The essential interactions in oxides and spectral weight transfer in doped manganites

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We calculate the value of the Fröhlich electron-phonon interaction in manganites, cuprates, and some other charge-transfer insulators and show that this interaction is much stronger than any relevant magnetic interaction. A polaron shift due to the Fröhlich interaction, which is about 1 eV, suggests that carriers in those systems are small (bi)polarons at all temperatures and doping levels, in agreement with the oxygen isotope effect and other data. An opposite conclusion, recently suggested in the literature, is shown to be incorrect. The frequency and temperature dependence of the optical conductivity of ferromagnetic manganites is explained within the framework of the bipolaron theory.

σ₀ = 360Ω⁻¹cm⁻¹ [18] yields the product of the Fermi wave vector and the mean free path kpℓ ≤ 1, which is below the Ioffe-Regel limit [21]. Hence, Fermi-liquid type theories appear to be inadequate even for a description of the low-temperature phase of manganites.

Among the major phenomena yet to be explained in the manganites are the spectral weight transfer with temperature [16–18] and the pronounced peak structure [18] in the optical conductivity both above and below Tc, observed in several systems.

In this paper we first calculate the value of the electron-phonon interaction in oxides. We then propose a theory of the optical conductivity, including the massive spectral weight transfer below the ferromagnetic transition, based on the idea of the current carrier density collapse. We show that the high-temperature optical conductivity is well described by the small polaron absorption, while the low temperature midinfrared band is due to absorption by small polarons. The bipolaron dissociation below Tc shifts the spectral weight from the bipolaronic peak to the polaronic one. We describe the optical spectra of the layered ferromagnetic (Tc = 125K) crystal La2−xSr1+2xMn2O7 [18] in the entire frequency and temperature range that has been studied experimentally, and show that the optical data provides strong evidence for CCDC.

I. INTRODUCTION

It is well understood that carriers in cuprates [1,2] and manganites [3,4] are strongly coupled to lattice vibrations. We have recently proposed a bipolaron theory of ferromagnetism and colossal magnetoresistance (CMR) based on the idea of a current carrier density collapse (CCDC) due to an interplay of the electron-phonon and exchange interactions in doped manganites [5]. Owing to the strong electron-phonon interaction, polaronic carriers are bound into almost immobile bipolarons in the paramagnetic phase of CMR materials. The non-degenerate polarons induce a polarization of localized Mn magnetic phase of CMR materials. The non-degenerate level is almost degenerate into an almost immobile bipolaron in the paramagnetic phase of CMR materials.
contrary to recent claims in the literature [24].

In order to assess the possibility of small polaron and bipolaron formation in manganites and cuprates, one has to calculate the electron-phonon interaction and compare it with the (inter-site) Coulomb repulsion. Fortunately, such an estimate is possible in oxides, which are highly polarizable materials with a substantial long-range Fröhlich interaction. Then the polaron binding energy \( E_p \) (polaronic shift) can be explicitly expressed through the well defined experimental parameters. In the long-wave limit the response of polarons at the optical phonon frequency is dynamic [24]. Their (renormalized) plasma frequency is lower than the optical phonon frequency due to the large static dielectric constant of oxides, an enhanced effective mass, and relatively low density of polarons. Therefore, the long-range character of the Fröhlich interaction is unaffected by screening. The optical phonon frequency is hardly affected as well [25]. Claims in the literature [26,27] that the Fröhlich interaction is unaffected by screening.

The correct value of\( E_p \) appears to be about 1eV in most cases, far exceeding the ad hoc estimates of \(~ 0.15 \text{ eV}\) [21], obtained from a numerically incorrect expression for \( E_p \) and wrong values of the dielectric constants. The large calculated value of the polaron shift (\(~ 1 \text{ eV}\)) is perfectly compatible with the small polaron theory. According to \[1\] the bare half-bandwidth is about \( W/2 \approx 0.8 \text{ eV} \) in manganites. Hence, even a simple variational criterion of the small polaron formation \( (E_p > W/2) \) is satisfied. In the case of the Fröhlich interaction the small polaron theory based on the Lang-Firsov canonical transformation is numerically accurate even in the weak coupling (large polaron) regime irrespective of the value of \( E_p \) [24]. It is much more important, irrespective of any theoretical arguments, that the existence of polarons in CMR materials is unambiguously confirmed experimentally, including: very low mobility [14,18], incompatible with Boltzmann-type approaches, activated dc and ac transport in the paramagnetic phase [18,33] and the giant isotope effect in manganites [14,18].

### TABLE I. Polaron shift \( E_p \) due to Fröhlich interaction.

| System     | \( \epsilon_{\infty} \) | \( \epsilon_0 \) | \( a \times b \times c \) (\( \text{Å}^3 \)) | \( E_p \) (eV) |
|------------|-----------------|-----------------|---------------------------------|---------------|
| BaBiO\(_3\) | 5.7\(^a\)       | 30.4\(^b\)      | 4.34\(^2\times\)4.32            | 0.579         |
| BaTiO\(_3\) | 5.1-5.3         | 1499.6          | 3.992\(^2\times\)4.032          | 0.842         |
| La\(_2\)CuO\(_4\) | 5.0         | 30               | 3.82\(^2\times\)6\(^c\)         | 0.647         |
| LaMnO\(_3\) | 3.9\(^d\)       | 16\(^d\)        | 3.86\(^3\)                      | 0.884         |
| La\(_{2.2}\)Sr\(_{1.2}\)O\(_7\) | 4.9\(^d\) | 38\(^d\)           | 3.86\(^2\times\)3.9\(^f\)       | 0.807         |
| SrTiO\(_3\) | 5.2             | 310              | 3.905\(^3\)                     | 0.852         |
| WO\(_3\)   | 5                | 100-300          | 7.31\(^2\times\)7.54\(^7\)      | 0.452         |
| CdO        | 5.4             | 21.9             | 4.7\(^a\)                        | 0.522         |
| EuS        | 5.0             | 11.1             | 5.968\(^3\)                     | 0.324         |
| EuSe       | 5.0             | 9.4              | 6.193\(^3\)                     | 0.266         |
| MgO        | 2.964           | 9.816            | 4.214\(^7\)                     | 0.982         |
| NaCl       | 2.44            | 5.90             | 5.64\(^3\)                      | 0.749         |
| NiO        | 5.4             | 12               | 4.18\(^p\)                      | 0.429         |
| TiO\(_2\)  | 6.7-7.2         | 89-173           | 4.59\(^2\times\)2.96            | 0.643         |

\(^a\)S. Uchida et al. J. Phys. Soc. Jap. \textbf{54}, 4395 (1985).
\(^b\)R.P.S.M. Lobo and F. Gervais, Phys. Rev. B \textbf{52}, 13294 (1995).
\(^c\)Distance between CuO\(_2\) planes.
\(^d\)T. Ishikawa (private communication, 1999).
\(^e\)J.H. Jung and T.W. Noh (private communication, 1999).
\(^f\)R.P.S.M. Lobo and F. Gervais, Phys. Rev. B \textbf{52}, 13294 (1995).

The effective polaron-polaron attraction, due to the overlap of the deformation fields, is about \( 2E_p \) [1], which appears to be more than sufficient to overcome the inter-site Coulomb repulsion, \( V_c \approx 0.7 - 0.8 \text{ eV} \), as also confirmed by the first-principles lattice minimization technique [24]. Hence, we can conclude that the electron-phonon interaction is comparable or even stronger than any relevant magnetic interaction (which is estimated as \( \lesssim 0.2 \text{ eV} \) [1]), so that small (bi)polarons are indeed the most probable quasiparticles in oxides, in contrast to the erroneous claims [1].

### III. OPTICAL CONDUCTIVITY OF INTERSITE BIPOLARONS

Now we show how the observed \textit{massive} weight transfer in optical conductivity with temperature [18] can be naturally explained within the bipolaron theory. The optical intraband conductivity of a charge-transfer doped insulator with (bi)polaronic carriers is the sum of the polaron \( \sigma_p(\nu) \) and bipolaron \( \sigma_b(\nu) \) contributions at a given frequency \( \nu \). Their frequency dependences are described in the literature [23,28,34,35]. In the leading saddle-point approximation both have almost a Gaussian shape given by
tion around a neighboring site “2”, lowering its energy by an amount $2E_p(1-\gamma)$.

\[ \sigma_{\text{intra}}(\nu) = \frac{\sigma_0 T^2}{\nu} \left[ \frac{n}{\gamma_p} \exp \left[ -\left( \nu - \nu_p \right)^2 / \gamma_p^2 \right] ight. \\
\left. + \frac{x - n}{\gamma_b} \exp \left[ -\left( \nu - \nu_b \right)^2 / \gamma_b^2 \right] \right] \]

where $\sigma_0 = 2\pi^{1/2}e^2/a$ is a constant with $a$ the lattice spacing, $T$ the hopping integral, $n$ the (atomic) polaron density, and $x$ the doping level. Here and further we take $\hbar = e = 1$.

We shall first determine the positions and widths of the (bi)polaron absorption peaks for oxides. Those are known for the Holstein model with local interactions, but it is unlikely to apply for oxides because of very large on-site Coulomb repulsion and the long-range (Fröhlich) electron-phonon interaction, which dominates in ionic solids.

Applying the Franck-Condon principle in the adiabatic regime, $\nu \gg \omega$ (phonon frequency), one can generalize the (bi)polaronic absorption, Eq. (3), to describe the optical conductivity of small Fröhlich (bi)polarons, Fig. 1. The electron ‘sitting’ on a site “1”, Fig. 1(a), lowers its energy by an amount $2E_p$, with respect to an atomic level in the undeformed lattice, owing to the lattice deformation. If the electron-phonon interaction has a finite radius, the electron also creates some deformation around a neighboring site “2”, lowering its energy level by an amount $2E_p(1 - \gamma)$, where

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

with $a$ the lattice vector connecting the neighboring sites. The coefficient $\gamma$ strongly depends on the radius of the interaction. In the Holstein model with $q$-independent electron-phonon coupling, $\gamma(q)$, this coefficient equals unity. Hence, there is no lattice deformation at the neighboring site. On the contrary, in the Fröhlich case, $\gamma(q) \propto 1/q$, and the coefficient is quite small, $\gamma \approx 0.2 - 0.4$ depending on the dimensionality of the system and the unit cell geometry. In that case, there is a significant lowering of the neighboring energy level and, as a result, of the polaron mass. Hence, generally, the peak energy in the polaron absorption is found at

\[ \nu_p = 2\gamma E_p, \quad (4) \]

and the activation energy of the high-temperature dc-conductivity is $E_a = \gamma E_p/2$ \[39\]. One can apply the same ‘frozen lattice distortion’ arguments to the intersite bipolaronic absorption, Fig. 1(b). The electron energy on site “2” is $-2E_p - 2E_p(1 - \gamma) + V_c$, where the first contribution is due to the lattice deformation created by the electron itself, while the second contribution is due to the lattice deformation around the site “2” created by the other electron of the pair on the site “1”, which is the polaron-polaron attraction \[1\]. After absorbing a quantum of radiation, the electron hops from site “2” to the empty site “3” into a state with the energy $-2E_p(1 - \gamma)$, which corresponds to an absorption frequency

\[ \nu_b = 2E_p - V_c, \quad (5) \]

where $V_c$ is the inter-site Coulomb repulsion. The quantum broadening of the polaronic and bipolaronic absorption is given by $\gamma_p = \gamma_b = (4\gamma E_p \omega)^{1/2}$. Since doped manganites are intrinsically disordered, their dielectric properties are inhomogeneous, and so is $E_p$, which fluctuates with a characteristic impurity broadening $\Gamma_{im}$. The convolution of the polaronic and bipolaronic absorption lines with the Gaussian distribution of $E_p$ results in their having different linewidths, $\gamma_p = 2(\gamma E_p \omega^2 + \gamma^2 \Gamma_{im}^2)^{1/2}$ and $\gamma_b = 2(\gamma E_p \omega^2 + \Gamma_{im}^2)^{1/2}$ for polaronic and bipolaronic absorption, respectively. The Coulomb repulsion $V_c$ can be readily estimated as $V_c = 2E_p - \nu_b$ from \[3\].

**IV. SPECTRAL WEIGHT TRANSFER IN MANGANITES**

The total absorption is the sum of the intraband polaronic and bipolaronic terms, Eq. (3), and the interband absorption, $\sigma(\nu) = \sigma_{\text{intra}}(\nu) + \sigma_{\text{inter}}(\nu)$. In the layered compounds like $\text{La}_{2-x-2z}\text{Sr}_{1+2z}\text{Mn}_2\text{O}_7$, the interband contribution to the out-of-plane conductivity is negligible \[18\]. Hence, one can take the $c$-axis optical conductivity $\sigma_{c}(\nu)$ as a measure of the interband contribution to the in-plane conductivity with a scaling factor, $s$, $\sigma_{\text{inter}}(\nu) \approx s\sigma_{c}(\nu)$. The scaling factor $s$ is the square of
the ratio of the in-plane components of the dipole matrix element for the interband transitions to its z component (z is the out-of-plane direction). It can be readily determined by comparing the in-plane and out-of-plane optical conductivities at high frequencies, where intraband absorption is irrelevant. The result of the comparison of the present theory with the experiment [18] is shown in Fig. 2. At temperatures above the transition \( T = 130K \) the polaron density is very low owing to CCDC [5], so the intraband conductivity is due to bipolarons only,

\[
\sigma(\nu) = \frac{\alpha_0 x T^2}{\nu_0} \exp \left[ -\left( \frac{\nu - \nu_0}{\gamma_b} \right)^2 \right] + s \sigma_c(\nu). \tag{6}
\]

This expression fits the experiment fairly well with \( \nu_b = 1.24 \) eV and \( \gamma_b = 0.6 \) eV, Fig. 2. The scaling factor is estimated as \( s = 0.6 \). When the temperature drops below \( T_c \), at least some of the bipolarons break apart by the exchange interaction with Mn sites, because one of the spin-polarized polaron bands falls suddenly below the bipolaron level by an amount \( (J_{pd} S - \Delta)/2 \), Fig. 3 [6]. The intraband optical conductivity is determined now by both the polaronic and bipolaronic contributions, Eq. (2), and that explains the sudden spectral weight transfer from \( \nu = \nu_b \) to \( \nu = \nu_p \) observed below \( T_c \) in the ferromagnetic manganites [13,14]. The experimental spectral shape at \( T = 10K \) is well described by Eq. (1) with \( n = x/5, \nu_p \approx 0.5 \) eV and \( \gamma_p \approx 0.3 \) eV (Fig. 2). Taking \( E_p = 1eV \) (Table) we find from Eq.(4) \( \gamma \approx 0.25 \) corresponding to the activation energy \( E_a = 125 \text{meV} \) and phonon frequency \( \omega \approx 75 - 90 \text{meV} \) [1]. With the use of Eq.(5) we find \( V_c \approx 0.76eV \) in good correspondence with previous estimates for cuprates [12].

\[
\sigma(\nu) = \frac{\alpha_0 x T^2}{\nu_0} \exp \left[ -\left( \frac{\nu - \nu_0}{\gamma_b} \right)^2 \right] + s \sigma_c(\nu).
\]

![FIG. 2](image)

**FIG. 2.** Optical conductivity of La\(_{2-x}\)Sr\(_{x}\)Mn\(_2\)O\(_7\) [17] compared with the theory (solid line) above \( T_c \) (top panel) and well below \( T_c \) (bottom panel). Inset: experimental c-axis optical conductivity [17].

\[
\sigma(\nu) = \frac{\alpha_0 x T^2}{\nu_0} \exp \left[ -\left( \frac{\nu - \nu_0}{\gamma_b} \right)^2 \right] + s \sigma_c(\nu).
\]

We note the weak temperature dependence of the optical conductivity at low temperatures \( T < 50K \) and above \( T > T_c \), in agreement with our theory. We do not expect any significant temperature dependence of the optical conductivity in the paramagnetic phase because the polaron density remains small compared with the bipolaron density above \( T_c \). Our results also suggest a double-peak structure of \( \sigma(\nu) \) below \( T_c \), which was clearly observed for La\(_{0.7}\)Ca\(_{0.3}\)Mn\(_2\)O\(_7\) [15]. This structure is not evident in the experiment on layered compound (Fig. 2), thus, more detailed studies of this low-temperature region are most desirable. It is worth mentioning that the scatter of the on-site energies of carriers may lead to an asymmetric broadening of both polaronic and bipolaronic absorption peaks [13], which can wash out the double-peak structure at low temperatures.

The temperature dependence of the polaron density can be found from our Hartree-Fock equations [4,15]. The polaron density at zero temperature is about the total (chemical) density of holes introduced by doping, while in the paramagnetic phase one obtains

\[
n(T > T_c) \sim \exp(-\Delta/2k_BT) \tag{7}
\]

for \( T \geq T_c \) with the prefactor depending on the ratio \( W_p/T_c \). Note that \( n \approx x \) below the Curie point, at \( T < T_c \). Therefore, there is a very abrupt drop of the polaron density at \( T_c \), which exponentially depends on the bipolaron binding energy \( \Delta = nu - nu - W_p \) (Fig. 3). It can reach many orders of magnitude if the relative strength of the exchange interaction \( J_{pd} S \) is close enough to the binding energy.

![FIG. 3](image)

**FIG. 3.** Spin-polarized polaron band (P) in the ferromagnetic phase \( T < T_c \) overlaps with the bipolaron (impurity) band (BP), breaking up a fraction of the bipolarons. In the paramagnetic phase \( T > T_c \) the spin-splitting of the polaron band is absent and system contains mainly localized bipolarons.
V. CONCLUSIONS

In conclusion, we have calculated the value of the electron-phonon interaction in oxides (Table) to show that the Fröhlich interaction plays the dominant role in comparison with any magnetic (exchange) interaction. The conditions in oxides are such that small polarons and bipolarons are the most probable quasiparticles both in insulating and superconducting compounds. An opposite conclusion, recently suggested in the literature [11,23], stems from (i) an incorrect expression for the polaron binding energy, (ii) misunderstanding of the criteria for small polaron formation, and from (iii) misunderstanding of the screening in polaronic conductors. We have developed the theory of the optical conductivity in doped magnetic charge-transfer insulators with a strong electron-phonon interaction. The spectral and temperature features of the optical conductivity of ferromagnetic manganites are well described by the bipolaron absorption in the paramagnetic phase and by the small polaron absorption in the ferromagnetic phase. The pair breaking by exchange interaction with the localized Mn spins explains the sudden spectral weight transfer in the optical conductivity below $T_c$. Therefore, the optical probe of the incoherent charge dynamics in manganites provides another strong evidence for the carrier density collapse, which we proposed earlier as the explanation of CMR. The anomalous specific heat and tunneling measurements also support CCDC [9]. The Mn localised spins give a major contribution into magnetic susceptibility peak around $T_c$, and a deviation of its temperature dependence from standard Curie behavior may better elucidate a (smaller) magnetic response of polarons and a character of the phase transition.

Our picture is further supported by recent ARPES data [4], clearly showing a pseudogap in the band dispersion in manganites at the Fermi level at low temperatures, incompatible with the presence of a metallic phase. In addition, strong antiferromagnetic correlations in the paramagnetic phase at $T > T_c$, found in polarized neutron scattering and in Raman spectra [17] support our idea of the singlet pairing above $T_c$. Outside the ferromagnetic regions (i) the bipolarons would tend to form charge density waves, usually observed as a charge ordering, and (ii) doped carriers would spill over to Mn sites from oxygen [22], thus allowing, at least in principle, a possibility for some orbital effects. The theory suggests that by replacing the magnetic ions (Mn) with nonmagnetic ions (Cu), one can transform a doped charge-transfer insulator into a high-temperature superconductor owing to the Bose-Einstein condensation of bipolarons [1], as was most likely observed experimentally [18].

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