \omega_0 - \xi(k + q)), \quad (28)$$

where the interval of integration (the Brillouin zone, BZ) is determined by lattice constants $a, b, c$. We calculate this integral for one-dimensional (1D), $\xi(k) = 2t\cos(k/a)$ and two-dimensional (2D), $\xi(k) = 2t\cos(k/a) + 2t'\cos(k/a)$ polaron bands in the tight-binding approximation with the (renormalised) nearest neighbour hopping integrals, $t = Z't, t' = Z't'$. The 1D single-phonon spectral function is reduced to

$$A^{(1)}(k, \omega) \sim (1-\omega^2)^{-1/2} \int_0^{\pi/3} dz f(z, \omega, k_x) + f(z, \omega, -k_x), \quad (29)$$

where

$$f(z, \omega, k_x) = \left\{ \begin{array}{ll} z^2 + (k_x - \cos^{-1}\omega_0)^2 \tan^{-1}\left( \frac{\pi}{z^2 + (k_x - \cos^{-1}\omega_0)^2} \right) & \text{for } 2t \ll \omega_0 \ll 2t' \\ 1 & \text{for } \omega_0 \ll 2t' \ll 2t \\ \end{array} \right. \quad (30)$$

Here and further we take $a = b = 1$ and $c = 3$, and $t' = t/4$. Its spectral and angular dependence are shown in Fig.1. Apart from two nondispersive 1D van-Hove singularities (vHs) at $\tilde{\omega} \equiv (\omega - \omega_0)/2t = \pm 1$ there is an interesting dispersive peak at $\tilde{\omega} = \cos k_x$, which is due entirely to the long-range character of the Fröhlich interaction. Indeed, approximating the Brillouin zone by a cylinder along $x$, one readily obtains a logarithmic singularity in the spectral function,

$$A^{(1)}(k, \omega) \sim (1-\tilde{\omega}^2)^{-1/2} \ln \left( \frac{(k_x - \cos^{-1}\tilde{\omega})^2 + q_0^2}{(k_x - \cos^{-1}\tilde{\omega})^2} \right). \quad (31)$$

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with $X_m = \exp\left(\sum_n u_i(q)\delta_q - H.c.\right)$, and $|\tilde{0}\rangle$ the ground state of $\tilde{H}$. Straightforward calculations yield

$$G_p(m, \omega) = \frac{N}{\omega - \xi(k')} + \frac{i\delta}{\omega - \xi(k')} \quad (21)$$

and

$$\sigma(m, \omega) = iZ \sum_{l=0}^{\infty} \frac{1}{\pi l(l+1)} \left( \frac{1}{2N} \sum_q |\gamma(q)|^2 \exp(iq \cdot m) \right)^l. \quad (22)$$

Here

$$Z = \exp(-\sum_q |\gamma(q)|^2). \quad (23)$$

In the following we consider dispersionless phonons, $\omega_q = \omega_0$, and the Fröhlich interaction with $\gamma(q) \sim 1/q$. The convolution of Eq.(19) and Eq.(20) yields

$$G_R(k, \omega) = \sum_{l=0}^{\infty} G_R^{(l)}(k, \omega), \quad (24)$$

where

$$G_R^{(l)}(k, \omega) = \frac{Z}{(2N)^l l!} \sum_{q_1,...,q_l} \frac{1}{\omega - \xi(k + q_1 + ... + q_l) + i\delta} \quad (25)$$

with $\delta = +0$.

Obviously, Eq.(22) is in the form of a perturbative multi-phonon expansion. A term with an index $l$ corresponds to a transition from the initial state $k$ of the polaron band to the final state $k + q_1 + ... + q_l$ with the emission of $l$ optical phonons.[2]
Here \( q_D \) is the radius of the cylinder. For \( \omega = \omega_0 + 2t \cos k_x \), the \( x \)-component of the phonon momentum is zero, and the singular matrix element squared (\( \sim 1/q^2 \)) integrated over \( q_y \) and \( q_z \) yields a singularity.

The 'long-range' dispersive features appear in the 2D single-phonon spectral function as well. This function is reduced to

\[
A^{(1)}(k, \omega) \sim \int_{a(\omega)}^{b(\omega)} dz \left[ 1 - \left( \bar{\omega} - \frac{1}{4} \cos z \right)^2 \right]^{-1/2} \\
\times \left[ \phi(z, \omega, k_x, k_y) + \phi(z, \omega, -k_x, -k_y) + \phi(z, \omega, -k_x, k_y) + \phi(z, \omega, k_x, -k_y) \right],
\]

with

\[
\phi(z, \omega, k_x, k_y) = \left[ (x - k_x)^2 + (z - k_y)^2 \right]^{-1/2} \\
\tan^{-1} \left( \frac{x}{y} \right) \left[ (x - k_x)^2 + (z - k_y)^2 \right]^{-1/2}
\]

and \( x = \cos^{-1}(\bar{\omega} - \frac{1}{4} \cos z) \). The integration limits are defined as \( a(\omega) = \cos^{-1}[4(\bar{\omega} + 1)] \) if \(-5/4 \leq \bar{\omega} \leq -3/4\), \( a(\omega) = 0 \) if \(-3/4 \leq \bar{\omega} \leq 5/4\), and \( b(\omega) = \cos^{-1}[4(\bar{\omega} - 1)] \) if \( 3/4 \leq \bar{\omega} \leq 5/4\). The numerical results are shown in Fig.2.

We do not expect any dispersive peaks in the multi-phonon \( (l \geq 2) \) contributions to the spectral function because of the additional integrations compared with the single-phonon term. However, the nondispersive vHs remains for all \( l \geq 1 \). They can be washed out by the phonon frequency dispersion in real crystals.

4. Conclusions

We have calculated the Green’s and spectral functions of a fermion on a lattice coupled with lattice vibrations via the long-range Fröhlich interaction. In a sharp contrast with the Holstein polaron the mass renormalisation exponent of Fröhlich polaron (\( Z' \)) differs from the renormalisation of the coherent spectral weight (\( Z \)). On the one hand this important result tells us that small polarons as well as intersite bipolarons are perfectly mobile and can account for the high-\( T_c \) values in cuprates. On the other hand the coherent spectral weight remains strongly suppressed in polaronic conductors, Eq.(24), because \( Z \) might be less than \( Z' \) by one or even a few orders of magnitude. These unusual spectral features provide an explanation for the apparent discrepancy between a very small Drude weight and a relatively moderate mass enhancement, \( m^* \sim 3m_c/10m_c \) (depending on doping) of carriers in manganites and cuprates. They also explain why the ‘extended’ vHs observed in angle-resolved photoemission can be hardly seen in angle-averaged photoemission. Indeed, the integrated spectral weight of the incoherent background is proportional to \( (1 - Z) \). It turns out to be much larger than the coherent contribution, proportional to \( Z << 1 \). Finally, the \( k \)-dependent incoherent background, Fig.1,2, might obscure the experimental determination of a Fermi-level crossing. We believe that our results can provide a quantitative approach to the experimental tunneling and photoemission spectra. In particular the coherent part of ARPES and tunneling in cuprates has been recently explained.

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24 Higher order corrections in $1/\lambda$, which do not depend on $k$ are also included in $\mu$.
27 Eq.(22) can be also derived from the polaronic Matsubara GF.

**Figure Captures**

Fig.1. 1D single-phonon contribution to the polaron spectral function.

Fig.2. 2D single-phonon contribution to the polaron spectral function along the $\Gamma$ – $Y$ direction.
1-D Spectral Fn. (arbitrary units)