

## Spectral properties and infrared absorption in manganites

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Within a recently proposed variational approach it has been shown that, in  $\text{La}_{1-x}\text{A}_x\text{MnO}_3$  perovskites with  $0 < x < 0.5$ , near the metal-insulator transition, the combined effect of the magnetic and electron-phonon interactions pushes the system toward a regime of two coexisting phases: a low electron density one made by itinerant large polarons forming ferromagnetic domains and a high electron density one made by localized small polarons giving rise to paramagnetic or antiferromagnetic domains depending on temperature. Employing the above-mentioned variational scheme, in this paper spectral and optical properties of manganites are derived for  $x=0.3$  at different temperatures. It is found that the phase separation regime induces a robust pseudogap in the excitation spectrum of the system. Then the conductivity spectra are characterized by a transfer of spectral weight from high to low energies, as the temperature  $T$  decreases. In the metallic ferromagnetic phase, at low  $T$  two types of infrared absorption come out: a Drude term and a broad absorption band due respectively to the coherent and incoherent motion of large polarons. The obtained results turn out in good agreement with experiments.

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In the last years the perovskite oxides  $\text{La}_{1-x}\text{A}_x\text{MnO}_3$  (A represents a divalent alkali element such as Sr or Ca) have become one of the main research areas of the condensed matter community because of the colossal magnetoresistance effect exhibited for  $0.2 \leq x \leq 0.5$ .<sup>1,2</sup> The study of these materials started in the 1950s showing the strong correlation between magnetization and resistivity.<sup>3</sup> Since in the electronically active Mn  $3d$  orbitals the mean number of  $d$  electrons per Mn is  $4-x$ , three electrons go into the  $t_{2g}$  core states and the remaining  $1-x$  electrons occupy the outer shell  $e_g$  orbitals. The ferromagnetic phase was explained by introducing the double-exchange mechanism<sup>4,5</sup> that takes into account the combined effect of the  $e_g$  electron hopping between nearest-neighbor sites and the very strong Hund's exchange with the localized  $t_{2g}$  electron spins. In the 1990s the discovery of the colossal magnetoresistance phenomenon has aroused a renewed interest for these compounds. In order to explain the experimental data, it has been suggested that, in addition to the double-exchange term, a strong interaction between electrons and lattice distortions plays a non-negligible role.<sup>6</sup> Next the relevance of the Jahn-Teller polaron formation has been confirmed experimentally by the giant isotope shift of the Curie temperature,<sup>7</sup> by measurements of the lattice distortions by extended x-ray-absorption fine structure<sup>8</sup> and by frequency shifts of the internal phonon modes.<sup>9</sup> Furthermore, both pseudogap features and conductivity spectra of these compounds have been discussed in terms of a strong coupling to lattice distortions.<sup>10-15</sup> Nowadays the role of the polaron is widely recognized.<sup>16</sup>

Recent studies have showed the presence of strong tendencies toward the phase separation in manganites.<sup>2,17</sup> At low temperatures for compositions near the metal-insulator transition the ferromagnetic and antiferromagnetic phases coexist while, close to the paramagnetic-to-ferromagnetic transition temperature, the phase separation occurs between paramagnetic and ferromagnetic domains. From a theoretical point of view, the coexistence between hole-poor and hole-

rich phases has been discussed by using exact numerical approaches on small lattices assuming classical Jahn-Teller phonons.<sup>2,18</sup> The pseudogap formation<sup>19</sup> and the optical properties<sup>20,21</sup> also have been studied neglecting the quantum nature of phonons. One of the drawbacks of this approximation in the optical properties is, for example, that at low temperatures in the metallic ferromagnetic phase a narrow Drude peak cannot be obtained.

In a recent paper,<sup>22</sup> some of us have shown, on the basis of a variational approach, that the quantum character of the Jahn-Teller phonons and the polaron formation can be important to explain the experimentally observed tendency of manganites to form inhomogeneous magnetic structures near the phase boundaries. We employ the scheme proposed in that paper to deduce spectral and optical properties of manganites for  $x=0.3$  at different temperatures. It is interesting to study the properties of these compounds at this doping since in the range of concentrations around  $x=0.3$  the colossal magnetoresistance effect is very pronounced in many manganese oxides.<sup>1</sup> In this paper we find that the phase separation regime induces a robust pseudogap in the excitation spectrum of the system. The pseudogap features show a direct relationship with the metal-insulator transition and with the cross-over from the coherent large polaron dynamics to the incoherent small polaron one. Our results turn out compatible with experimental findings.<sup>10,11</sup> Next, by using the formalism of generalized Matsubara Green's functions,<sup>23-27</sup> we determine the scattering rate of charges affected by the interaction with the lattice and the spin fluctuations. The single phonon emission and absorption represent the main mechanism of scattering. However, the damping due to spin fluctuations is effective in the energy range around the chemical potential  $\mu$ . The scattering rate turns out fundamental to derive the very interesting optical properties of the system. With decreasing  $T$ , our spectra are characterized by a transfer of spectral weight from high to low energies filling up the low-frequency optical gap present in the high-temperature phase.

Indeed at high  $T$  the infrared absorption is due to the incoherent small polaron dynamics. Instead, at low temperatures, in the ferromagnetic phase the system shows two types of optical response: a Drude term and a broad absorption band due respectively to the coherent and incoherent motion of large polarons. The results obtained are consistent with experimental data.<sup>12,13,28</sup>

In Sec. I the model and the variational approach are reviewed; in Sec. II the spectral properties are deduced; in Sec. III the damping of the particle motion is calculated; in Sec. IV the optical properties are discussed. In appendix details of the calculations employed to derive the optical properties are reported.

### I. THE MODEL AND THE VARIATIONAL APPROACH

The model takes into account the double-exchange mechanism, the coupling of the  $e_g$  electrons to lattice distortions, and the superexchange interaction between neighboring localized  $t_{2g}$  electrons. The coupling to longitudinal optical phonons arises from the Jahn-Teller effect that splits the  $e_g$  double degeneracy.<sup>29</sup> Adopting the single-orbital approximation (reasonable for  $x < 0.5$ ), the Hamiltonian reads

$$\begin{aligned}
 H = & -t \sum_{\langle i,j \rangle} \left( \frac{S_0^{i,j} + 1/2}{2S + 1} \right) c_i^\dagger c_j + \omega_0 \sum_i a_i^\dagger a_i \\
 & + g \omega_0 \sum_i c_i^\dagger c_i (a_i + a_i^\dagger) \\
 & + \epsilon \sum_{\langle i,j \rangle} \vec{S}_i \cdot \vec{S}_j - \mu \sum_i c_i^\dagger c_i. \quad (1)
 \end{aligned}$$

Here  $t$  is the transfer integral between nearest-neighbor (NN) sites  $\langle i,j \rangle$  for electrons occupying  $e_g$  orbitals,  $S_0^{i,j}$  is the total spin of the subsystem consisting of two localized spins on NN sites and the conduction electron,  $S$  is the spin of the  $t_{2g}$  core states ( $S = 3/2$ ), and  $c_i^\dagger (c_i)$  creates (destroys) an electron with spin parallel to the ionic spin at the  $i$ th site. The first term of the Hamiltonian describes the double-exchange mechanism in the limit where the intra-atomic exchange integral  $J$  is far larger than the transfer integral  $t$ . Furthermore, in Eq. (1)  $\omega_0$  denotes the frequency of the local optical phonon mode,  $a_i^\dagger (a_i)$  is the creation (annihilation) phonon operator at the site  $i$ , the dimensionless parameter  $g$  indicates the strength of the electron-phonon interaction in the Holstein model,<sup>30</sup>  $\epsilon$  represents the antiferromagnetic superexchange coupling between two NN  $t_{2g}$  spins, and  $\mu$  is the chemical potential.

The hopping of electrons is supposed to take place between the equivalent NN sites of a simple cubic lattice separated by the distance  $|n - n'| = a$ . The units are such that the Planck's constant  $\hbar = 1$ , the Boltzmann's constant  $k_B = 1$  and the lattice parameter  $a = 1$ .

Following the recently proposed variational scheme,<sup>22</sup> we perform two successive canonical transformations to treat the electron-phonon interaction variationally.

The first is the variational Lang-Firsov unitary transformation<sup>31,32</sup>

$$U_1 = \exp \left[ -g \sum_j (f c_j^\dagger c_j + \Delta) (a_j - a_j^\dagger) \right] \quad (2)$$

where  $f$  and  $\Delta$  are variational parameters. The quantity  $f$  represents the strength of the coupling between an electron and the phonon displacement on the same site, hence it measures the degree of the polaronic effect. On the other hand, the site-independent parameter  $\Delta$  denotes a displacement field describing static lattice distortions that are not influenced by the instantaneous position of the electrons.

The second canonical transformation is of Bogoliubov type<sup>33</sup>

$$U_2 = \exp \left[ -\alpha \sum_j (a_j^\dagger a_j^\dagger - a_j a_j) \right], \quad (3)$$

where  $\alpha$  is a variational parameter. It introduces correlations between the emission of successive virtual phonons by the electrons and it is responsible of the phonon frequency renormalization.

The transformed Hamiltonian  $\tilde{H} = U_2 U_1 H U_1^{-1} U_2^{-1}$  becomes

$$\begin{aligned}
 \tilde{H} = & \sum_{\langle i,j \rangle} C_{i,j} c_i^\dagger c_j + \bar{\omega}_0 \sum_i a_i^\dagger a_i + N \omega_0 \sinh^2(2\alpha) + N \omega_0 g^2 \Delta^2 \\
 & + \omega_0 \sinh(2\alpha) \cosh(2\alpha) \sum_i (a_i^\dagger a_i^\dagger + a_i a_i) - \omega_0 g e^{2\alpha} \Delta \\
 & \times \sum_i (a_i + a_i^\dagger) + \epsilon \sum_{\langle i,j \rangle} \vec{S}_i \cdot \vec{S}_j + \sum_i c_i^\dagger c_i (\tilde{C}_i + \eta - \mu), \quad (4)
 \end{aligned}$$

where the operator  $C_{i,j}$  reads

$$C_{i,j} = -t \left( \frac{S_0^{i,j} + 1/2}{2S + 1} \right) X_i^\dagger X_j \quad (5)$$

and  $X_i$  is the phononic operator

$$X_i = \exp[ g f e^{-2\alpha} (a_i - a_i^\dagger) ]. \quad (6)$$

Furthermore, in the Hamiltonian (4) we specify the renormalized phonon frequency  $\bar{\omega}_0 = \omega_0 \cosh(4\alpha)$ , the number of lattice sites  $N$ , the phonon operator  $\tilde{C}_i$ ,

$$\tilde{C}_i = \omega_0 g (1 - f) e^{2\alpha} (a_i + a_i^\dagger), \quad (7)$$

and the quantity  $\eta$ ,

$$\eta = \omega_0 g^2 f (f - 2) + 2 \omega_0 g^2 (f - 1) \Delta \quad (8)$$

that measures the electronic band shift due to the electron-phonon interaction.

To obtain the free energy in a variational scheme, we introduce a test Hamiltonian characterized by electron, phonon, and spin degrees of freedom nonmutually interacting

$$\begin{aligned}
H_{test} = & -t_{eff} \sum_{\langle i,j \rangle} c_i^\dagger c_j + \bar{\omega}_0 \sum_i a_i^\dagger a_i + N\omega_0 \sinh^2(2\alpha) \\
& + N\omega_0 g^2 \Delta^2 - g_s \mu_B \sum_i \vec{h}_{eff} \cdot \vec{S}_i + (\eta - \mu) \sum_i c_i^\dagger c_i.
\end{aligned} \tag{9}$$

The quantity  $t_{eff}$  denotes the effective transfer integral

$$t_{eff} = t \left\langle \left( \frac{S_0 + 1/2}{2S + 1} \right) \right\rangle e^{-S_T}, \tag{10}$$

where the symbol  $\langle \rangle$  indicates a thermal average and the quantity  $S_T$  is

$$S_T = g^2 f^2 e^{-4\alpha} (2N_0 + 1) \tag{11}$$

with  $N_0$  being the average number of phonons with frequency  $\bar{\omega}_0$ . In the test Hamiltonian (9)  $g_s$  is the dimensionless electron-spin factor ( $g_s \approx 2$ ),  $\mu_B$  is the Bohr magneton, and  $h_{eff}$  is the effective molecular magnetic field in a cell containing two neighboring sites. The magnetic field is induced by a partial orientation of ionic spins around this cell and is determined by the variational approach.<sup>34</sup> We notice that in Eq. (10) the factor  $e^{-S_T}$  controls the band renormalization due to the polaron formation.

To obtain the variational free energy of the system, we employ the Bogoliubov inequality

$$F \leq F_{test} + \langle \tilde{H} - H_{test} \rangle_t, \tag{12}$$

where  $\langle \rangle_t$  indicates a thermodynamic average made using the test Hamiltonian. This approach allows to treat the local spin dynamics in a variational mean field theory.<sup>34</sup> The free energy per site becomes

$$\begin{aligned}
\frac{F}{N} = & f_{test}^{el} + T \ln(1 - e^{-\beta \bar{\omega}_0}) + \omega_0 \sinh^2(2\alpha) + \omega_0 g^2 (1-f)^2 \rho^2 \\
& - T \ln \nu_S \pm \frac{\epsilon}{2} Z S^2 m_S^2 + T \lambda m_S,
\end{aligned} \tag{13}$$

where the electron free energy reads

$$\begin{aligned}
f_{test}^{el} = & \left( \frac{1}{N} \right) \sum_{\mathbf{k}} \xi_{\mathbf{k}} n_{\mathbf{k}} + T \left( \frac{1}{N} \right) \sum_{\mathbf{k}} [n_{\mathbf{k}} \ln n_{\mathbf{k}} \\
& + (1 - n_{\mathbf{k}}) \ln(1 - n_{\mathbf{k}})]
\end{aligned} \tag{14}$$

with  $n_{\mathbf{k}} = n_F(\xi_{\mathbf{k}})$  the Fermi distribution function. We get  $\xi_{\mathbf{k}} = \bar{\epsilon}_{\mathbf{k}} - \mu$ , where  $\bar{\epsilon}_{\mathbf{k}} = \epsilon_{\mathbf{k}} + \eta$  is the renormalized electronic band and  $\epsilon_{\mathbf{k}}$  the band dispersion

$$\epsilon_{\mathbf{k}} = -2t_{eff} [\cos(k_x) + \cos(k_y) + \cos(k_z)]. \tag{15}$$

Furthermore, in Eq. (13)  $\beta$  is the inverse of the temperature,  $\nu_S$  is the partition function of the localized spins, the top and bottom sign of  $\epsilon$  hold, respectively, for the ferromagnetic and antiferromagnetic solutions of the localized spins,  $Z$  indicates the number of nearest neighbors,  $m_S$  represents the magnetization of the localized spins, and  $\lambda$  is a dimensionless variational parameter proportional to the effective magnetic field.

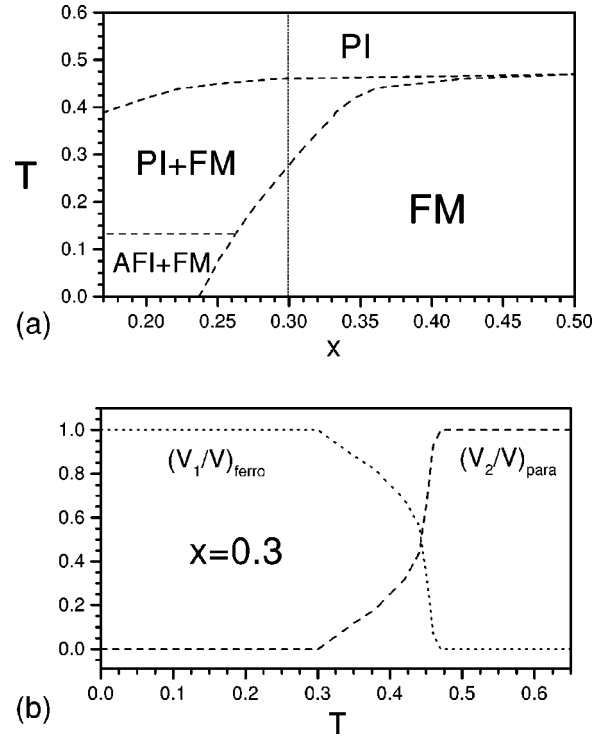


FIG. 1. (a) The phase diagram corresponding to  $t=2\omega_0$ ,  $g=2.8$  and  $\epsilon=0.01t$ . PI means paramagnetic insulator, FM ferro-magnetic metal, and AFI antiferromagnetic insulator. The areas PI+FM and AFI+FM indicate regions where localized (PI and AFI) and delocalized (FM) phases coexist. (b) The fractions of volume  $(V_1/V)_{ferro}$  and  $(V_2/V)_{para}$  at  $x=0.3$  as a function of the temperature ( $T$  is expressed in units of  $\omega_0$ ).

In order to simplify the calculations, we consider a semi-circular electronic density of states

$$g(\xi) = \left( \frac{2}{\pi W^2} \right) \theta(W - |\xi - \eta + \mu|) \sqrt{W^2 - (\xi - \eta + \mu)^2}, \tag{16}$$

where  $W = Zt_{eff}$  is the renormalized band half-width and  $\theta(x)$  is the Heaviside function. Actually  $g(\xi)$  represents a simple approximate expression for the exact density of states and it is generally used for a three-dimensional lattice.<sup>35,36</sup> Therefore the electron free energy becomes

$$\begin{aligned}
f_{test}^{el} = & \int_{-\infty}^{\infty} d\xi g(\xi) \xi n_F(\xi) + T \int_{-\infty}^{\infty} d\xi g(\xi) \{n_F(\xi) \ln n_F(\xi) \\
& + [1 - n_F(\xi)] \ln[1 - n_F(\xi)]\}.
\end{aligned} \tag{17}$$

As shown in Ref. 22, in the intermediate electron-phonon coupling regime, the free energy (13) gives rise to a region of coexisting phases characterized by different electron densities  $\rho_1$  and  $\rho_2$  but the same value of chemical potential  $\mu$ . The quantities  $\rho_1$  and  $\rho_2$  depend on the temperature and correspond to homogeneous phases of the system constituted by large and small polarons, respectively. Hence, near the metal-insulator transition [see Fig. 1(a)], the system segregates into ferromagnetic and antiferromagnetic or paramag-

netic domains of large and small polarons. We observe that the quantity  $x$  denotes the hole concentration, that is  $x=1-\rho$ , where  $\rho$  is the electron concentration.

In the region of coexistence, the fractions of volume,  $V_1/V$  and  $V_2/V$ , filled with density  $\rho_1$  and  $\rho_2$ , are determined by the two conditions:  $V_1/V+V_2/V=1$  and  $(V_1/V)\rho_1+(V_2/V)\rho_2=\rho$ . In Fig. 1(b) we report the fractions of volume  $(V_1/V)_{ferro}$  and  $(V_2/V)_{para}$  for  $x=0.3$  as a function of the temperature [the dotted line in Fig. 1(a) allows us to recognize the different phases of the system at this doping].

In the regime of phases coexisting,<sup>37,38</sup> it is possible to calculate any property  $B$  of the system by means of the linear combination of the properties  $B_1$  and  $B_2$  characteristic of the single phases assuming the respective volume fractions  $(V_1/V)$  and  $(V_2/V)$  as weights,

$$B = \left(\frac{V_1}{V}\right)B_1 + \left(\frac{V_2}{V}\right)B_2. \quad (18)$$

Hence we assume that in this regime the properties of the system are independent of morphology of coexisting domains.

## II. SPECTRAL PROPERTIES

In this section we calculate the spectral properties of the system.

Performing the two canonical transformations (2),(3) and exploiting the cyclic properties of the trace, the electron Matsubara Green's function becomes<sup>39</sup>

$$\mathcal{G}(\mathbf{k}, \tau) = -\left(\frac{1}{N}\right) \sum_{i,j} e^{i\mathbf{k}\cdot(\mathbf{R}_i - \mathbf{R}_j)} \langle T_\tau \tilde{c}_i(\tau) \tilde{X}_i(\tau) c_j^\dagger X_j^\dagger \rangle_t, \quad (19)$$

where  $\tilde{c}_i(\tau) = e^{\tau\tilde{H}} c_i e^{-\tau\tilde{H}}$  and  $\tilde{X}_i(\tau) = e^{\tau\tilde{H}} X_i e^{-\tau\tilde{H}}$ , with  $\tilde{H}$  given by Eq. (4).

The correlation function in Eq. (19) can be disentangled into electronic and phononic terms by using the test Hamiltonian (9), hence

$$\mathcal{G}(\mathbf{k}, \tau) = -\left(\frac{1}{N}\right) \sum_{i,j} e^{i\mathbf{k}\cdot(\mathbf{R}_i - \mathbf{R}_j)} \langle T_\tau \bar{c}_i(\tau) c_j^\dagger \rangle_t \langle T_\tau \bar{X}_i(\tau) X_j^\dagger \rangle_t, \quad (20)$$

where now we have  $\bar{c}_i(\tau) = e^{\tau H_i} c_i e^{-\tau H_i}$  and  $\bar{X}_i(\tau) = e^{\tau H_i} X_i e^{-\tau H_i}$ .

The Green's function in Matsubara frequencies  $\omega_n$  reads

$$\begin{aligned} \mathcal{G}(\mathbf{k}, i\omega_n) &= e^{-S_T} \mathcal{G}^{(0)}(\mathbf{k}, i\omega_n) \\ &+ e^{-S_T} \left(\frac{1}{N}\right) \sum_{\mathbf{k}_1} \left(\frac{1}{\beta}\right) \sum_m \mathcal{G}^{(0)}(\mathbf{k}_1, i\omega_m) \\ &\times \int_0^\beta \exp[(i\omega_n - i\omega_m)\tau] \\ &\times \left( \exp\left[ s \cosh\left[ \bar{\omega}_0 \left( \tau - \frac{\beta}{2} \right) \right] \right] - 1 \right), \quad (21) \end{aligned}$$

where  $\mathcal{G}^{(0)}(\mathbf{k}, i\omega_n)$  is the Green's function of noninteracting particles and

$$s = 2f^2 g^2 e^{-4\alpha} [N_0(N_0+1)]^{1/2}. \quad (22)$$

We notice that two physically distinct terms appear in Eq. (21), the coherent one and the incoherent one.<sup>40,41</sup> The first derives from the coherent motion of electrons and their surrounding phonon cloud (this process is called *diagonal transition*). The renormalization factor  $e^{-S_T}$  appearing in this term plays the role of the residue  $Z$  at the pole in a Fermi-liquid description.<sup>42</sup> The second contribution describes the possibility of changing the number of phonons in the phonon cloud during the electron hopping (this process is known as *nondiagonal transition*). This is confirmed by the presence of the sum over all electron momenta that reflects the fact that the electron momentum is not conserved separately from the phonon one.

In the second term of the expression (21), we can perform the expansion

$$\begin{aligned} \exp\left[ s \cosh\left[ \bar{\omega}_0 \left( \tau - \frac{\beta}{2} \right) \right] \right] &= 2 \sum_{l=1}^{+\infty} I_l(s) \cosh\left[ \bar{\omega}_0 \left( \tau - \frac{\beta}{2} \right) \right] \\ &+ I_0(s) \quad (23) \end{aligned}$$

into multiphonon contributions, where the symbols  $I_l(s)$  denote the modified Bessel functions.

Making the analytic continuation  $i\omega_n \rightarrow \omega + i\delta$ , one obtains the retarded Green's function  $G_{ret}(\mathbf{k}, \omega)$  and the spectral function

$$\begin{aligned} A(\mathbf{k}, \omega) &= -2 \operatorname{Im} G_{ret}(\mathbf{k}, \omega) \\ &= 2\pi e^{-S_T} \delta(\omega - \xi_{\mathbf{k}}) + 2\pi e^{-S_T} \sum_{l(\neq 0)}^{+\infty} I_l(s) e^{\beta l \bar{\omega}_0/2} \\ &\times [1 - n_F(\omega - l\bar{\omega}_0)] g(\omega - l\bar{\omega}_0) + 2\pi e^{-S_T} \\ &\times \sum_{l(\neq 0)}^{+\infty} I_l(s) e^{\beta l \bar{\omega}_0/2} n_F(\omega + l\bar{\omega}_0) g(\omega + l\bar{\omega}_0) \\ &+ 2\pi e^{-S_T} [I_0(s) - 1] g(\omega). \quad (24) \end{aligned}$$

The first term represents the purely polaronic band contribution and shows a delta behavior. The remaining terms provide the incoherent contribution and spread over a wide energy range. Thus we get the renormalized density of states

$$N(\omega) = \frac{1}{2\pi} \left(\frac{1}{N}\right) \sum_{\mathbf{k}} A(\mathbf{k}, \omega), \quad (25)$$

where the spectral function  $A(\mathbf{k}, \omega)$  is given by Eq. (24). We note that the sum rule  $\int_{-\infty}^{+\infty} (d\omega/2\pi) A(\mathbf{k}, \omega) = 1$  is satisfied and  $N(\omega)$  is normalized to unity.

So we can evaluate the renormalized electron distribution function

$$\begin{aligned}
n_{\mathbf{k}}^{ren} &= \int_{-\infty}^{+\infty} \frac{d\omega}{2\pi} A(\mathbf{k}, \omega) n_F(\omega) \\
&= e^{-S_T} n_F(\xi_{\mathbf{k}}) + e^{-S_T} \sum_{l(\neq 0)}^{+\infty} I_l(s) e^{\beta l \bar{\omega}_0/2} \\
&\quad \times \int_{-\infty}^{\infty} d\xi g(\xi) [1 - n_F(\xi)] n_F(\xi + l \bar{\omega}_0) + e^{-S_T} \\
&\quad \times \sum_{l(\neq 0)}^{+\infty} I_l(s) e^{\beta l \bar{\omega}_0/2} \int_{-\infty}^{\infty} d\xi g(\xi) n_F(\xi) n_F(\xi - l \bar{\omega}_0) \\
&\quad + e^{-S_T} [I_0(s) - 1] \int_{-\infty}^{\infty} d\xi g(\xi) n_F(\xi). \tag{26}
\end{aligned}$$

At zero temperature it is found that  $n^{ren}(\mu - \delta) - n^{ren}(\mu + \delta) = e^{-S_T}$ , so the factor  $e^{-S_T}$  determines the jump in the Fermi distribution function.<sup>40</sup> For low temperatures and  $x > 0.25$ ,  $e^{-S_T}$  is about 0.9, therefore the spectral properties indicate a polaronic Fermi-liquid state in the metallic ferromagnetic phase.

On the other hand, for the small polaron excitations and high temperatures, we expect that the spectral properties are dominated only by the incoherent term. It is well known<sup>39</sup> that in this limit the spectral function (24) consists of a sum of delta functions on the points  $n\omega_0$ . This is caused by the choice of a dispersionless phonon frequency. To overcome this drawback, there are two ways of proceeding: the first is to introduce a phononic dispersion relation,<sup>30</sup> whereas the second is to make a high-temperature expansion.<sup>43</sup> In this paper we concentrate on the latter approach performing in Eq. (21) the following approximation<sup>43</sup>

$$e^{-S_T} \exp \left\{ s \cosh \left[ \bar{\omega}_0 \left( \tau - \frac{\beta}{2} \right) \right] \right\} \approx \exp(-\bar{s} \tau) \exp \left( \frac{\bar{s}}{\beta} \tau^2 \right), \tag{27}$$

where  $\bar{s}$  is

$$\bar{s} = \frac{2f^2 g^2 e^{-4\alpha} \bar{\omega}_0^2 \beta}{4 \sinh \left( \frac{\beta \bar{\omega}_0}{2} \right)} \approx g^2 \omega_0. \tag{28}$$

This expansion is valid as long as  $(\beta \bar{s}/12)(\beta \bar{\omega}_0/4)^2 < 1$ , hence  $T > 0.3\omega_0$ . In this limit the Green's function becomes

$$\begin{aligned}
\mathcal{G}(\mathbf{k}, i\omega_n) &= e^{-S_T} \mathcal{G}^{(0)}(\mathbf{k}, i\omega_n) - e^{-S_T} \left( \frac{1}{N} \right) \sum_{\mathbf{k}_1} \mathcal{G}^{(0)}(\mathbf{k}_1, i\omega_n) \\
&\quad + \left( -\frac{i}{2} \right) \sqrt{\frac{\pi\beta}{\bar{s}}} \left( \frac{1}{N} \right) \sum_{\mathbf{k}_1} (1 - n_{\mathbf{k}_1}) W \\
&\quad \times \left[ \frac{1}{2} \sqrt{\frac{\beta}{\bar{s}}} (i\omega_n - \xi_{\mathbf{k}_1} - \bar{s}) \right] \\
&\quad + \left( -\frac{i}{2} \right) \sqrt{\frac{\pi\beta}{\bar{s}}} \left( \frac{1}{N} \right) \sum_{\mathbf{k}_1} n_{\mathbf{k}_1} W \\
&\quad \times \left[ \frac{1}{2} \sqrt{\frac{\beta}{\bar{s}}} (i\omega_n - \xi_{\mathbf{k}_1} + \bar{s}) \right], \tag{29}
\end{aligned}$$

where  $W(z)$  is

$$W(z) = e^{-z^2} \left[ 1 + \frac{2i}{\sqrt{\pi}} \int_0^z d\xi e^{\xi^2} \right]. \tag{30}$$

In the small polaron case, we get  $\xi_{\mathbf{k}} \approx -g^2 \omega_0 - \mu$ . The incoherent term yields

$$\begin{aligned}
A(\mathbf{k}, \omega) \approx A(\omega) &= [1 - n_F(-g^2 \omega_0 - \mu)] \sqrt{\frac{\pi\beta}{\bar{s}}} \\
&\quad \times \exp \left[ -\frac{\beta}{4\bar{s}} (\omega + g^2 \omega_0 + \mu - \bar{s})^2 \right] + n_F(-g^2 \omega_0 \\
&\quad - \mu) \sqrt{\frac{\pi\beta}{\bar{s}}} \exp \left[ -\frac{\beta}{4\bar{s}} (\omega + g^2 \omega_0 + \mu + \bar{s})^2 \right]. \tag{31}
\end{aligned}$$

We stress that this spectral function represents the envelope of the delta functions obtained by Eq. (24) for small polaron excitations in the limit of high temperatures. Furthermore, using the spectral function (31), we can derive the renormalized density of states

$$N(\omega) = \frac{A(\omega)}{2\pi} \tag{32}$$

that turns out normalized to unity.

In Fig. 2 we report the spectral function for two different temperatures at  $x = 0.3$ . At  $T = 0.05\omega_0$ , the spectral function is deduced by Eq. (24) and the case  $\xi_{\mathbf{k}} = 0$  is considered. It is apparent that in the large polaron ferromagnetic phase the weight of the coherent term is prevalent. For  $T = 0.47\omega_0$ , in the paramagnetic phase, the spectral function is provided by



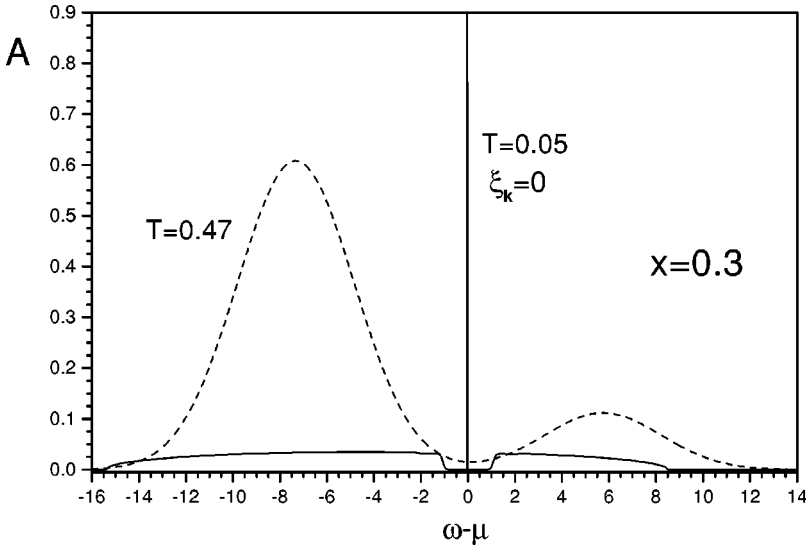


FIG. 2. The spectral function (corresponding to  $t=2\omega_0$  and  $g=2.8$ ) at  $x=0.3$  as a function of the energy (in units of  $\omega_0$ ) for two different temperatures: (solid line)  $T=0.05$  and  $\xi_{\mathbf{k}}=0$ , (dashed line)  $T=0.47$  ( $T$  is expressed in units of  $\omega_0$ ).

Eq. (31) and it is determined by the incoherent dynamics of the small polaron excitations. It is made up of two bands peaked approximately around  $-g^2\omega_0$  and  $g^2\omega_0$ , whose different heights are due to the fixed doping. At high temperatures the spectral function is characterized by a large depression in correspondence of  $\mu$ . With decreasing temperature and fixed doping, a transfer of spectral weight occurs crossing the regime of coexisting phases. To stress this important point, in Fig. 3 we present the renormalized density of states

for  $x=0.3$  at different temperatures. In the large-polaron ferromagnetic phase,  $N(\omega)$  is given by Eq. (25), in the small-polaron paramagnetic phase by Eq. (31), in the regime of coexisting phases by the two preceding densities by means of the Eq. (18). We stress that  $T_c$  denotes the ferromagnetic transition temperature that coincides with the beginning of formation of large polaron domains. For  $x=0.3$ ,  $T_c$  is about  $0.46\omega_0$ . For low temperatures incoherent processes of phonon creation and annihilation lead to a depression in the

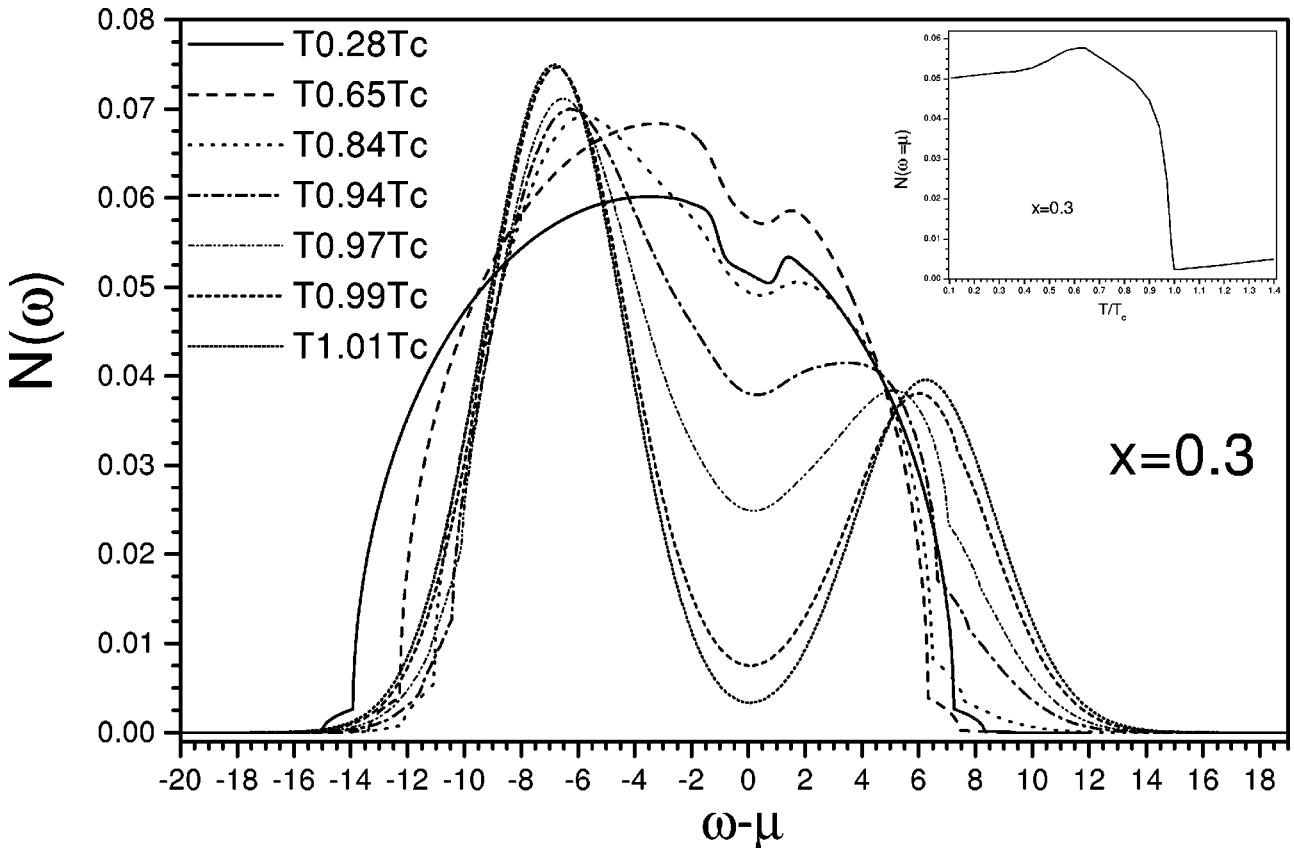


FIG. 3. The renormalized density of states at  $x=0.3$  as a function of the energy (in units of  $\omega_0$ ) for different temperatures. In the inset the renormalized density of states at  $\mu$  as a function of the temperature. The results are obtained for  $t=2\omega_0$  and  $g=2.8$ .

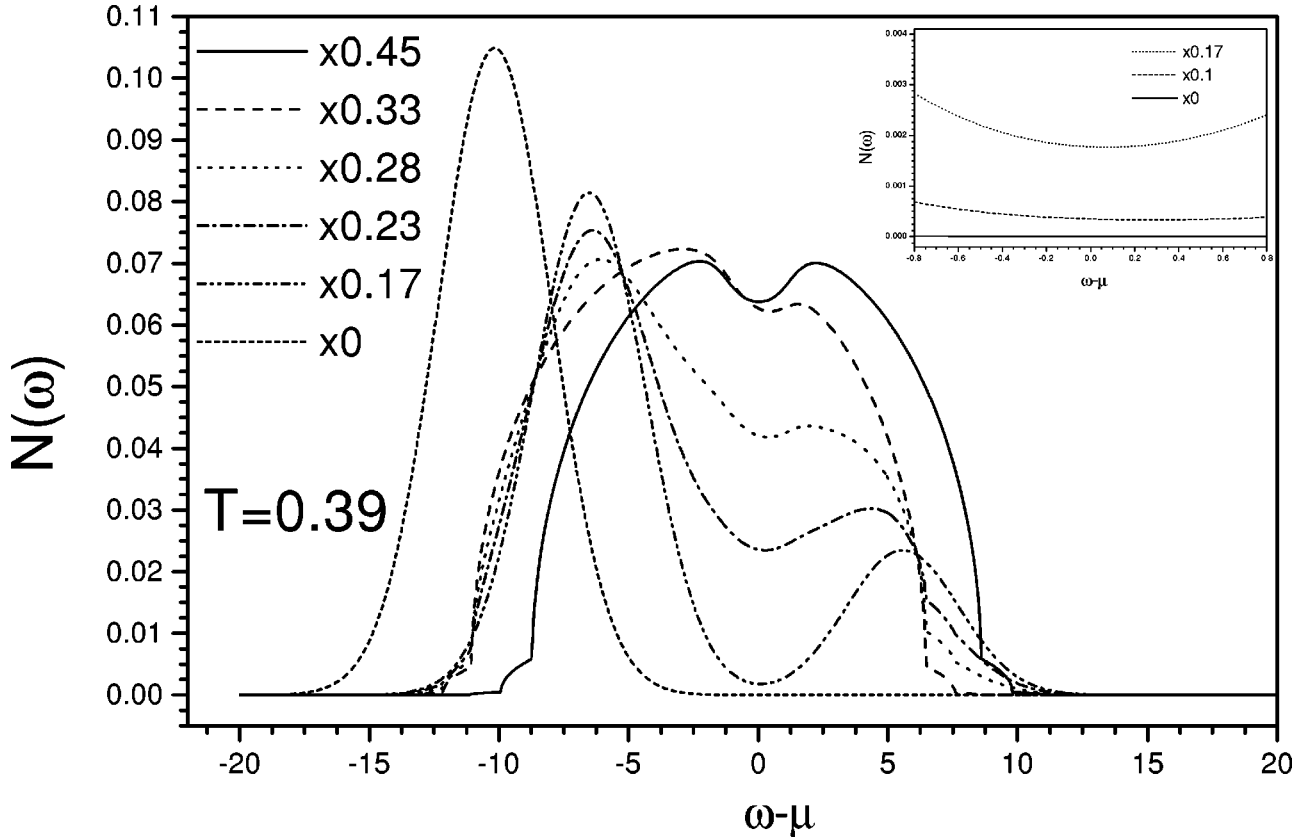


FIG. 4. The renormalized density of states at  $T=0.39\omega_0$  as a function of the energy (in units of  $\omega_0$ ) for different densities. In the inset the renormalized density of states in a small range of energy around the chemical potential  $\mu$ .

density of states around the chemical potential  $\mu$ . When the temperature increases, the effect of band renormalization driven by  $e^{-S\tau}$  is stronger than this decrease of electronic states at  $\mu$ . Since the density of states is normalized, the net effect is a rise of states at  $\mu$  (compare graphs at  $T=0.28T_c$  and  $T=0.65T_c$ ). However, when the phase separation regime takes place (starting from  $T=0.65T_c$ ), the formation of paramagnetic insulating domains in the metallic background causes a robust pseudogap in the excitation spectrum. The subtraction of levels below  $\mu$  is strongly temperature dependent and, as we see in the inset of Fig. 3, it finishes at  $T_c$ . Therefore the pseudogap features show a direct relationship with the metal-insulator transition and the coherent-incoherent crossover of charge dynamics. Above  $T_c$  the density of states at  $\mu$  is low and with increasing temperature it is nearly constant. It is important to note that these results are compatible with photoemission experiments. Indeed the density of states at  $\mu$  in the inset of Fig. 3 bears a strong resemblance with the spectral weights reported in Refs. 10, and 11.

Finally it is interesting to study the pseudogap formation fixing the temperature and varying the hole concentration  $x$  (see Fig. 4). At  $T=0.39\omega_0$ , the system is in the insulating paramagnetic phase from  $x=0$  to  $x=0.17$ , in the regime of the insulating paramagnetic and metallic ferromagnetic coexisting phases from  $x=0.17$  to  $x=0.33$ , and in the metallic ferromagnetic phase from  $x=0.33$ . With  $x$  increasing, holes are added to the system and ferromagnetic large polaron domains are created yielding pseudogap features. As soon as

the phase separation regime is set up at  $x=0.17$  (inset of Fig. 4), electronic levels are occupied in the energy range around the chemical potential  $\mu$ . Thus, the charge carriers prefer to be located in the metallic ferromagnetic domains in order to increase the kinetic energy gain.

### III. DAMPING

In this section we deal with the polaron self-energy. This quantity allows us to determine the scattering rate of particles affected by lattice distortions and spin fluctuations. The latter, in turn, will play an essential role in the infrared absorption calculations.

Retaining only the dominant autocorrelation terms at the second step of iteration,<sup>23-27</sup> we can derive the self-energy

$$\begin{aligned} \Sigma^{(2)}(\mathbf{k}, i\omega_n) = & \varepsilon_{\mathbf{k}} + \left(\frac{1}{N}\right) \sum_{\mathbf{k}_1} \left(\frac{1}{\beta}\right) \sum_m \mathcal{G}^{(0)}(\mathbf{k}_1, i\omega_m) \\ & \times \int_0^\beta d\tau \exp[(i\omega_n - i\omega_m)\tau] [f_1(\tau) + f_2(\tau)], \end{aligned} \quad (33)$$

where  $\varepsilon_{\mathbf{k}}$  is given by Eq. (15),  $f_1(\tau)$  is

$$f_1(\tau) = Zt^2 e^{-2S_T} \left\{ \left\langle \left( \frac{S_0 + 1/2}{2S+1} \right)^2 \right\rangle \exp \left\{ z \cosh \left[ \bar{\omega}_0 \left( \tau - \frac{\beta}{2} \right) \right] \right\} - \left\langle \left( \frac{S_0 + 1/2}{2S+1} \right)^2 \right\rangle \right\} \quad (34)$$

with

$$z = 4g^2 f^2 e^{-4\alpha} [N_0(N_0 + 1)]^{1/2} \quad (35)$$

and  $f_2(\tau)$  is

$$f_2(\tau) = 2g^2 \omega_0^2 e^{4\alpha} (1-f)^2 [N_0(N_0 + 1)]^{1/2} \cosh \left[ \bar{\omega}_0 \left( \tau - \frac{\beta}{2} \right) \right]. \quad (36)$$

Making the analytic continuation  $i\omega_n \rightarrow \omega + i\delta$ , we can calculate the scattering rate

$$\Gamma(\mathbf{k}) = \bar{\Gamma}(\mathbf{k}, \omega = \xi_{\mathbf{k}}) = -2 \text{Im} \Sigma_{ret}^{(2)}(\mathbf{k}, \omega = \xi_{\mathbf{k}}). \quad (37)$$

We realize that the dependence of  $\Gamma(\mathbf{k})$  on  $\mathbf{k}$  is due only to  $\xi_{\mathbf{k}}$ . Hence, performing the expansion (23) into the series of multiphonon processes, we can express the scattering rate in the following way:

$$\Gamma(\mathbf{k}) = \Gamma(\xi_{\mathbf{k}}) = \Gamma_{1\text{-phon}}(\xi_{\mathbf{k}}) + \Gamma_{\text{multi-phon}}(\xi_{\mathbf{k}}) + \Gamma_{\text{Spin-Fluct}}(\xi_{\mathbf{k}}), \quad (38)$$

where  $\Gamma_{1\text{-phon}}$  is the contribution due to single-phonon processes only,

$$\begin{aligned} \Gamma_{1\text{-phon}}(\xi_{\mathbf{k}}) &= 2Zt^2 e^{-2S_T} \left\langle \left( \frac{S_0 + 1/2}{2S+1} \right)^2 \right\rangle I_1(z) \\ &\quad \times \sinh \left( \frac{\beta \bar{\omega}_0}{2} \right) g_{1,l=1}(\xi_{\mathbf{k}}) \\ &\quad + g^2 \omega_0^2 e^{4\alpha} (1-f)^2 g_2(\xi_{\mathbf{k}}), \end{aligned} \quad (39)$$

$\Gamma_{\text{multi-phon}}$  represents the scattering rate by multiphonon processes,

$$\begin{aligned} \Gamma_{\text{multi-phon}}(\xi_{\mathbf{k}}) &= 2Zt^2 e^{-2S_T} \left\langle \left( \frac{S_0 + 1/2}{2S+1} \right)^2 \right\rangle \sum_{l=2}^{+\infty} I_l(z) \\ &\quad \times \sinh \left( \frac{\beta \bar{\omega}_0 l}{2} \right) g_{1,l}(\xi_{\mathbf{k}}), \end{aligned} \quad (40)$$

and  $\Gamma_{\text{Spin-Fluct}}$  denotes the damping term by spin fluctuations,

$$\begin{aligned} \Gamma_{\text{Spin-Fluct}}(\xi_{\mathbf{k}}) &= Zt^2 e^{-2S_T} \left[ I_0(z) \left\langle \left( \frac{S_0 + 1/2}{2S+1} \right)^2 \right\rangle \right. \\ &\quad \left. - \left\langle \left( \frac{S_0 + 1/2}{2S+1} \right)^2 \right\rangle \right] B(\xi_{\mathbf{k}}). \end{aligned} \quad (41)$$

Here  $g_{1,l}$  reads

$$g_{1,l}(\xi_{\mathbf{k}}) = [N_0(l\bar{\omega}_0) + n_F(\xi_{\mathbf{k}} + l\bar{\omega}_0)] B(\xi_{\mathbf{k}} + l\bar{\omega}_0) + [N_0(l\bar{\omega}_0) + 1 - n_F(\xi_{\mathbf{k}} - l\bar{\omega}_0)] B(\xi_{\mathbf{k}} - l\bar{\omega}_0) \quad (42)$$

and  $g_2$  reads

$$g_2(\xi_{\mathbf{k}}) = [N_0 + n_F(\xi_{\mathbf{k}} + \bar{\omega}_0)] B(\xi_{\mathbf{k}} + \bar{\omega}_0) + [N_0 + 1 - n_F(\xi_{\mathbf{k}} - \bar{\omega}_0)] B(\xi_{\mathbf{k}} - \bar{\omega}_0). \quad (43)$$

The function  $B(x)$  is defined as

$$B(x) = 2\pi g(x), \quad (44)$$

where  $g(x)$  is the density of states (16).

We stress that the decomposition of the scattering rate in three distinct terms has been introduced in order to simplify the analysis of our results. Furthermore, we observe that the scattering rate for small-polaron excitations can be correctly calculated only within a self-consistent treatment.<sup>24,25</sup> This can be carried out substituting in Eq. (33)  $\mathcal{G}^{(0)}$  for a new Green's function  $\tilde{\mathcal{G}}$ . Using the Lehmann representation we can write

$$\tilde{\mathcal{G}}(\mathbf{k}, i\omega_n) = \int_{-\infty}^{+\infty} d\omega \frac{\tilde{A}(\mathbf{k}, \omega)}{2\pi i\omega_n - \omega}, \quad (45)$$

where the spectral function  $\tilde{A}$  is assumed to be

$$\tilde{A}(\mathbf{k}, \omega) = \frac{\Gamma(\mathbf{k})}{[\Gamma(\mathbf{k})]^2/4 + (\omega - \xi_{\mathbf{k}})^2}. \quad (46)$$

So, employing this self-consistent procedure, we get the scattering rate  $\Gamma^{(sc)}(\xi_{\mathbf{k}})$ ,

$$\Gamma^{(sc)}(\xi_{\mathbf{k}}) = \Gamma_{1\text{-phon}}^{(sc)}(\xi_{\mathbf{k}}) + \Gamma_{\text{multi-phon}}^{(sc)}(\xi_{\mathbf{k}}) + \Gamma_{\text{Spin-Fluct}}^{(sc)}(\xi_{\mathbf{k}}), \quad (47)$$

where  $\Gamma_{1\text{-phon}}^{(sc)}$ ,  $\Gamma_{\text{multi-phon}}^{(sc)}$  and  $\Gamma_{\text{Spin-Fluct}}^{(sc)}$  are calculated by Eqs. (39), (40), and (41), respectively, substituting the function  $B(x)$  for  $B^{(sc)}(x)$

$$B^{(sc)}(x) = \left( \frac{1}{N} \right) \sum_{\mathbf{k}_1} \tilde{A}(\mathbf{k}_1, x) = \int_{-\infty}^{+\infty} d\xi_1 g(\xi_1) \tilde{A}(\xi_1, x). \quad (48)$$

Since the spectral function  $\tilde{A}$  (46) depends on  $\Gamma$ , the Eq. (47) allows to evaluate self-consistently the damping due to polaron formation and spin fluctuations. Clearly, when in the right-hand side of Eq. (47) the limit  $\Gamma \rightarrow 0$  is made, the equation (38) is obtained.

In Fig. 5(a) we report the scattering rate  $\Gamma$  of large polaron excitations for different temperatures. Because of the augmented number of phonons and the enhanced role of spin fluctuations with rising temperature, the quantity  $\Gamma$  increases. At low temperatures we notice that the scattering rate is zero for energies within  $\omega_0$  of the chemical potential  $\mu$ . This means that at these temperatures the spin fluctuations are too feeble to scatter electrons and the main mechanism of energy loss is the emission or the absorption of phonons with frequency  $\omega_0$  (one-phonon processes are prevalent). Thus the



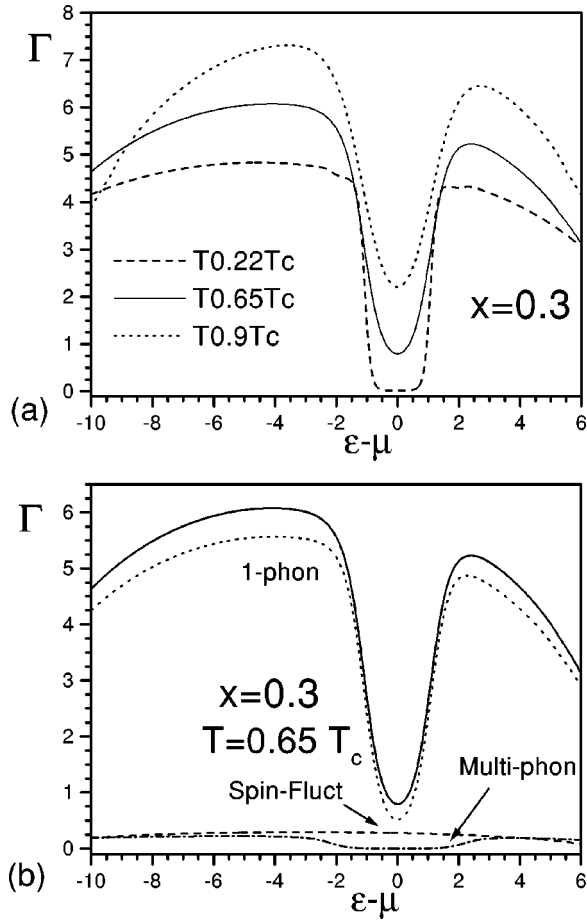


FIG. 5. (a) The scattering rate (corresponding to  $t=2\omega_0$  and  $g=2.8$ ) at  $x=0.3$  as a function of the energy (in units of  $\omega_0$ ) for different temperatures. (b) The scattering rate (solid line), 1-phonon rate (dotted line), spin-fluctuation rate (dashed line), and multiphonon rate (dotted-dashed line) at  $x=0.3$  and  $T=0.65T_c$  as a function of the energy (in units of  $\omega_0$ ).

behavior of the scattering rate is explained by the fact that the quasiparticle excitations within  $\omega_0$  of the Fermi energy cannot lose energy because of the Fermi statistics.<sup>39</sup>

In Fig. 5(b) we concentrate on the components of the scattering rate at a fixed temperature ( $T=0.65T_c$ ). It is confirmed that also at higher temperatures the single-phonon emission and absorption give an important contribution to the quantity  $\Gamma$ . Nevertheless we stress that the spin fluctuations are effective to enhance the scattering rate just in the important energy range around the chemical potential  $\mu$ . Finally, the multiphonon contribution is dominated by two-phonon processes and it is negligible.

We stress that for the small-polaron excitations the quantity  $\Gamma$  decreases when  $T$  increases resembling the behavior of  $t_{eff}$ . We note, however, that, for  $T > 0.3\omega_0$ ,  $\Gamma$  is always larger than  $t_{eff}$ , making clear that the electronic states lose their individual characteristics and the electron motion is predominantly a diffusive process.

The introduction of the damping allows us to improve the approximations of calculation for the spectral properties: indeed we can determine the spectral function,

$$\begin{aligned}
 A(\mathbf{k}, \omega) &= e^{-S_T} \tilde{A}(\mathbf{k}, \omega) + 2e^{-S_T} \sum_{l=1}^{+\infty} I_l(s) \sinh\left(\frac{\beta l \bar{\omega}_0}{2}\right) \\
 &\times \left(\frac{1}{N}\right) \sum_{\mathbf{k}_1} [N_0(l\bar{\omega}_0) + n_F(\omega + l\bar{\omega}_0)] \tilde{A}(\mathbf{k}_1, \omega \\
 &+ l\bar{\omega}_0) + 2e^{-S_T} \sum_{l=1}^{+\infty} I_l(s) \sinh\left(\frac{\beta l \bar{\omega}_0}{2}\right) \\
 &\times \left(\frac{1}{N}\right) \sum_{\mathbf{k}_1} [N_0(l\bar{\omega}_0) + 1 - n_F(\omega - l\bar{\omega}_0)] \tilde{A}(\mathbf{k}_1, \omega \\
 &+ l\bar{\omega}_0) + e^{-S_T} [I_0(s) - 1] \left(\frac{1}{N}\right) \sum_{\mathbf{k}_1} \tilde{A}(\mathbf{k}_1, \omega).
 \end{aligned} \tag{49}$$

In the limit  $\Gamma \rightarrow 0$ , the result of Eq. (24) is recovered. We have checked that the quantity  $\Gamma$  does not change the spectral properties in a considerable manner, although it allows to eliminate the delta behavior in the expression (24).

On the other hand, as mentioned, the damping is essential for determining the optical properties of the system.

#### IV. OPTICAL PROPERTIES

In this section we focus our attention on the optical properties.

To determine the linear response to an external field of frequency  $\omega$ , we derive the conductivity tensor  $\sigma_{\alpha,\beta}$  by means of the Kubo formula

$$\sigma_{\alpha,\beta}(\omega) = \left(\frac{ie^2}{\omega}\right) \langle \hat{T}_{\alpha,\beta} \rangle + \left(\frac{i}{\omega}\right) \Pi_{\alpha,\beta}^{ret}(\omega), \tag{50}$$

where  $\hat{T}_{\alpha,\beta}$  is the kinetic energy and  $\Pi_{\alpha,\beta}^{ret}(\omega)$  is the retarded current-current correlation function. The transport of electrons will be supposed to take place between the equivalent NN sites of the cubic lattice, hence the tensor  $\sigma_{\alpha,\beta}$  is assumed to be diagonal with mutually equal elements  $\sigma_{\alpha,\alpha}$ . To calculate the infrared absorption, we need only the real part of the conductivity,

$$\text{Re } \sigma_{\alpha,\alpha}(\omega) = -\frac{\text{Im } \Pi_{\alpha,\alpha}^{ret}(\omega)}{\omega}. \tag{51}$$

Therefore our problem reduces to evaluating the current-current correlation function that in Matsubara frequencies is defined as

$$\Pi_{\alpha,\alpha}(i\omega_n) = \left(-\frac{1}{N}\right) \int_0^\beta d\tau e^{i\omega_n \tau} \langle T_\tau j_\alpha^\dagger(\tau) j_\alpha(0) \rangle, \tag{52}$$

where the current operator  $j_\alpha$  suitable for the Hamiltonian (1) is

$$j_\alpha = ite \sum_{i,\delta} \left(\frac{S_0^{i+\delta\hat{\alpha},i} + 1/2}{2S+1}\right) c_{i+\delta\hat{\alpha}}^\dagger c_i \tag{53}$$

and  $j_\alpha^\dagger(\tau) = e^{H\tau} j_\alpha^\dagger e^{-H\tau}$ . Performing the two canonical transformations (2),(3) and making the decoupling of the correlation function (52) in the electron, phonon, and spin terms through the introduction of  $H_{test}$  (9), we get

$$\begin{aligned} \Pi_{\alpha,\alpha}(i\omega_n) &= e^2 t^2 \left( -\frac{1}{N} \right) \sum_{i,\delta} \sum_{i',\delta'} (\delta \cdot \delta') \\ &\quad \times \int_0^\beta d\tau e^{i\omega_n \tau} \Delta(i, i', \delta, \delta') \\ &\quad \times \langle T_\tau \bar{c}_i^\dagger(\tau) \bar{c}_{i+\delta\hat{\alpha}}(\tau) c_{i'+\delta'\hat{\alpha}}^\dagger c_{i'} \rangle_t, \end{aligned} \quad (54)$$

where the function  $\Delta$ ,

$$\Delta(i, i', \delta, \delta') = \Phi(i, i', \delta, \delta') \Upsilon(i, i', \delta, \delta'), \quad (55)$$

denotes the product of the phonon correlation function  $\Phi$ ,

$$\Phi(i, i', \delta, \delta') = \langle T_\tau \bar{X}_i^\dagger(\tau) \bar{X}_{i+\delta\hat{\alpha}}(\tau) X_{i'+\delta'\hat{\alpha}}^\dagger X_{i'} \rangle_t \quad (56)$$

times the spin correlation function  $\Upsilon$ ,

$$\Upsilon(i, i', \delta, \delta') = \left\langle \left( \frac{S_0^{i, i+\delta\hat{\alpha}} + 1/2}{2S+1} \right) \left( \frac{S_0^{i'+\delta'\hat{\alpha}} + 1/2}{2S+1} \right) \right\rangle_t. \quad (57)$$

We calculate the electron correlation function

$$\begin{aligned} &\langle T_\tau \bar{c}_i^\dagger(\tau) \bar{c}_{i+\delta\hat{\alpha}}(\tau) c_{i'+\delta'\hat{\alpha}}^\dagger c_{i'} \rangle_t \\ &= \left( -\frac{1}{N^2} \right) \sum_{\mathbf{k}, \mathbf{k}_1} \exp[i\mathbf{k} \cdot \mathbf{R}_i e - i\mathbf{k}_1 \cdot (\mathbf{R}_i + \delta\hat{\alpha})] \\ &\quad \times \exp[i\mathbf{k}_1 \cdot (\mathbf{R}_{i'} + \delta'\hat{\alpha})] \\ &\quad \times e^{-i\mathbf{k} \cdot \mathbf{R}_{i'}} \mathcal{G}^{(0)}(\mathbf{k}, -\tau) \mathcal{G}^{(0)}(\mathbf{k}_1, \tau) \end{aligned} \quad (58)$$

and we separate  $\Delta$  into two terms,

$$\begin{aligned} \Delta(i, i', \delta, \delta') &= \Phi(i, i', \delta, \delta') \Upsilon(i, i', \delta, \delta') \\ &= [\langle X_i^\dagger X_{i+\delta\hat{\alpha}} \rangle_t]^2 \left\langle \left( \frac{S_0^{i, i+\delta\hat{\alpha}} + 1/2}{2S+1} \right) \right\rangle_t^2 \\ &\quad + \left\{ \Phi(i, i', \delta, \delta') \Upsilon(i, i', \delta, \delta') \right. \\ &\quad \left. - [\langle X_i^\dagger X_{i+\delta\hat{\alpha}} \rangle_t]^2 \left\langle \left( \frac{S_0^{i, i+\delta\hat{\alpha}} + 1/2}{2S+1} \right) \right\rangle_t^2 \right\}, \end{aligned} \quad (59)$$

hence

$$\begin{aligned} \Delta(i, i', \delta, \delta') &= e^{-2S_T} \left\langle \left( \frac{S_0 + 1/2}{2S+1} \right) \right\rangle^2 \\ &\quad + \left[ \Phi(i, i', \delta, \delta') \Upsilon(i, i', \delta, \delta') \right. \\ &\quad \left. - e^{-2S_T} \left\langle \left( \frac{S_0 + 1/2}{2S+1} \right) \right\rangle^2 \right]. \end{aligned} \quad (60)$$

To derive the optical properties, the role of the damping  $\Gamma$  of the particle motion is fundamental. Furthermore, the defect scattering rate  $\Gamma_0$  is added to the quantity  $\Gamma$  calculated in the previous section. This scattering rate  $\Gamma_0$  is assumed to be a small perturbation, thus it has been fixed equal to  $0.1\omega_0$ . The effect of the damping can enter our calculation substituting in Eq. (58)  $\mathcal{G}^{(0)}$  for  $\tilde{\mathcal{G}}$  given by Eq. (45).

Considering the two terms of Eq. (60), the correlation function can be written as

$$\Pi_{\alpha,\alpha}(i\omega_n) = \Pi_{\alpha,\alpha}^{(1)}(i\omega_n) + \Pi_{\alpha,\alpha}^{(2)}(i\omega_n). \quad (61)$$

The first term reads

$$\begin{aligned} \Pi_{\alpha,\alpha}^{(1)}(i\omega_n) &= 4e^2 t^2 e^{-2S_T} \left\langle \left( \frac{S_0 + 1/2}{2S+1} \right) \right\rangle^2 \left( \frac{1}{N} \right) \sum_{\mathbf{k}} \sin^2(k_\alpha) \\ &\quad \times \int_0^\beta d\tau e^{i\omega_n \tau} \tilde{\mathcal{G}}(\mathbf{k}, -\tau) \tilde{\mathcal{G}}(\mathbf{k}, \tau) \end{aligned} \quad (62)$$

and the second one is obtained retaining only the main auto-correlation term  $i=i'$  and  $\delta=\delta'$ ,

$$\begin{aligned} \Pi_{\alpha,\alpha}^{(2)}(i\omega_n) &= \left( \frac{2e^2}{Z} \right) \left\langle \left( \frac{S_0 + 1/2}{2S+1} \right) \right\rangle^2 \left( \frac{1}{N^2} \right) \\ &\quad \times \sum_{\mathbf{k}, \mathbf{k}_1} \int_0^\beta d\tau e^{i\omega_n \tau} \tilde{\mathcal{G}}(\mathbf{k}, -\tau) \tilde{\mathcal{G}}(\mathbf{k}_1, \tau) f_1(\tau), \end{aligned} \quad (63)$$

where  $f_1(\tau)$  is given by Eq. (34). We stress that substituting  $\mathcal{G}^{(0)}$  for  $\tilde{\mathcal{G}}$  is necessary to have a nonvanishing  $\Pi_{\alpha,\alpha}^{(1)}(i\omega_n)$ . We perform the analytic continuation  $i\omega_n \rightarrow \omega + i\delta$ , and, clearly, the conductivity can be expressed as sum of two terms,

$$\begin{aligned} \text{Re } \sigma_{\alpha,\alpha}(\omega) &= -\frac{\text{Im}[\Pi_{\alpha,\alpha}^{ret(1)}(\omega) + \Pi_{\alpha,\alpha}^{ret(2)}(\omega)]}{\omega} \\ &= \text{Re } \sigma_{\alpha,\alpha}^{(band)}(\omega) + \text{Re } \sigma_{\alpha,\alpha}^{(incoh)}(\omega). \end{aligned} \quad (64)$$

The first term  $\text{Re } \sigma_{\alpha,\alpha}^{(band)}$  represents the band conductivity. Here the charge transfer is affected by the damping due to interactions with the lattice and spin fluctuations, but it is not accompanied by processes changing the number of phonons. On the other hand, the second term  $\text{Re } \sigma_{\alpha,\alpha}^{(incoh)}$  in Eq. (64) derives from inelastic scattering processes of emission and absorption of phonons and it is marked by the apex ‘‘incoherent’’ since the particles lose their phase coherence be-

cause of this phonon-assisted hopping. As in the spectral properties, we stress the appearance of two physically distinct contributions.

In appendix we report the details of calculation for the optical conductivity. The band conductivity is derived as

$$\begin{aligned} \text{Re } \sigma_{\alpha,\alpha}^{(band)}(\omega) &= \left( \frac{4e^2 t^2}{\omega} \right) e^{-2S_T} \left\langle \left( \frac{S_0 + 1/2}{2S + 1} \right) \right\rangle^2 \\ &\times \int_{-\infty}^{+\infty} d\xi [n_F(\xi - \omega) - n_F(\xi)] \tilde{C}(\xi, \omega) h(\xi), \end{aligned} \quad (65)$$

where  $\tilde{C}(\xi, \omega)$  is

$$\tilde{C}(\xi, \omega) = \frac{\Gamma(\xi)}{\Gamma^2(\xi) + \omega^2} \quad (66)$$

and  $h(\xi)$  reads

$$h(\xi) = \left( \frac{1}{N} \right) \sum_{\mathbf{k}} \sin^2(k_\alpha) \delta(\xi - \xi_{\mathbf{k}}). \quad (67)$$

The latter term of the conductivity becomes

$$\begin{aligned} \text{Re } \sigma_{\alpha,\alpha}^{(incoh)}(\omega) &= \left( \frac{2e^2 t^2}{\omega} \right) e^{-2S_T} \left\langle \left( \frac{S_0 + 1/2}{2S + 1} \right)^2 \right\rangle \\ &\times \int_{-\infty}^{+\infty} d\xi \int_{-\infty}^{+\infty} d\xi_1 g(\xi) g(\xi_1) R(\xi, \xi_1, \omega) \\ &+ \left( \frac{2e^2 t^2}{\omega} \right) e^{-2S_T} \left[ I_0(z) \left\langle \left( \frac{S_0 + 1/2}{2S + 1} \right)^2 \right\rangle \right. \\ &\left. - \left\langle \left( \frac{S_0 + 1/2}{2S + 1} \right) \right\rangle^2 \right] \\ &\times \int_{-\infty}^{+\infty} d\xi \int_{-\infty}^{+\infty} d\xi_1 g(\xi) g(\xi_1) [n_F(\xi - \omega) \\ &- n_F(\xi)] C(\xi, \xi_1, \omega), \end{aligned} \quad (68)$$

where  $g(\xi)$  is the density of states (16) and the function  $R(\xi, \xi_1, \omega)$  is given by

$$\begin{aligned} R(\xi, \xi_1, \omega) &= 2 \sum_{l=1}^{+\infty} I_l(z) \sinh \left( \frac{\beta \bar{\omega}_0 l}{2} \right) [J_l(\xi, \xi_1, \omega) \\ &+ H_l(\xi, \xi_1, \omega)]. \end{aligned} \quad (69)$$

We notice that  $J_l(\xi, \xi_1, \omega)$ ,

$$\begin{aligned} J_l(\xi, \xi_1, \omega) &= C(\xi, \xi_1, \omega + l\bar{\omega}_0) [n_F(\xi - l\bar{\omega}_0 - \omega) \\ &- n_F(\xi - l\bar{\omega}_0)] [N_0(l\bar{\omega}_0) + n_F(\xi)], \end{aligned} \quad (70)$$

and  $H_l(\xi, \xi_1, \omega)$ ,

$$\begin{aligned} H_l(\xi, \xi_1, \omega) &= C(\xi, \xi_1, \omega - l\bar{\omega}_0) [n_F(\xi + l\bar{\omega}_0 - \omega) \\ &- n_F(\xi + l\bar{\omega}_0)] [N_0(l\bar{\omega}_0) + 1 - n_F(\xi)], \end{aligned} \quad (71)$$

describe phonon absorption and emission processes, respectively.

In the limit of high temperatures ( $T > 0.39\omega_0$ ) and for small polaron excitations, the incoherent absorption is prevalent. In this case the conductivity consists of a sum of narrow Lorentzian functions centered on the points  $n\omega_0$ , respectively.<sup>39,43</sup> We perform an expansion suitable for high temperatures and in appendix we derive the approximate expression of the conductivity,

$$\begin{aligned} \text{Re } \sigma_{\alpha,\alpha}(\omega) &= \left( \frac{e^2 t^2}{\omega} \right) \left\langle \left( \frac{S_0 + 1/2}{2S + 1} \right)^2 \right\rangle \sqrt{\frac{\pi\beta}{z}} \\ &\times \left\{ \exp \left[ -\frac{\beta}{4z} (\omega - \bar{z})^2 \right] \right. \\ &\left. - \exp \left[ -\frac{\beta}{4z} (\omega + \bar{z})^2 \right] \right\} \rho(1 - \rho), \end{aligned} \quad (72)$$

where  $\rho$  is the electron concentration.

We have checked the internal consistency of our approach by means of the sum rule

$$\int_0^\infty d\omega \text{Re } \sigma_{\alpha,\alpha}(\omega) = -\frac{\pi}{2} e^2 \langle \hat{T}_{\alpha,\alpha} \rangle, \quad (73)$$

where  $\hat{T}_{\alpha,\alpha}$  is the component of the kinetic energy. The left-hand side of Eq. (73) is calculated by using the conductivity spectra, whereas the mean value  $\langle \hat{T}_{\alpha,\alpha} \rangle$  can be evaluated by performing the two canonical transformations (2),(3) and making the decoupling in the electron, phonon, and spin terms through the introduction of  $H_{test}$  (9). We get

$$\langle \hat{T}_{\alpha,\alpha} \rangle = \frac{1}{3} \int_{-\infty}^{+\infty} d\xi g(\xi) \xi n_F(\xi) \quad (74)$$

realizing that the two sides of Eq. (73) differ by a few percent.

In Fig. 6 we report the calculated conductivity spectra for  $x=0.3$  at different temperatures. In the large polaron ferromagnetic phase,  $\sigma$  is given by Eq. (64), in the small polaron paramagnetic phase by Eq. (72), in the regime of coexisting phases by the two preceding conductivities by means of the Eq. (18). With rising temperature, a transfer of spectral weight from low to high energies takes place. The crossover energy can be estimated about  $\omega_c = 9\omega_0$ . When the temperature increases, the large polaron Drude term makes smaller and at  $T = T_c$  the optical response is characterized by the small-polaron absorption band peaked approximately around  $2g^2\omega_0$ . Thus changes in the quasiparticle excitations of the system are traceable in the optical response that, for  $T > T_c$ , show the opening of an optical gap. At low temperatures the conductivity shows a clear Drude peak below  $\omega_0$

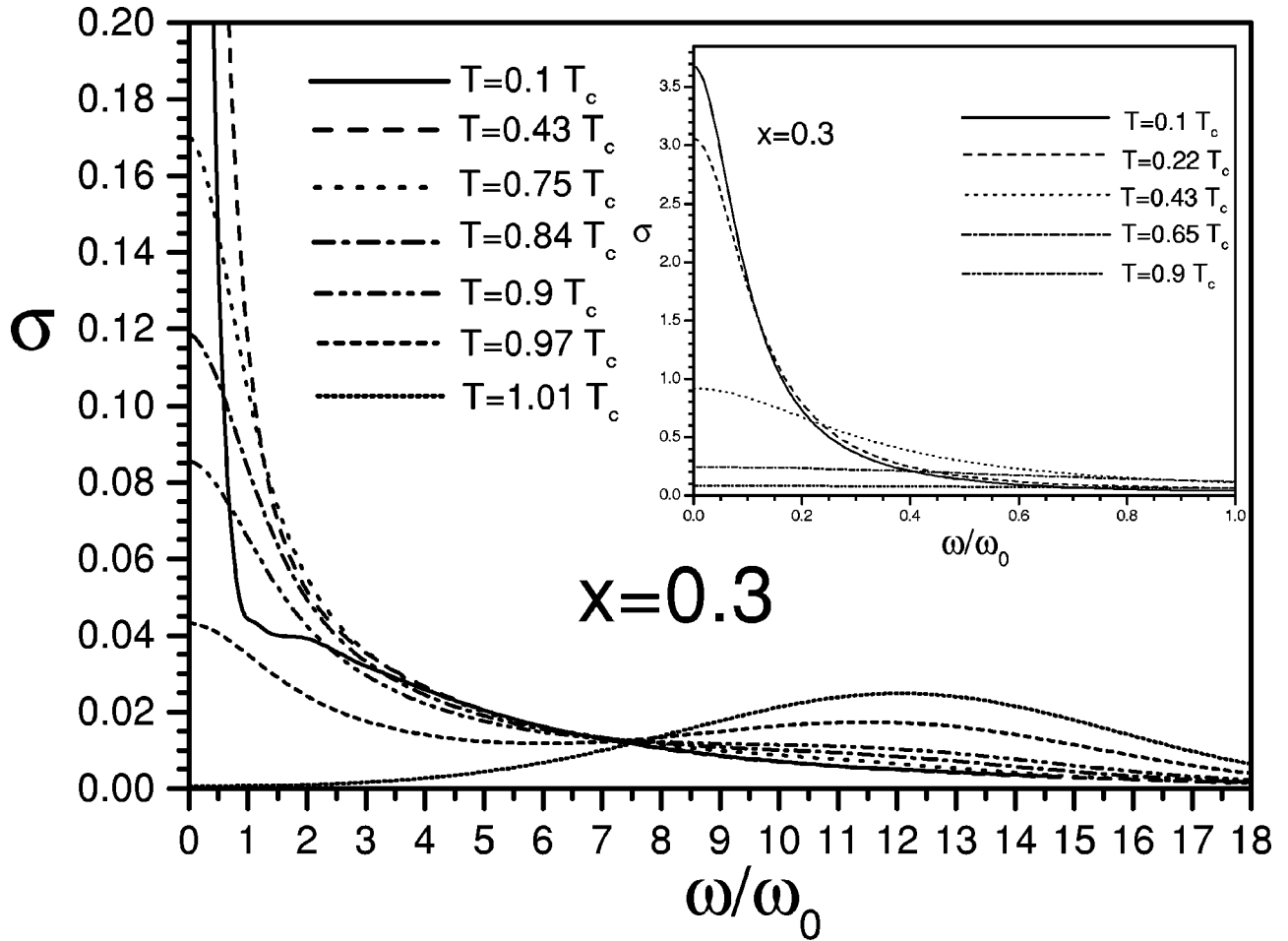


FIG. 6. The conductivity up to  $18 \omega_0$  at different temperatures. In the inset the conductivity up to  $1 \omega_0$  at different temperatures. The conductivities are expressed in units of  $e^2 c / m \omega_0$ , with  $c$  hole concentration and  $m = 1/2t$ , and they are obtained for  $t = 2\omega_0$  and  $g = 2.8$ .

and a quiet asymmetric absorption band with a long tail above its peak position at higher energies. These two types of optical response are due, respectively, to a coherent motion and an incoherent absorption band of large polarons. In the inset of Fig. 6 the Drude peak is shown. As  $T$  decreases, the coherent response increases continuously. We stress that the results shown in this figure are all consistent with experimental spectra.<sup>12,13</sup>

In Fig. 7 at  $T = 0.1 T_c$  the optical conductivity is decomposed into two components: band (dotted line) and incoherent conductivity (dashed line). It is worthwhile to notice that the band conductivity provides also the main contribution to the polaronic absorption band. In the inset of Fig. 7 the Drude term is fully shown. At low temperatures it can be obtained by means of the equation

$$\text{Re } \sigma_{\text{Drude}}(\omega) = A \frac{\Gamma(\mu)}{\Gamma^2(\mu) + \omega^2}, \quad (75)$$

where  $A$  is a proportionality constant and  $\Gamma(\mu)$  is the scattering rate of the preceding section calculated at  $\mu$ . This inset emphasizes the spectral weight of the absorption band too.

In order to be more quantitative in the comparison with experimental data, we have evaluated the effective carrier number below the crossover energy  $\omega_c$ ,

$$N_{\text{eff}}(\omega) = \frac{2m_e}{\pi e^2 N_{\text{Mn}}} \int_0^{\omega_c} d\omega \text{Re } \sigma_{\alpha,\alpha}(\omega), \quad (76)$$

that is proportional to the spectral weight below this cutoff energy [see Fig. 8(a)]. In Eq. (76)  $m_e$  is the free electron mass,  $e$  is the electron charge, and  $N_{\text{Mn}}$  is the number of Mn atoms per unit volume. As  $T$  increases,  $N_{\text{eff}}$  decreases assuming a nearly constant value for  $T > T_c$ . We stress that the calculated quantity (circles) agrees with the experimental effective carrier number deduced by Refs. 12,13 and reported in the same figure (diamonds). We note that the agreement gets worse for  $T > T_c$ . Indeed, within this temperature range, in addition to the small-polaron band, other terms, such as interband transitions between the exchange-split conduction bands,<sup>28</sup> could contribute to the optical absorption.

We have also estimated the quantity  $\langle \omega \rangle$  (the first moment),

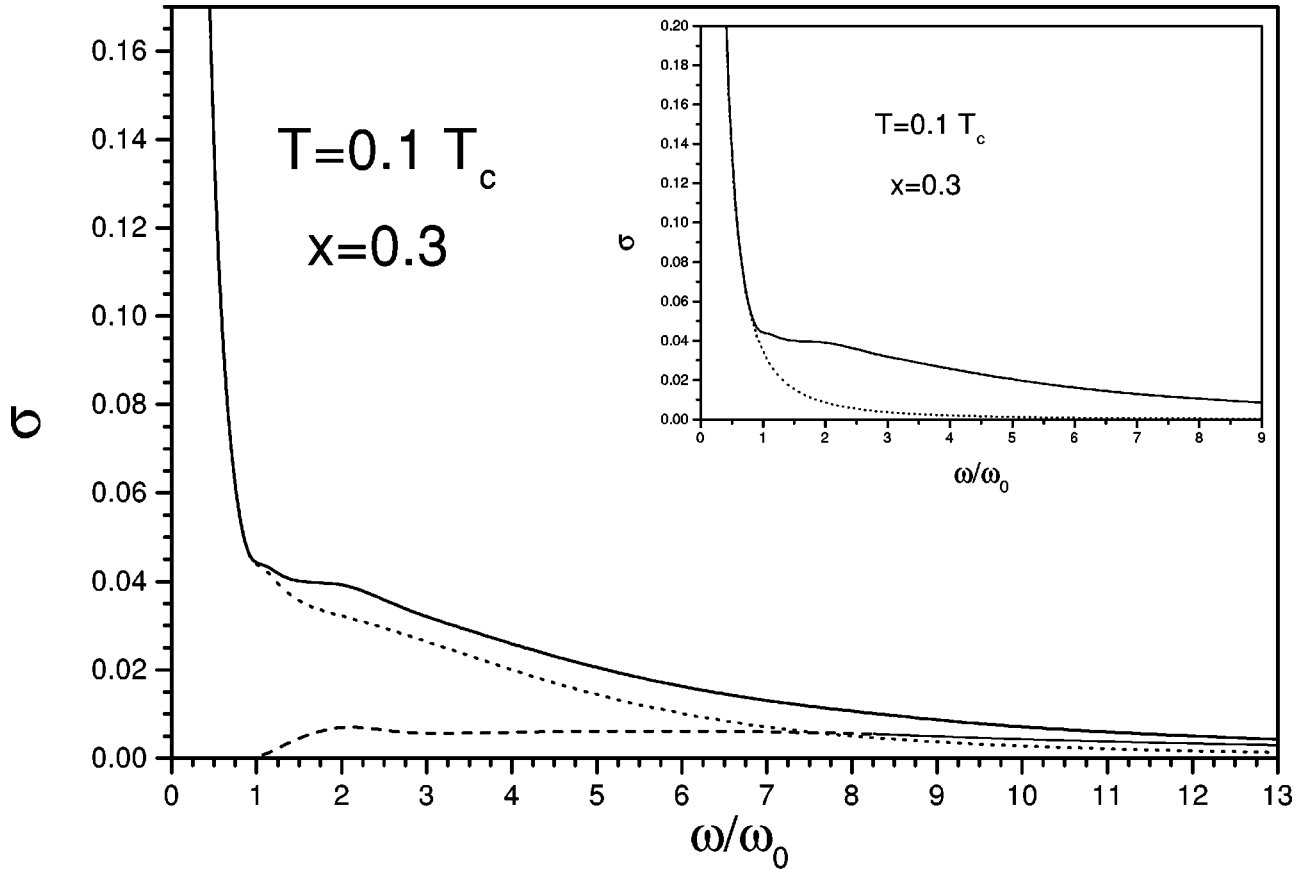


FIG. 7. The conductivity (solid line), the band conductivity (dotted line), and the “incoherent” conductivity (dashed line) at  $T=0.1T_c$  in units of  $e^2c/m\omega_0$ , where  $c$  is the hole concentration and  $m=1/2t$ . In the inset the conductivity (solid line) and the Drude term (dotted line) at  $T=0.1T_c$  in the same units of the figure.

$$\langle \omega \rangle = \frac{\int_0^\infty d\omega \omega \operatorname{Re} \sigma_{\alpha,\alpha}(\omega)}{\int_0^\infty d\omega \operatorname{Re} \sigma_{\alpha,\alpha}(\omega)}, \quad (77)$$

as a function of the temperature [see Fig. 8(b)]. It measures the characteristic energy of the polaron excitations.<sup>44</sup> Thus  $\langle \omega \rangle$  assumes small values in the large-polaron phase at low temperatures and increases gradually in the phase separation regime tending to the value  $g^2\omega_0$  in the small-polaron phase at high temperatures.

Finally in Fig. 9 we plot the resistivity  $\rho=1/\sigma$ , where  $\sigma$  results from the conductivity  $\sigma(\omega)$  in the limit  $\omega \rightarrow 0$ . We obtain a further confirmation of two different behaviors: metallic ( $d\rho/dT > 0$ ) and insulating ( $d\rho/dT < 0$ ). In logarithmic scale the behavior of activated resistivity<sup>45</sup> for  $T > T_c$  is emphasized.

## V. SUMMARY AND CONCLUSIONS

We have discussed spectral and optical properties mainly for  $x=0.3$  and as functions of the temperature. The polaron formation and the small-to-large polaron crossover through the phase separation regime turn out to play a crucial role in understanding the properties of manganites.

It has been stressed that the phase separation regime induces a robust pseudogap in the excitation spectrum of the system. Furthermore, the pseudogap features show a direct relationship with the metal-insulator transition and with the crossover from the coherent large-polaron dynamics to the incoherent small-polaron one. Our results turn out to be compatible with the spectral weights found experimentally.<sup>10,11</sup>

With regard to the infrared absorption, we have observed that, with decreasing  $T$ , our optical spectra are characterized by a transfer of spectral weight from high to low energies filling up the low-frequency optical gap present in the high-temperature phase.

In the metallic ferromagnetic phase, at low temperatures, the system shows two types of optical response: a narrow Drude term and a broad absorption band due respectively to the coherent and incoherent motion of large polarons. These results obtained at low  $T$  are consistent with experimental conductivity spectra.<sup>12,13,28</sup> The agreement improves when a temperature-independent part is extracted from the experimental optical spectra.<sup>28,46</sup> This part is considered to be due to the “background” interband transitions between the O  $2p$  and Mn  $3d$  band. The reduced optical conductivity obtained by subtracting the temperature-independent term<sup>28</sup> points out that the broad band at low  $T$  is very similar to that shown in this paper. We also stress that orbital degrees of freedom could be responsible for the effect of enhancing the incoher-



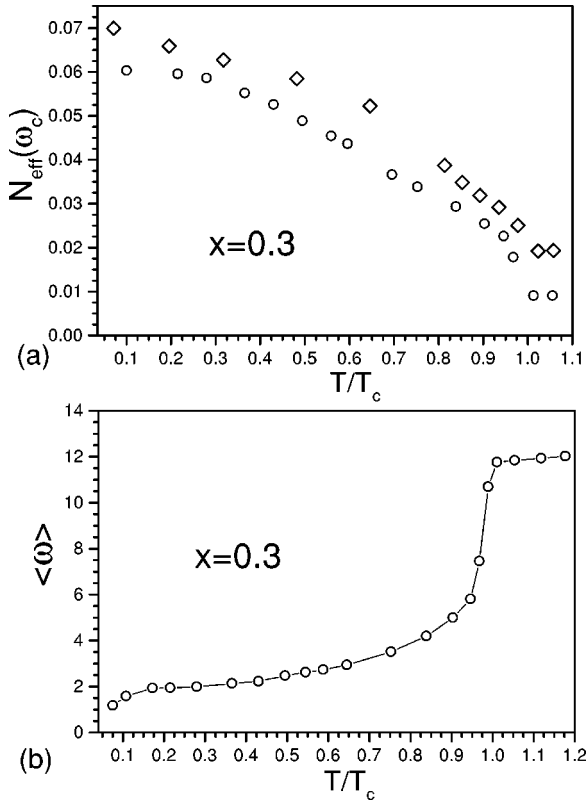


FIG. 8. (a) The effective carrier number (circles) below the crossover energy  $\omega_c$  compared to the experimental effective carrier number deduced by Refs. 12,13 as a function of the temperature. (b) The first moment (in units of  $\omega_0$ ) as a function of the temperature. The results are obtained for  $t=2\omega_0$  and  $g=2.8$ , in the case (a) specifying  $\omega_0=50$  meV and the lattice constant  $a=0.4$  nm.

ent absorption.<sup>47,48</sup> On the other hand, we observe that electronic correlations can change the spectral properties but are somehow obscured in the optical conductivity within the ferromagnetic metallic phase.<sup>49</sup>

At high temperatures, the infrared absorption is due to the incoherent small-polaron motion. Spectroscopic evidences of

the small-polaron formation have been reported in the high- $T$  paramagnetic phase.<sup>14,15</sup> However for frequencies higher than the peak of small-polaron absorption band, the effects due to the exchange-split bands and to the local Coulomb repulsion cannot be neglected.<sup>50</sup> Indeed the experimental data can be fitted reasonably well with the small-polaron band below 0.8 eV.<sup>12</sup> Thus we can consider our results meaningful for frequencies up to the absorption peak.

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#### APPENDIX

In this appendix we report on the calculations leading to the expressions of the optical conductivity

$$\text{Re } \sigma_{\alpha,\alpha}(\omega) = \text{Re } \sigma_{\alpha,\alpha}^{(band)}(\omega) + \text{Re } \sigma_{\alpha,\alpha}^{(incoh)}(\omega). \quad (\text{A1})$$

This decomposition originates from the current-current correlation function that in Matsubara frequencies can be written as

$$\Pi_{\alpha,\alpha}(i\omega_n) = \Pi_{\alpha,\alpha}^{(1)}(i\omega_n) + \Pi_{\alpha,\alpha}^{(2)}(i\omega_n), \quad (\text{A2})$$

where the first term is

$$\begin{aligned} \Pi_{\alpha,\alpha}^{(1)}(i\omega_n) = & 4e^2 t^2 e^{-2S\tau} \left\langle \left( \frac{S_0 + 1/2}{2S + 1} \right) \right\rangle^2 \left( \frac{1}{N} \right) \\ & \times \sum_{\mathbf{k}} \sin^2(k_\alpha) \int_0^\beta d\tau e^{i\omega_n \tau} \tilde{\mathcal{G}}(\mathbf{k}, -\tau) \tilde{\mathcal{G}}(\mathbf{k}, \tau) \end{aligned} \quad (\text{A3})$$

and the second one reads

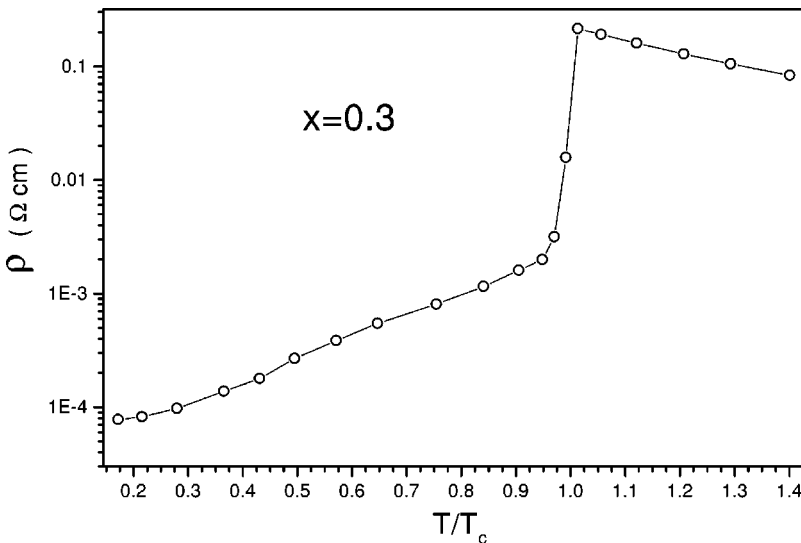


FIG. 9. The resistivity as a function of the temperature (we have used  $\omega_0=50$  meV and the lattice constant  $a=0.4$  nm).

$$\begin{aligned} \Pi_{\alpha,\alpha}^{(2)}(i\omega_n) &= \left( \frac{2e^2}{Z} \right) \left\langle \left( \frac{S_0+1/2}{2S+1} \right) \right\rangle^2 \left( \frac{1}{N^2} \right) \\ &\times \sum_{\mathbf{k}, \mathbf{k}_1} \int_0^\beta d\tau e^{i\omega_n \tau} \tilde{\mathcal{G}}(\mathbf{k}, -\tau) \tilde{\mathcal{G}}(\mathbf{k}_1, \tau) f_1(\tau) \end{aligned} \quad (\text{A4})$$

with  $f_1(\tau)$  given by Eq. (34). Adopting the Lehmann representation, the spectral function  $\tilde{A}(\mathbf{k}, \omega)$  can be introduced, therefore, making the analytic continuation  $i\omega_n \rightarrow \omega + i\delta$ , the band conductivity becomes

$$\begin{aligned} \text{Re } \sigma_{\alpha,\alpha}^{(band)}(\omega) &= - \frac{\text{Im } \Pi_{\alpha,\alpha}^{(1)ret}(\omega)}{\omega} \\ &= \left( \frac{4e^2 t^2}{\omega} \right) e^{-2S_T} \left\langle \left( \frac{S_0+1/2}{2S+1} \right) \right\rangle^2 \left( \frac{1}{N} \right) \\ &\times \sum_{\mathbf{k}} \sin^2(k_\alpha) \tilde{D}(\mathbf{k}), \end{aligned} \quad (\text{A5})$$

where  $\tilde{D}(\mathbf{k}) = D(\mathbf{k}, \mathbf{k})$  with

$$\begin{aligned} D(\mathbf{k}, \mathbf{k}_1) &= \int_{-\infty}^{+\infty} \frac{d\omega'}{4\pi} \tilde{A}(\mathbf{k}, \omega') \tilde{A}(\mathbf{k}_1, \omega' - \omega) \\ &\times [n_F(\omega' - \omega) - n_F(\omega')]. \end{aligned} \quad (\text{A6})$$

We also derive the latter term of the conductivity,

$$\begin{aligned} \text{Re } \sigma_{\alpha,\alpha}^{(incoh)}(\omega) &= - \frac{\text{Im } \Pi_{\alpha,\alpha}^{(2)ret}(\omega)}{\omega} \\ &= \left( \frac{2e^2 t^2}{\omega} \right) e^{-2S_T} \left\langle \left( \frac{S_0+1/2}{2S+1} \right)^2 \right\rangle \\ &\times \left( \frac{1}{N^2} \right) \sum_{\mathbf{k}, \mathbf{k}_1} f(\mathbf{k}, \mathbf{k}_1) + \left( \frac{2e^2 t^2}{\omega} \right) e^{-2S_T} \\ &\times \left[ I_0(z) \left\langle \left( \frac{S_0+1/2}{2S+1} \right)^2 \right\rangle - \left\langle \left( \frac{S_0+1/2}{2S+1} \right) \right\rangle^2 \right] \\ &\times \left( \frac{1}{N^2} \right) \sum_{\mathbf{k}, \mathbf{k}_1} D(\mathbf{k}, \mathbf{k}_1), \end{aligned} \quad (\text{A7})$$

where

$$f(\mathbf{k}, \mathbf{k}_1) = 2 \sum_{l=1}^{+\infty} I_l(z) \sinh\left( \frac{\beta \bar{\omega}_0 l}{2} \right) [J_l(\mathbf{k}, \mathbf{k}_1, \omega) + H_l(\mathbf{k}, \mathbf{k}_1, \omega)] \quad (\text{A8})$$

with

$$\begin{aligned} J_l(\mathbf{k}, \mathbf{k}_1, \omega) &= \int_{-\infty}^{+\infty} \frac{d\omega'}{4\pi} \tilde{A}(\mathbf{k}, \omega') \tilde{A}(\mathbf{k}_1, \omega' - l\bar{\omega}_0 - \omega) \\ &\times [n_F(\omega' - l\bar{\omega}_0 - \omega) - n_F(\omega' - l\bar{\omega}_0)] \\ &\times [N_0(l\bar{\omega}_0) + n_F(\omega')] \end{aligned} \quad (\text{A9})$$

and

$$\begin{aligned} H_l(\mathbf{k}, \mathbf{k}_1, \omega) &= \int_{-\infty}^{+\infty} \frac{d\omega'}{4\pi} \tilde{A}(\mathbf{k}, \omega') \tilde{A}(\mathbf{k}_1, \omega' + l\bar{\omega}_0 - \omega) \\ &\times [n_F(\omega' + l\bar{\omega}_0 - \omega) - n_F(\omega' + l\bar{\omega}_0)] \\ &\times [N_0(l\bar{\omega}_0) + 1 - n_F(\omega')]. \end{aligned} \quad (\text{A10})$$

We observe that  $J_l(\mathbf{k}, \mathbf{k}_1, \omega)$  and  $H_l(\mathbf{k}, \mathbf{k}_1, \omega)$  describe phonon absorption and emission processes, respectively. Using the convolution product of Lorentzian functions, the band conductivity becomes

$$\begin{aligned} \text{Re } \sigma_{\alpha,\alpha}^{(band)}(\omega) &= \left( \frac{4e^2 t^2}{\omega} \right) e^{-2S_T} \left\langle \left( \frac{S_0+1/2}{2S+1} \right) \right\rangle^2 \\ &\times \int_{-\infty}^{+\infty} d\xi [n_F(\xi - \omega) - n_F(\xi)] \tilde{C}(\xi, \omega) h(\xi), \end{aligned} \quad (\text{A11})$$

where  $\tilde{C}(\xi, \omega) = C(\xi, \xi, \omega)$  with

$$\begin{aligned} C(\xi, \xi_1, x) &= \int_{-\infty}^{+\infty} \frac{d\omega'}{4\pi} \tilde{A}(\xi, \omega') \tilde{A}(\xi_1, \omega' - x) \\ &= \frac{1}{2} \frac{[\Gamma(\xi) + \Gamma(\xi_1)]}{[\Gamma(\xi) + \Gamma(\xi_1)]^2/4 + (\xi_1 - \xi + x)^2} \end{aligned} \quad (\text{A12})$$

and

$$h(\xi) = \left( \frac{1}{N} \right) \sum_{\mathbf{k}} \sin^2(k_\alpha) \delta(\xi - \xi_{\mathbf{k}}). \quad (\text{A13})$$

Since  $\psi(y)$ , the Fourier transform of  $h(\xi)$ , is calculated as

$$\psi(y) = \int_{-\infty}^{+\infty} d\xi e^{iy\xi} h(\xi) = \frac{[J_0(2yt_{eff})]^2 J_1(2yt_{eff})}{2yt_{eff}}, \quad (\text{A14})$$

we can express  $h(\xi)$  in the following manner

$$h(\xi) = \frac{1}{2\pi t_{eff}} \int_{-\infty}^{+\infty} dy \cos\left( \frac{\xi}{2t_{eff}} \right) \frac{[J_0(y)]^2 J_1(y)}{y}. \quad (\text{A15})$$

On the other hand, the latter part of the conductivity reads

$$\begin{aligned} \text{Re } \sigma_{\alpha,\alpha}^{(incoh)}(\omega) &= \left( \frac{2e^2 t^2}{\omega} \right) e^{-2S_T} \left\langle \left( \frac{S_0+1/2}{2S+1} \right)^2 \right\rangle \\ &\times \int_{-\infty}^{+\infty} d\xi \int_{-\infty}^{+\infty} d\xi_1 g(\xi) g(\xi_1) R(\xi, \xi_1, \omega) \\ &+ \left( \frac{2e^2 t^2}{\omega} \right) e^{-2S_T} \left[ I_0(z) \left\langle \left( \frac{S_0+1/2}{2S+1} \right)^2 \right\rangle \right. \\ &\left. - \left\langle \left( \frac{S_0+1/2}{2S+1} \right) \right\rangle^2 \right] \end{aligned}$$

$$\times \int_{-\infty}^{+\infty} d\xi \int_{-\infty}^{+\infty} d\xi_1 g(\xi) g(\xi_1) [n_F(\xi - \omega) - n_F(\xi)] C(\xi, \xi_1, \omega), \quad (\text{A16})$$

where  $g(\xi)$  is the density of states (16) and the function  $R(\xi, \xi_1, \omega)$  is given by

$$R(\xi, \xi_1, \omega) = 2 \sum_{l=1}^{+\infty} I_l(z) \sinh\left(\frac{\beta \bar{\omega}_0 l}{2}\right) [J_l(\xi, \xi_1, \omega) + H_l(\xi, \xi_1, \omega)] \quad (\text{A17})$$

with

$$J_l(\xi, \xi_1, \omega) = C(\xi, \xi_1, \omega + l\bar{\omega}_0) [n_F(\xi - l\bar{\omega}_0 - \omega) - n_F(\xi - l\bar{\omega}_0)] [N_0(l\bar{\omega}_0) + n_F(\xi)] \quad (\text{A18})$$

and

$$H_l(\xi, \xi_1, \omega) = C(\xi, \xi_1, \omega - l\bar{\omega}_0) [n_F(\xi + l\bar{\omega}_0 - \omega) - n_F(\xi + l\bar{\omega}_0)] [N_0(l\bar{\omega}_0) + 1 - n_F(\xi)]. \quad (\text{A19})$$

In the limit of high temperatures, we concentrate on  $f_1(\tau)$  of Eq. (A4) carrying out the following approximation,

$$e^{-2s\tau} \exp\left\{z \cosh\left[\bar{\omega}_0 \left(\tau - \frac{\beta}{2}\right)\right]\right\} \approx \exp(-\bar{z}\tau) \exp\left(\frac{\bar{z}}{\beta} \tau^2\right), \quad (\text{A20})$$

where

$$\bar{z} = 2\bar{s} \approx 2g^2 \omega_0. \quad (\text{A21})$$

This expansion is valid as long as  $(\beta\bar{z}/12)(\beta\bar{\omega}_0/4)^2 < 1$ , hence  $T > 0.39\omega_0$ . The response of the small-polaron excitations is characterized by the incoherent behavior, therefore the conductivity becomes

$$\begin{aligned} \text{Re } \sigma_{\alpha, \alpha}(\omega) &= \left(\frac{e^2 t^2}{\omega}\right) \left\langle \left(\frac{S_0 + 1/2}{2S + 1}\right)^2 \right\rangle \sqrt{\frac{\pi\beta}{\bar{z}}} \left(\frac{1}{N^2}\right) \\ &\times \sum_{\mathbf{k}, \mathbf{k}_1} \int_{-\infty}^{+\infty} \frac{d\omega'}{2\pi} \int_{-\infty}^{+\infty} \frac{d\omega''}{2\pi} \tilde{A}(\mathbf{k}, \omega') \tilde{A}(\mathbf{k}_1, \omega'') \\ &\times [1 - n_F(\omega'')] n_F(\omega') \left\{ \exp\left[-\frac{\beta}{4\bar{z}}(\omega + \omega' - \omega'' - \bar{z})^2\right] - \exp\left[-\frac{\beta}{4\bar{z}}(\omega + \omega' - \omega'' + \bar{z})^2\right] \right\}. \end{aligned} \quad (\text{A22})$$

We consider  $\xi_{\mathbf{k}} \approx -g^2 \omega_0 - \mu$  obtaining

$$\begin{aligned} \text{Re } \sigma_{\alpha, \alpha}(\omega) &= \left(\frac{e^2 t^2}{\omega}\right) \left\langle \left(\frac{S_0 + 1/2}{2S + 1}\right)^2 \right\rangle \sqrt{\frac{\pi\beta}{\bar{z}}} \left\{ \exp\left[-\frac{\beta}{4\bar{z}}(\omega - \bar{z})^2\right] - \exp\left[-\frac{\beta}{4\bar{z}}(\omega + \bar{z})^2\right] \right\} \rho(1 - \rho), \end{aligned} \quad (\text{A23})$$

where  $\rho$  is the electron concentration.

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