

Optical absorption from a nondegenerate polaron gas

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We apply the dynamical mean field theory to calculate the low-density limit of the optical conductivity in the Holstein model. This nonperturbative treatment allows continuous spanning from the weak-coupling quasi-free-electron behavior to the strong-coupling small polaron picture. Different features arise in the intermediate coupling regime: (i) a noticeable transfer of spectral weight toward low frequencies occurs at low temperatures, when coherent processes acquire importance; (ii) thermally activated polaron interband transitions appear at nonzero temperature, whose spectral weight has a nonmonotonic temperature dependence. We propose an interpretation of the infrared active vibrations observed in the optical spectra of underdoped superconducting cuprates in terms of small polarons in the intermediate coupling regime.

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The calculation of the optical conductivity of small polarons is an important and unsolved problem in solid state physics. Although many approaches exist that are appropriate in specific limiting cases, an analytical treatment able to give the correct physical results in all the regimes of parameters is still lacking. The simplest model that describes small polarons is the Holstein molecular crystal model,¹ where the carriers interact with phonon modes localized on each lattice cell. The Holstein model was recently solved² in the single-electron case by dynamical mean field theory (DMFT),³ which becomes exact in the limit of infinite lattice coordination. When applied to real lattices, this approach can be viewed as a valid interpolation scheme, provided that an appropriate finite dimensional density of states (DOS) is chosen.^{4,5} The solution reported in Ref. 2 is essentially analytical, and it overcomes the well-known difficulties inherent to numerical methods.

The aim of this work is to generalize the exact DMFT solution of the Holstein model to the calculation of the optical conductivity. We shall focus on the most interesting *intermediate coupling regime* near the adiabatic limit, where unusual features have been found in the single-particle propagator, so that interesting properties can be expected in the optical spectra. Unlike the usual strong-coupling situation,⁶ where the polaron absorption is concentrated in a "band" located at frequencies much higher than the phonon frequency, we will show that at intermediate coupling strengths narrow absorption peaks appear close to the phonon frequencies, which are due to transitions between different subbands in the polaron excitation spectrum. The presence of such low-frequency features is a very general property of finite band electron-phonon (*e-ph*) systems in the intermediate coupling regime, which could help in interpreting the far infrared spectra of the superconducting cuprates at low doping levels, as discussed in the conclusion.

In the Holstein Hamiltonian,¹ tight-binding electrons with hopping amplitude t are coupled locally to Einstein phonons with energy ω_0 .⁷

$$H = \omega_0 \sum_i a_i^\dagger a_i - g \sum_{i,\sigma} c_{i,\sigma}^\dagger c_{i,\sigma} (a_i^\dagger + a_i) - \frac{t}{2\sqrt{z}} \sum_{i,j,\sigma} (c_{i,\sigma}^\dagger c_{j,\sigma} + \text{H.c.}) \quad (1)$$

At zero temperature, a crossover occurs from a quasi-free-electron to a small polaron as the *e-ph* coupling constant g increases. This crossover depends strongly on the adiabaticity parameter ω_0/t .^{4,8} If we restrict consideration to the nearly adiabatic regime ($\omega_0/t < 1$), the crossover takes place when the polaron energy $E_p = g^2/\omega_0$ is of the order of the free electron bandwidth, i.e., for $\lambda = g^2/\omega_0 t \sim 1$. Although these arguments are based on the ground-state properties alone, it was pointed out in Ref. 2 that the mechanism of polaron formation can be better understood by analyzing the whole excitation spectrum, as deduced from knowledge of the interaction propagator G . It was shown that the polaron crossover is characterized by the successive opening of energy gaps in the low-energy part of the spectral density as the *e-ph* coupling is increased. At intermediate values of λ , although the polaron mass is still close to the free electron value, the spectral density starts to exhibit typical polaronic features, i.e., narrow subbands at low energies, together with a broad contribution at higher energies. The width W of the first subband, which gives a measure of the coherent (band-like) behavior of the polaron, is rapidly reduced as λ moves toward the strong-coupling limit. This picture of the polaron crossover has important consequences on the optical properties.

The calculation of the optical conductivity within the DMFT is simplified since, for symmetry reasons and due to the locality of vertices, no current vertex corrections enter in the Kubo formula.^{9,10} Writing the electron spectral function as

$$\rho_\epsilon(\nu) = -\frac{1}{\pi} \text{Im} \frac{1}{\nu + i0^+ - \epsilon - \Sigma(\nu)}, \quad (2)$$

the optical conductivity per particle takes the form

$$\sigma(\omega) = \frac{\zeta\pi}{n\omega} \int d\epsilon N_\epsilon \phi_\epsilon \int d\nu (f_\nu - f_{\omega+\nu}) \rho_\epsilon(\nu) \rho_\epsilon(\nu+\omega), \quad (3)$$

where $N_\epsilon = (2/\pi) \sqrt{t^2 - \epsilon^2}$ is the free DOS of the real lattice, $\phi_\epsilon = (t^2 - \epsilon^2)/3$ is the corresponding current vertex¹¹ (which ensures a sum rule for the total spectral weight¹²), f is the Fermi function, n is the particle density, and ζ is a numerical constant that we shall drop in the following discussion.¹³ Taking the low-density limit ($n \rightarrow 0$) at finite temperatures, following the procedure described in Ref. 14, Eq. (3) becomes

$$\sigma(\omega) = \frac{\pi}{\omega} (1 - e^{-\beta\omega}) \frac{\mathcal{D}(\omega, \beta)}{\mathcal{N}(\beta)}, \quad (4)$$

where we have defined

$$\mathcal{D}(\omega, \beta) = \int d\epsilon N_\epsilon \phi_\epsilon \int d\nu e^{-\beta(\nu - E_0)} \rho_\epsilon^{(0)}(\nu) \rho_\epsilon^{(0)}(\nu + \omega), \quad (5)$$

$$\mathcal{N}(\beta) = \int d\epsilon N_\epsilon \int d\nu e^{-\beta(\nu - E_0)} \rho_\epsilon^{(0)}(\nu), \quad (6)$$

and $\rho^{(0)}$ is the spectral function at zero density (these quantities have been rescaled by $e^{\beta E_0}$ to obtain a finite result in the low-temperature limit). The integrals in ϵ can be performed analytically, while the remaining integrals in ν are computed numerically, once the self-energy Σ is obtained as a continued fraction expansion by applying the iteration scheme reported in Ref. 2.

Our treatment applies to a dilute *nondegenerate* polaron gas, where exchange effects can be neglected. Turning to real systems, we expect that our results will be relevant at low enough density, i.e., when the Fermi temperature is negligible compared to the important temperature scales of the problem (e.g., the temperature $\hbar\omega_0/k_B$, below which phonon quantum effects become important⁶).

Figure 1 shows the results obtained at $\omega_0/t = 0.1$ and $\lambda = 0.9$, i.e., close to the polaron crossover.¹⁵ The optical response shows a typical pattern of subbands roughly spaced by ω_0 , which tends to become smooth at higher frequencies. This behavior is a characteristic signature of the polaron crossover.² At temperatures well above ω_0 , the results are in good agreement with Reik's formula,⁶ showing that the physics in this limit is dominated by activated hopping processes. As the temperature is decreased, a substantial transfer of spectral weight takes place, the low-frequency features becoming more pronounced.

To get some insight into this peculiar behavior, it is convenient to analyze the polaron excitation spectrum by explicitly separating the contributions $\rho^>$ and $\rho^<$ at energies respectively above and below E_0 . At low temperatures, $\rho^>$ has quasiparticle poles at energies below $E_0 + \omega_0$ plus a more or less incoherent background due to phonon scattering at higher energies. On the other hand, $\rho^<$ represents incoherent states where one or more phonons are taken from the thermal

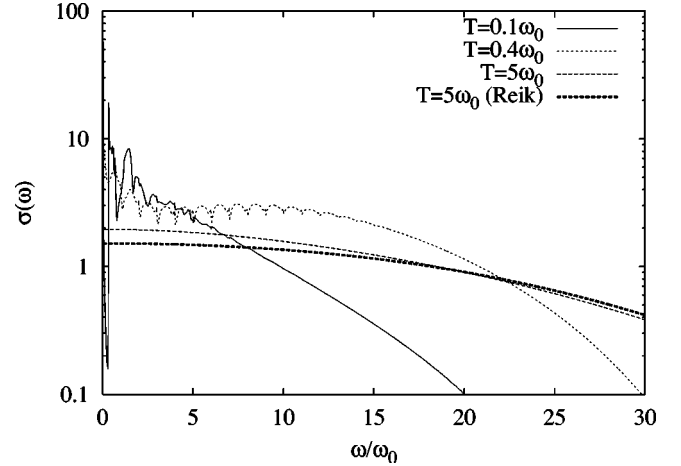


FIG. 1. Optical conductivity per particle at $\lambda = 0.9$ and $\omega_0/t = 0.1$, at different temperatures. Reik's result is shown at $T = 5\omega_0$ for comparison. Note the transfer of spectral weight occurring at low T (for the definition of T_W , see text). The curve marked $T \ll T_W$ was obtained with Eq. (7).

bath. Correspondingly, by looking at the definitions (5) and (6), one can define $\mathcal{N} = \mathcal{N}^> + \mathcal{N}^<$ and $\mathcal{D} = \mathcal{D}^{>>} + \mathcal{D}^{<>} + \mathcal{D}^{<<}$. The term $\mathcal{D}^{>>}$ has contributions associated with both coherent band motion and phonon emission processes, while $\mathcal{D}^{<>}$ and $\mathcal{D}^{<<}$ account for the thermally activated hopping.

An estimate for the temperature T_W below which coherent processes acquire importance can be obtained by analyzing the relative weights of $\mathcal{N}^>$ and $\mathcal{N}^<$. Due to the exponential factor in Eq. (6), we see that at low temperatures $\mathcal{N}^>$ weights the coherent states close to E_0 . Using the semicircular free DOS, we obtain $\mathcal{N}^> = \sqrt{2(m^*/m)/\pi} (T/t)^{3/2}$, where m^*/m is the ratio of the polaron mass with the free electron value. On the other hand, being associated with incoherent states, the spectral function $\rho_\epsilon^<(\nu)$ is essentially ϵ independent and peaked around a discrete set of energies $\nu = E_0 + W - p\omega_0$, with a weight proportional to the thermal occupation factor¹⁶ $e^{-p\beta\omega_0}$ (recall that W is the width of the first subband in the spectral density, and $p = 1, 2, \dots$ is the number of thermal phonons). When evaluating the integral in $\mathcal{N}^<$, such vanishing occupation factors are partly compensated by the exponential $e^{-\beta(\nu - E_0)}$, which gives rise to an overall behavior $\mathcal{N}^< \propto e^{-\beta W}$.

We find that the crossover between the two regimes occurs at $T_W \sim W/\ln(t/W)$. This is not surprising, since, as was noted in the introduction, W is the typical energy scale for coherent excitations (in the particular case studied here, where $m^*/m \approx 1.5$ and $W = 0.7\omega_0$, we get $T_W \approx 0.1\omega_0$). For $T \ll T_W$, only the terms $\mathcal{D}^{>>}$ and $\mathcal{N}^>$ survive in formula (4), which becomes

$$\sigma(\omega) = \pi t \frac{m^*}{m} \left[\frac{1 - e^{-\omega/T}}{\omega/T} \right] \rho_{q=0}(\omega + E_0) \quad (7)$$

where the spectral function ρ is evaluated at $\epsilon = \epsilon(q = 0)$. In this regime, the conductivity scales as T/ω for any $\omega > T$, so that most of the spectral weight is transferred to the Drude peak $\sigma(\omega) \sim \delta(\omega)$.

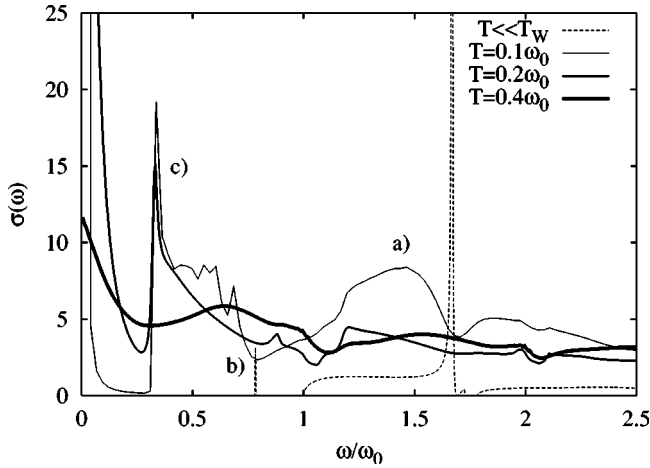


FIG. 2. The same as Fig. 1 but in the region close to ω_0 and with a finer temperature scan. The curve $T \leq T_W$ corresponds to $T = 0.025\omega_0$ (the Drude peak is not shown).

At higher temperatures, the main contribution to the optical absorption involves phonons that are already present in the thermal bath, so that one expects some spectral weight to move toward higher frequencies as the temperature is raised above T_W . This is indeed the case, as can be seen in Fig. 1. Let us stress that this crossover from *coherent* to *activated* conductivity is a signature of the intermediate coupling regime. In the strong-coupling limit, where $W \rightarrow 0$, the crossover temperature vanishes and phonon-assisted contributions always dominate.

In Fig. 2 we report the optical conductivity in the frequency range around ω_0 , at temperatures close to T_W . Despite its apparent complexity, the observed pattern can be qualitatively understood by looking at the excitation spectrum close to the ground state, which is sketched in Fig. 3 versus the free particle energy ϵ [see Eq. (2)]. The dark curves are the quasiparticle states in the lowest-order polaron subbands close to the ground state, while the region above ω_0 represents the incoherent background due to scattering from Einstein phonons (the states below E_0 , which give rise to the hoppinglike behavior at $T \gg T_W$, are not shown in Fig.

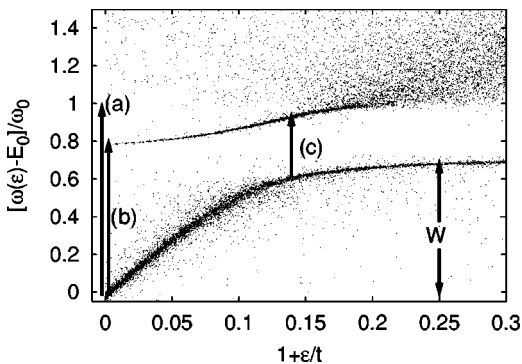


FIG. 3. The dispersion of the lowest-order polaronic subbands versus the free particle energy ϵ (parameters are the same as in the preceding figures, and $T = 0.2\omega_0$). The points are distributed according to the spectral function $\rho_\epsilon(\nu)$.

3). The transitions from the ground state to the incoherent background, denoted by (a), give rise to a continuum of absorption above ω_0 . A transition from the ground state to the bottom of the second subband is also possible (b), but this is hardly visible in Fig. 2 owing to its negligible spectral weight. In addition, the Drude peak corresponding to coherent band motion is clearly visible in Fig. 2.

The most striking feature in Fig. 2 is probably the appearance of a resonance *below* ω_0 (c), which is due to *polaron interband transitions* (PIT's), as can be seen in Fig. 3. Indeed, because of thermal activation, long-lived states at energy $E_0 < \nu < E_0 + \omega_0$ can be occupied with a probability $e^{-\beta(\nu - E_0)}$. According to the Fermi golden rule, the transition probability at a given frequency ω is proportional to the joint density of the initial and final states separated by ω . If we focus on transitions between different subbands, this function has a Van Hove singularity at that value ω^* where the band dispersions are parallel, which identifies an *interband threshold* [see arrow (c)]. This resonance has a nonmonotonic temperature dependence, which can be understood as follows: the occupation probability of the initial state gives rise to an activated behavior at low T ; as the temperature is raised above ω^* , the difference in population of the initial and final states is balanced, which depresses the optical absorption according to the prefactor $\sim (1 - e^{-\beta\omega^*})$ in formula (4). According to the discussion above, we argue that PIT's are not peculiar to the Holstein model, nor are they a consequence of the approximations used. Rather, they are closely related to the presence of several subbands in the spectral density at low energy, which should be a common feature of any *finite band* model in the intermediate e -ph coupling regime. Such interband transitions are obviously absent at weak coupling, where there are no subbands in the excitation spectrum, while in strong coupling they can hardly be resolved from the incoherent multiphonon absorption, due to their relatively small spectral weight. On the other hand, the results are not qualitatively affected by variations of the adiabaticity parameter, provided that $\omega_0/t < 1^2$.

In this work, we have analyzed the optical conductivity of a dilute gas of small polarons in the framework of the DMFT, focusing on the crossover regime between quasi-free-electrons and small polarons which takes place at intermediate values of the e -ph coupling strength. The spectra at high temperatures do not differ significantly from the classical picture of hoppinglike conductivity, which predicts a featureless absorption band at finite frequencies. However, interesting results appear at low temperatures, where the physics is governed by two different energy scales. At temperatures below ω_0 , phonon *quantum* effects become important, and the optical spectra start to exhibit narrow peaks at low frequencies, together with a broad background at higher frequencies. Upon further decreasing the temperature, a noticeable transfer of spectral weight takes place toward low frequencies. This occurs at a temperature T_W that is related to W , the width of the first polaron subband in the spectral density, i.e., the typical energy scale governing *coherent* behavior.

Our results in the low-frequency region (see Fig. 2) bear a strong resemblance to the far infrared spectra of both hole- and electron-doped cuprates at low doping levels, where narrow peaks are observed at typical phonon frequencies, usually called infrared active vibrations (IRAV's).^{17,18} From a theoretical point of view, if we focus on such low-energy features, i.e., much lower than the typical energies of electronic correlations (e.g., J in the t - J model), the inclusion of correlation effects would only enter through a renormalization of the hopping parameter. Therefore, we expect that a description in terms of the Holstein model with renormalized parameters should correctly account for all the observed features in the frequency range of interest.¹⁹ On the other hand, the present single-polaron treatment, which neglects polaron-polaron interactions, is appropriate at low doping levels, i.e., precisely where the IRAV's are best resolved.

In the measured optical spectra, the IRAV peaks appear as

shoulders on both sides of the main phonon lines, and they have been ascribed to the lowest-order transitions inside the polaron potential well.²⁰ We have shown here that in the intermediate e -ph coupling regime the polaron absorption exhibits narrow peaks located above and below the phonon frequency,²¹ whose T dependence is nonmonotonic. These peaks come from two different processes: transitions to incoherent scattering states [denoted by (a) in Fig. 3], and thermally activated polaron interband transitions [denoted by (c)]. Although indications of a nonmonotonic behavior of some IRAV's already exist,^{18,22} a more detailed experimental analysis of their temperature dependence is needed to confirm these ideas.

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