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LETTER TO THE EDITOR

The essential interactions in oxides and spectral weight transfer in doped manganitesA S Alexandrov[†] and A M Bratkovsky[‡][†] Department of Physics, Loughborough University, Loughborough, Leics LE11 3TU, UK[‡] Hewlett-Packard Laboratories, 3500 Deer Creek Road, Palo Alto, CA 94304-1392, USA

E-mail: asa21@cus.cam.ac.uk (A S Alexandrov) and alexb@hp1.hp.com (A M Bratkovsky)

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Abstract. 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.

(Some figures in this article appear in colour in the electronic version; see www.iop.org)**1. Introduction**

It is well understood that carriers in cuprates [1, 2] and manganites [3–5] 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 d electrons. As a result, the exchange interaction dissociates bipolarons below T_c if the p–d exchange energy $J_{pd}S$ of the polaronic carriers with the localized Mn d electrons is larger than the bipolaron binding energy Δ . Hence, the density of current carriers (polarons) suddenly increases below T_c , which explains the resistivity peak and CMR, observed in many ferromagnetic oxides [6–8]. We have shown [9] that CCDC also explains the giant isotope effect [10, 11], the tunnelling gap [12], the specific heat anomaly [13], and the temperature dependence of the dc resistivity [14].

The non-metallic nature of the ferromagnetic low-temperature phase of the doped manganites has been unambiguously confirmed in recent studies of the optical conductivity [15–18] and photoemission [19]. In particular, a broad incoherent spectral feature [15–18] in the mid-infrared region and a pseudogap in the excitation spectrum [19] were observed, while the coherent Drude weight appeared to be one or even two orders of magnitude smaller [16, 17] than expected for a metal, or almost *absent* [18]. These and other studies [20] prove that carriers retain their polaronic character well below T_c , in agreement with our theory of CMR [5]. For example, the measured residual conductivity (at $T = 0$) $\sigma_0 = 360 \Omega^{-1} \text{cm}^{-1}$ [18] yields the

product of the Fermi wave vector and the mean free path $k_F l \lesssim 1$, which is below the Ioffe–Regel limit[†]. 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 T_c , observed in several systems.

In this letter 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-bipolaron absorption, while the low-temperature mid-infrared band is due to absorption by small polarons. The bipolaron dissociation below T_c shifts the spectral weight from the bipolaronic peak to the polaronic one. We describe the optical spectra of the layered ferromagnetic ($T_c = 125$ K) crystal $\text{La}_{2-2x}\text{Sr}_{1+2x}\text{Mn}_2\text{O}_7$ [18] for the entire frequency and temperature range that has been studied experimentally, and show that the optical data provide strong evidence for CCDC.

2. Fröhlich interaction and small-(bi)polaron formation in oxides

The carriers in both cuprates and manganites are O p (*not* Cu or Mn d) holes [21], and their pairing in the paramagnetic phase due to strong electron–phonon interaction would most likely be in the form of *intersite* bipolarons localized on a pair of O sites [1]. Clearly, the formation of these bipolarons cannot be hindered by repulsive on-site Hubbard- U and/or Hund’s rule coupling, contrary to recent claims in the literature [22].

In order to assess the possibility of small-polaron and small-bipolaron formation in manganites and cuprates, one has to calculate the electron–phonon interaction and compare it with the (intersite) Coulomb repulsion. Fortunately, making such an estimate is possible for 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-wavelength limit the response of polarons at the optical phonon frequency is dynamic [23]. 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 the 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 [24]. Claims in the literature [25, 26] that the Fröhlich interaction is reduced to a local Holstein interaction in doped oxides disregard the established fact that the mobility of carriers determines the screening, rather than their density [1].

We shall apply the exact expression for the polaronic shift E_p [1, 27–29], which for the Fröhlich interaction reads

$$E_p = \frac{1}{2\kappa} \int_{BZ} \frac{d^3q}{(2\pi)^3} \frac{4\pi e^2}{q^2}. \quad (1)$$

Here the dielectric constants, $\kappa^{-1} = \epsilon_\infty^{-1} - \epsilon_0^{-1}$, are known from experiment, and the size of the integration region (the Brillouin zone, BZ) is determined by the lattice constants a, b, c (table 1). The data in the table represent the *lower* boundary for the polaron binding energies, equation (1), since taking account of coupling with acoustic and/or Jahn–Teller modes can only increase the polaronic shift E_p . The correct value of E_p appears to be about 1 eV in most cases, far exceeding the *ad hoc* estimates of ~ 0.15 eV [30], obtained from a numerically incorrect

[†] In two dimensions, $\sigma_0 = (e^2/h)(k_F l)$.

Table 1. The polaron shift E_p due to Fröhlich interaction; the data are from [48] and [49]. The value $\epsilon_\infty = 5$ for WO_3 is an estimate.

System	ϵ_∞	ϵ_0	$a \times b \times c$ (\AA^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	1499.0	$3.992^2 \times 4.032$	0.842
La ₂ CuO ₄	5.0	30	$3.8^2 \times 6^c$	0.647
LaMnO ₃	3.9 ^d	16 ^{d,e}	3.86^3	0.884
La _{2-2x} Sr _{1+2x} Mn 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	$7.31 \times 7.54 \times 7.7$	0.445
CdO	5.4	21.9	4.7^3	0.522
EuS	5.0	11.1	5.968^3	0.324
EuSe	5.0	9.4	6.1936^3	0.266
MgO	2.964	9.816	4.2147^3	0.982
NaCl	2.44	5.90	5.643^3	0.749
NiO	5.4	12	4.18^3	0.429
TiO ₂	6–7.2	89–173	$4.59^2 \times 2.96$	0.643

^a Reference [50].^b Reference [51].^c The distance between CuO₂ planes.^d Reference [52].^e Jung and Noh [53] have estimated from their data $\epsilon_\infty = 3.4$ and $\epsilon_0 = 21$ for LaMnO₃.^f The distance between MnO₂ planes.

expression for E_p and wrong values of the dielectric constants. The large calculated value of the polaron shift (~ 1 eV) is perfectly compatible with the small-polaron theory. According to [30] the bare half-bandwidth is about $W/2 \simeq 0.8$ eV in manganites. Hence, even a simple variational criterion [30] 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 [31]. 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, 32], and the giant isotope effect in manganites [10, 11].

The effective polaron–polaron attraction, due to the overlap of the deformation fields, is about $2E_p$ [1]. This appears to be more than sufficient to overcome the intersite Coulomb repulsion, $V_c \simeq 0.7$ – 0.8 eV, as is also confirmed by the first-principles lattice-minimization technique [33]. Hence, we can conclude that the electron–phonon interaction is comparable to or even stronger than any relevant magnetic interaction (which is estimated as $\lesssim 0.2$ eV [30]), so small (bi)polarons are indeed the most probable quasiparticles in oxides, in contrast to the erroneous claims [30].

3. Optical conductivity of intersite bipolarons

Now we show how the observed *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 ν . Their frequency dependences are described in the literature [1, 34–39]. In the leading saddle-point

approximation both have almost a Gaussian shape given by

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

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

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 [35] in the adiabatic regime, $\nu \gg \omega$ (phonon frequency), one can generalize the (bi)polaronic absorption, equation (2), to describe the optical conductivity of small Fröhlich (bi) polarons; see figure 1. The electron ‘sitting’ at a site ‘1’, figure 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 neighbouring site ‘2’, lowering its energy level by an amount $2E_p(1 - \gamma)$, where [41]

$$\gamma = \sum_q |\gamma(\mathbf{q})|^2 [1 - \cos(\mathbf{q} \cdot \mathbf{a})] / \sum_q |\gamma(\mathbf{q})|^2 \quad (3)$$

with \mathbf{a} the lattice vector connecting the neighbouring sites. The coefficient γ strongly depends on the radius of the interaction. In the Holstein model with \mathbf{q} -independent electron–phonon coupling, $\gamma(\mathbf{q})$, this coefficient equals unity. Hence, there is no lattice deformation at the neighbouring site. On the contrary, in the Fröhlich case, $\gamma(\mathbf{q}) \propto 1/q$, and the coefficient is quite small, $\gamma \approx 0.2\text{--}0.4$ [41] depending on the dimensionality of the system and the unit cell geometry. In that case, there is a significant lowering of the neighbouring energy level and, as a result, of the polaron mass [31]. Hence, generally, the peak energy in the polaron absorption is found at

$$\nu_p = 2\gamma E_p \quad (4)$$

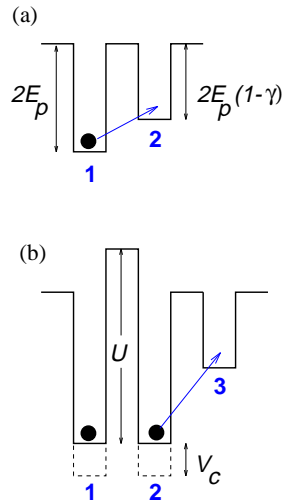


Figure 1. Adiabatic energy levels and optical transitions for the small polaron (a) and the intersite small bipolaron (b).

and the activation energy of the high-temperature dc conductivity is $E_a = \gamma E_p/2$ [35]. One can apply the same ‘frozen-lattice distortion’ arguments to the intersite bipolaron absorption; see figure 1(b). The electron energy at 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 at 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 intersite 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 Γ_{im} . The convolution of the polaronic and bipolaronic absorption lines with the Gaussian distribution of E_p results in their having different linewidths, $\gamma_p = 2(\gamma E_p \omega + \gamma^2 \Gamma_{\text{im}}^2)^{1/2}$ and $\gamma_b = 2(\gamma E_p \omega + \Gamma_{\text{im}}^2)^{1/2}$ for polaronic and bipolaronic absorption, respectively. The Coulomb repulsion V_c can be readily estimated as $V_c = 2E_p - \nu_b$ from (5).

4. Spectral weight transfer in manganites

The total absorption is the sum of the intraband polaronic and bipolaronic terms, equation (2), and the interband absorption, $\sigma(\nu) = \sigma_{\text{intra}}(\nu) + \sigma_{\text{inter}}(\nu)$. In the layered compounds like $\text{La}_{2-2x}\text{Sr}_{1+2x}\text{Mn}_2\text{O}_7$, the intraband 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 , $\sigma_{\text{inter}}(\nu) \simeq s\sigma_c(\nu)$. 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 figure 2.

At temperatures above the transition ($T = 130$ K) the polaron density is very low owing to CCDC [5], so the intraband conductivity is due to bipolarons only:

$$\sigma(\nu) = \frac{\sigma_0 x T^2}{\nu \gamma_b} \exp[-(\nu - \nu_b)^2 / \gamma_b^2] + s\sigma_c(\nu). \quad (6)$$

This expression fits the experiment fairly well with $\nu_b = 1.24$ eV and $\gamma_b = 0.6$ eV; see figure 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 $(J_{pd}S - \Delta)/2$; see figure 3 [5]. The intraband optical conductivity is determined now by both the polaronic and bipolaronic contributions, equation (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 [16–18]. The experimental spectral shape at $T = 10$ K is well described by equation (1) with $n = x/5$, $\nu_p \simeq 0.5$ eV, and $\gamma_p \simeq 0.3$ eV (figure 2). Taking $E_p = 1$ eV (table 1) we find from equation (4) $\gamma \simeq 0.25$ corresponding to the activation energy $E_a = 125$ meV and phonon frequency $\omega \simeq 75$ – 90 meV [40]. With the use of equation (5) we find $V_c \simeq 0.76$ eV in good correspondence with previous estimates for cuprates [41].

We note the weak temperature dependence of the optical conductivity at low temperatures $T < 50$ K and above T_c [18] in agreement with our theory. We do not expect any significant

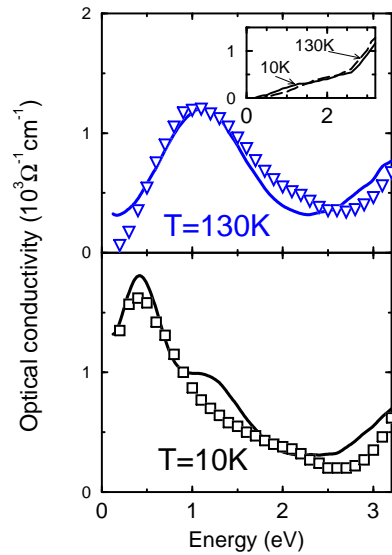


Figure 2. Optical conductivity of $\text{La}_{2-2x}\text{Sr}_{1+2x}\text{Mn}_2\text{O}_7$ [17] compared with the theory (solid line) above T_c (top panel) and well below T_c (bottom panel). Inset: the experimental c -axis optical conductivity [17].

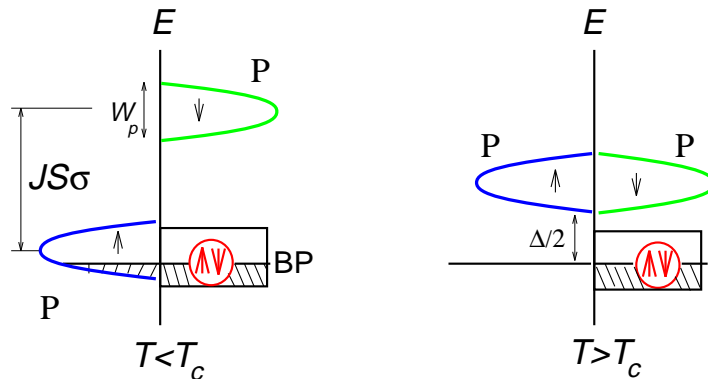


Figure 3. The spin-polarized polaron band (P) in the ferromagnetic phase ($T < T_c$) overlaps with the bipolaron (impurity) band (BP), breaking up a fraction of the bipolarons. In the paramagnetic phase ($T > T_c$) the spin splitting of the polaron band is absent and system contains mainly localized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

temperature dependence of the optical conductivity in the paramagnetic phase because the polaron density remains small compared with the bipolaron density above T_c [5]. Our results also suggest a double-peak structure of $\sigma(\nu)$ below T_c , which was clearly observed for $\text{La}_{0.7}\text{Ca}_{0.3}\text{MnO}_3$ [42]. This structure is not evident in the experiment on the layered compound (figure 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 [43], 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 [5,44]. The polaron density at zero temperature is about the total (chemical) density of holes introduced by doping, while in the paramagnetic phase one obtains

$$n(T > T_c) \sim \exp(-\Delta/2k_B T) \quad (7)$$

for $T \geq T_c$ with the prefactor depending on the ratio W_p/T_c . Note that $n \sim x$ 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 $\Delta = v_b - v_p - W_p$ (figure 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.

5. Conclusions

In conclusion, we have calculated the value of the electron–phonon interaction in oxides (table 1) 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 [25, 30], stems from (i) an incorrect expression for the polaron binding energy, (ii) misunderstanding of the criteria for small-polaron formation, and (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 further strong evidence for the carrier-density collapse, which we proposed earlier as the explanation of the CMR. The anomalous specific heat and tunnelling measurements also support CCDC [9]. The Mn localized spins give a major contribution to the magnetic susceptibility peak around T_c , and a deviation of its temperature dependence from standard Curie behaviour may better elucidate a (smaller) magnetic response of polarons and the character of the phase transition.

Our picture is further supported by recent ARPES data [45], 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 [46], 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 carries would spill over to Mn sites from oxygen [21], thus allowing, at least in principle, a possibility for some orbital effects. The theory suggests that by replacing the magnetic ions (Mn) with non-magnetic 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 [47].

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