Journal of Physics and Chemistry of Solids 69 (2008) 2164–2167

Contents lists available at ScienceDirect

Journal of Physics and Chemistry of Solids

journal homepage: www.elsevier.com/locate/jpcs

Optical properties of lattice/magnetic small polarons from DMFT

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ARTICLE INFO

PACS:
71.10.Fd
71.38.Ht
78.20.Bh
75.30.Ds

Keywords:
A. Magnetic materials
A. Super conductors
D. Optical properties

ABSTRACT

We calculate the optical conductivity of small polarons in the Holstein and Holstein-*t*-*J* models, by applying the dynamical mean field theory. We show that the antiferromagnetic correlations tend to increase the region of the parameters where polaronic signatures are found in the optical spectra, and shift the polaronic absorption band to higher frequencies compared to the case of purely lattice polarons. On the other hand, the electron–lattice interaction is essential in order to have polaronic features in the optical absorption.

1. Introduction

In systems with strong electron–phonon (e–ph) interactions, the motion of the carriers is significantly reduced by the formation of polarons. A polaron is a state in which the phonon and electron degrees of freedom are tightly entangled: the presence of an electron is associated to a finite lattice distortion, which in turn binds the electron, leading to the so-called *self-trapping* effect. Magnetic interactions, such as those acting on an hole moving in antiferromagnetic (AF) background, also tend to localize the charges. In this case the localizing effect is due to the energy cost that the moving hole has to pay as it modifies the local magnetic environment. In transition metal oxides, these two mechanisms often coexist, leading to an interesting interplay which will be the subject of the present study.

Typical signatures of polarons are seen in photoemission spectra [1]—where the lattice degrees of freedom can show up as characteristic multi-peaked structures—and transport measurements—where a thermally activated behavior is often observed at room temperature [2,3]. Optical absorption measurements [4,5] may also detect a polaronic band at a frequency related to the polaron binding energy [6] as well as polaronic interband transitions in the range of the phonon frequencies [7]. Another less classical indication of polaronic formation comes from the analysis of lattice displacements associated to the excess charge as obtained by the distribution of distances between atoms [8].

In studies of polaron formation, the difficulty consists in describing the dressing of the moving electron by a cloud of phonons, in a complex state that can coherently move as a quasiparticle [9]. Here we use dynamical mean field theory (DMFT), a non-perturbative technique originally developed as the exact solution of a interacting electron problem on an infinite dimensional lattice [10,11]. In its original formulation, DMFT solves a general tight-binding problem in the presence of local interactions by mapping it onto an effective local model. The latter is embedded in a quantum medium, whose properties are determined self-consistently.

The results for the optical absorption of small lattice polarons are presented in Section 2. These are generalized in Section 3 to include an additional coupling with the magnetic background. The main findings are briefly summarized in Section 4.

2. Optical conductivity for Holstein polarons

As a paradigmatic model for the study of small lattice polarons we choose the Holstein molecular crystal model whose Hamiltonian reads

$$H = \omega_0 \sum_i a_i^+ a_i - g \sum_{i,\sigma} c_i^+ c_i (a_i^+ + a_i) - t \sum_{i,j} (c_i^+ c_j + \text{H.c.}), \qquad (1)$$

where tight-binding electrons $(c_i, c_i^\dagger)$ with hopping amplitude t are coupled locally to Einstein bosons $(a_i, a_i^\dagger)$ with energy ω_0. We choose as dimensionless parameters of the model the e–ph coupling constant $\lambda = g^2/\omega_0 D$, where D is the half-bandwidth of our lattice, and the adiabatic ratio $\gamma = \omega_0/D$.

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doi:10.1016/j.jpcs.2008.03.023

The single electron case can be solved semi-analytically within DMFT at any temperature [9]. The solution is given in terms of the local Green function $G(\omega)$ [or equivalently a local self-energy $\Sigma(\omega)$] in the form of a continued fraction expansion [9,12]. The self-energy enters in the single particle spectral function

$$\rho(\varepsilon, \nu) = -\frac{1}{\pi}\mathrm{Im}\frac{1}{\nu - \Sigma(\nu) - \varepsilon}. \quad (2)$$

Its integral over the band dispersion ε equals the local Green's function $G(\omega)$, which provides the self-consistency equation that closes the DMFT loop.

The conductivity at finite frequency is related to the current–current correlation function through the appropriate Kubo formula. In DMFT, due to the absence of vertex corrections [13,14], the optical conductivity per particle can be expressed in the low density limit as

$$\sigma(\omega) = \frac{\zeta\pi}{\omega}(1 - \mathrm{e}^{-\beta\omega})\int \mathrm{d}\varepsilon N(\varepsilon)\phi(\varepsilon)\int \mathrm{d}\nu\, \mathrm{e}^{-\beta\nu}\rho^{\mathrm{w}}(\varepsilon, \nu)\rho(\varepsilon, \nu + \omega), \quad (3)$$

where $\beta = 1/T$ is the inverse temperature, and the constant $\zeta = \mathrm{e}^2a^2/\hbar v$, that we shall omit in the following, carries the appropriate dimensions of conductivity (a being the lattice spacing, v the volume of the unit cell). In Eq. (3)

$$\rho^{\mathrm{w}}(\varepsilon, \omega) = \frac{\mathrm{e}^{-\beta\omega}\rho(\varepsilon, \omega)}{\int \mathrm{d}\varepsilon N(\varepsilon)\int \mathrm{d}\nu\, \mathrm{e}^{-\beta\nu}\rho(\varepsilon, \nu)} \quad (4)$$

defines the "weighted spectral function", which carries information on thermally excited states.

The density of states $N(\varepsilon)$ of the unperturbed lattice is assumed semi-elliptical of half-bandwidth D, namely $N(\varepsilon) = (2/\pi D^2)\sqrt{D^2 - \varepsilon^2}$. In Eq. (3), $\phi(\varepsilon) = (D^2 - \varepsilon^2)/3$ is the current vertex function [15–17]. With this choice for $N(\varepsilon)$ and $\phi(\varepsilon)$, the ε-integral can be performed analytically, leaving only one integral to be computed numerically [15].

Let us focus on the *adiabatic* regime $\gamma \ll 1$, where the polaron crossover is very sharp (it becomes a true localization transition at $\gamma = 0$) [18]. A key parameter which controls the optical properties in this regime is the variance of the phonon quantum fluctuations $s = \lambda D\omega_0 \coth \omega_0/2T$ [19]. In the strong coupling regime, the photoexcitation of the electron is much faster than the lattice dynamics, which is virtually frozen during the absorption process. Since the lattice energy cannot be relaxed, the dominant optical transition corresponds to the difference in *electronic* energy between the initial and final states (Franck–Condon principle) which, in the Holstein model, equals twice the ground state energy $E_P = \lambda D$. The shape of the optical absorption will depend on the ratio between the width of the non-interacting band $\sim D$, and the variance s of the phonon field.

When $s \gg D$, i.e. when the phonon-induced broadening of electronic levels is much larger than the electronic dispersion, the absorption by localized polarons takes the form of a skewed Gaussian peak centered at $\omega_{max} = 2E_P$ [20,21]

$$\sigma(\omega) = \frac{\pi}{\omega}\frac{D^2}{4}\frac{1 - \mathrm{e}^{-\omega/T}}{\sqrt{4\pi s^2}}\exp\left[-\frac{(\omega - 2E_P)^2}{4s^2}\right]. \quad (5)$$

In the opposite limit $s \ll D$, the lineshape is dominated by the electronic dispersion. The absorption is due to transitions from a polaronic state whose electronic energy is $\simeq -2E_P$ to the continuum of free-electron states [22,23]. From Eq. (3) we get in this limit [15]

$$\sigma(\omega) = \frac{4\pi E_P^2}{\omega^2}\frac{1 - \mathrm{e}^{-\omega/T}}{\omega}\phi(\omega - 2E_P)N(\omega - 2E_P). \quad (6)$$

We see that in this case the absorption of photons occurs as a threshold phenomenon above $2E_P - D$ and vanishes above $2E_P + D$. The lineshape strongly differs from Eq. (5) and is rather similar to the weak coupling case (the absorption vanishing as a power law at the edges). For the above semi-elliptical DOS, which can be taken as representative of a three-dimensional lattice, we find that the absorption maximum $\omega_{max} = 2E_P - D^2/2E_P$ is shifted to lower frequencies compared to the usual estimate $2E_P$. This softening is entirely due to finite bandwidth effects. Note that Eq. (6) is valid at all temperatures below the polaron dissociation temperature $T \sim E_P$. In particular, contrary to Eq. (5), nothing happens here at temperatures $T \sim \omega_0/2$, provided that the condition $s \ll D$ is not violated (in other words, a temperature independent sub-threshold absorption is not necessarily in contradiction with polaron formation [24]).

Fig. 1 shows the DMFT results at $\gamma = 0.1$ and $\lambda = 1.1$. This value of the coupling strength lies in the polaronic regime, in a region where the electronic dispersion and the phonon-induced broadening are comparable ($s/D = 0.33$), so that none of Eqs. (5) and (6) is expected to hold. DMFT results shows a qualitative difference between low ($\omega < 2E_P$) and high ($\omega > 2E_P$) frequency regions: characteristic phonon resonances are evident at low frequencies, that merge in a broad continuum at higher frequencies [9,15]. This behavior, obtained in DMFT, cannot be reproduced by the usual approximate formulas, which either predict a smooth continuum or a multi-peaked spectrum depending on their range of validity [21].

Concerning the overall shape of the optical conductivity, the DMFT spectrum at $s/D = 0.33$ is intermediate between the two asymptotic formulas above. It is softer, broader and more asymmetric than Eq. (5) (full line). In fact, the position of the absorption maximum agrees better with Eq. (6) (dashed line). However, the peak height is much reduced compared to Eq. (6) and the absorption edge is completely washed out by phonon fluctuations.

Our results show that detectable deviations from the commonly used Eq. (5) arise as soon as the non-interacting bandwidth is larger than the broadening s, a condition that is often realized in real systems. For example, taking typical values $\omega_0 \simeq 0.01-0.05\,\mathrm{eV}$ and $E_p \simeq 0.1-0.5\,\mathrm{eV}$ yields a zero temperature broadening $s \simeq 0.03-0.16\,\mathrm{eV}$, in which case electron bandwidths of few tenths of meV are already sufficient to invalidate the standard Gaussian lineshape equation (5).

3. Optical conductivity for Holstein-*t*-*J* polarons

We consider the case of a single hole in an AF background, interacting with local Holstein phonons. Using the linear spin-

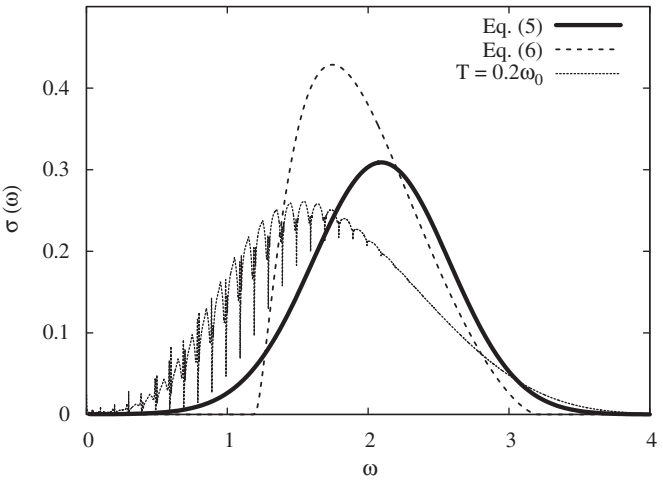

Fig. 1. Optical conductivity for $\lambda = 1.1, \gamma = 0.1$ and $T/\omega_0 = 0.2$ (dashed lines) compared with $T = 0$ predictions of Eq. (5) (full line) and Eq. (6) (bold dashed line).