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Composite polarons in ferromagnetic narrow-band metallic manganese oxides

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Abstract. Moving electrons accompanied by Jahn–Teller phonon and spin-wave clouds may form composite polarons in ferromagnetic narrow-band manganites. The ground-state and finite-temperature properties of such composite polarons are studied in the present paper. By using a variational method, it is shown that the energy of the system at zero temperature decreases with the formation of the composite polaron and the composite polaron behaves as a Jahn–Teller phononic polaron; the energy spectrum of the composite polaron at finite temperatures is found to be strongly renormalized by the temperature and the magnetic field. It is suggested that the composite polaron contributes significantly to the transport and the thermodynamic properties in ferromagnetic narrow-band metallic manganese oxides.

1. Introduction

Because of the electron–lattice interaction, an electron moving in a polarized or a dynamically distorted lattice surrounded by phonon clouds at low temperature is damped and forms a phononic polaron [1]. Similarly, a moving electron in a ferromagnetic (or antiferromagnetic) background disturbs the nearby local spin field and excites spin waves; therefore an electron surrounded by spin-wave clouds can form a magnetic polaron [2]. One could anticipate that in the presence of strong electron–lattice interaction and electron–spin coupling, a moving electron clouded both by phonons and spin waves can form a kind of new quasiparticle, which we call a composite polaron. In the presence of a magnetic field and for a certain temperature, the physical properties of a system with composite polarons differ from those for systems with either magnetic polarons or phononic polarons. Since there exist strong electron–phonon interactions, arising from the Jahn–Teller effect of MnO_6, and strong electron–local spin interactions, arising from Hund's coupling, this kind of new quasiparticle might be found in ferromagnetic manganese oxide materials, such as the doped lanthanum manganites and neodymium manganites.

Recently, the colossal magnetoresistance (CMR) effect has been found in manganites, such as $La_{1-x}Ca_xMnO_3$ and $Nd_{1-x}Sr_xMnO_3$ [3–6]. The huge magnetoresistance change of more than 3–6 orders of magnitude has potential technical applications. Other unusual transport, magnetic, and thermodynamic properties, and the microscopic mechanism of the

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CMR effect have attracted great interest theoretically and experimentally. The crystal and magnetic structures of $La_{1-x}R_xMnO_3$ systems had been studied in 1950s by the x-ray crystallography and neutron diffraction techniques [7, 8]. In manganites, an Mn ion is surrounded by six O^{2-} ions and forms an octahedron. Due to the crystalline-field effect, the 3d energy level of the Mn ion is split into a low-lying triplet (t_{2g}) and a high-energy doublet (e_g). Therefore, in $La_{1-x}R_xMnO_3$, the three d electrons of Mn^{3+} and Mn^{4+} ions fill the d t_{2g} band (the filled band) and form a localized core spin $S = \frac{3}{2}$ via strong intra-atomic Hund's rule coupling. The extra d electrons in Mn^{3+} fill the higher d e_g band and interact with t_{2g} electrons through strong Hund coupling. These two bands are separated by about 1.5 eV [9]. The localized spins tend to align parallel through the double-exchange interaction between Mn^{3+} and Mn^{4+} ions, and form a ferromagnetic background [10, 11]. Electrons in the d e_g band hop between Mn ions as itinerant ones, and are responsible for the electrical conduction in these systems. The degenerate d e_g doublet of Mn^{3+} in the MnO_6 octahedron will be further split due to the Jahn–Teller effect [12, 13]; the distortion of oxygen atoms reduces the symmetry of the MnO_6 octahedron and stabilizes it. An electron moving together with the distorted crystalline field (dynamical Jahn–Teller distortion) will form a polaron. Recent experiments and theories [13–18] have confirmed the existence and the importance of the Jahn–Teller distortion in $La_{1-x}R_xMnO_3$ compounds. Millis *et al* [13] suggested that the dynamic Jahn–Teller effect plays a crucial role in the physical properties of manganites. In the meantime, doped $La_{1-x}R_xMnO_3$ compounds ($0.1 < x < 0.5$) are ferromagnetically ordered at temperatures below the Curie temperature T_c. The exchange coupling between the mobile electron and ordered localized spins can form a magnetic polaron. Therefore the motion of an electron at the e_g level in lanthanum manganites may form a composite quasiparticle, with the simultaneous coexistence of phononic and magnetic polarons at the same place; this called the composite polaron.

The composite polaron (CP) in the present situation is small in the metallic regime. An earlier study [19] had shown that lattice deformation can stabilize a small magnetic polaron, so the CP formed in ferromagnetic narrow-band metallic manganese oxides is stable.

In this paper, in section 2 and section 3, the framework of the composite polaron is developed for perovskite-type manganese oxides, and its dependence on the temperature and magnetic field are discussed. We found that the formation of a CP in manganese oxide systems is favourable in energy, and propose that the CP plays an essential role in the transport and the thermodynamic properties of lanthanum manganites. The formation of the CP could be responsible for the microscopic mechanism of the CMR effect. Finally, we draw conclusions in section 4.

2. The composite polaron at zero temperature

We first discuss the possibility of the formation of a CP in ferromagnetic narrow-band manganese oxides at zero temperature. The typical physical processes in manganites can be decomposed into three parts: the electron–electron interaction (the hopping process and the Coulomb interaction), H_0, the electron–lattice interaction (the Jahn–Teller effect), H_{e-ph}, and the interaction between localized spins and mobile electrons (Hund's coupling), H_{e-m}. So the Hamiltonian is

$$H = H_0 + H_{e-m} + H_{e-ph} \tag{1}$$

where

$$H_0 = \sum_{ia\sigma}[\epsilon_d^a - \sigma\mu_B B]d_{ia\sigma}^\dagger d_{ia\sigma} + \sum_{\langle ij\rangle ab\sigma} t^{ab} d_{ia\sigma}^\dagger d_{jb\sigma} \tag{2}$$

$$H_{e-ph} = \sqrt{\alpha\hbar\omega}\sum_{ia} n_{ia}\beta_a[b_i^\dagger + b_i] + \sum_i \hbar\omega b_i^\dagger b_i \tag{3}$$

$$H_{e-m} = -J_H \sum_{ia\mu\nu} \boldsymbol{S}_i \cdot d_{ia\mu}^\dagger \sigma_{\mu\nu} d_{ia\nu} - \sum_{\langle ij\rangle} A_{ij}\boldsymbol{S}_i \cdot \boldsymbol{S}_j - \sum_i g\mu_B B S_i^z. \tag{4}$$

In equation (2), $d_{ia\sigma}^\dagger$ creates an e_g electron at site $\boldsymbol{R}_i$ at level a with spin σ, t^{ab} denotes the hopping integral of the e_g electron from one site to its nearest neighbour, ϵ_d is the site energy of the mobile electron with respect to the chemical potential, and a (or b) denotes the energy-level index of the Jahn–Teller splitting in the e_g level. In equation (3), $b_i^\dagger$ creates a Jahn–Teller phonon at site $\boldsymbol{R}_i$ with mode $\hbar\omega$, α represents the electron–phonon coupling constant, and β_a is a two-component constant vector; it is -1 or 1 for the low and the high levels of the Jahn–Teller splittings, respectively. Several experiments have shown that in the metallic regime, the magnetic ordering of the local spins is of Heisenberg ferromagnetic type [20, 21]. In equation (4), A_{ij} is the effective ferromagnetic exchange constant of manganese spins with only the nearest-neighbour interaction taken into consideration, and $-g\mu_B B$ represents the Zeeman energy in the magnetic field $\boldsymbol{B}$. The mobile electron is scattered from state $i\nu$ to state $i\mu$ by the localized spin $\boldsymbol{S}_i$ due to the Hund coupling J_H between the mobile electrons and the core spins. As suggested by Kubo and Ohata [22], Millis *et al* [13] and Zang *et al* [14], $J_H \gg t^{ab}$, so the bandwidth of the metallic manganites is narrow.

In doped perovskite manganites, the double-exchange interaction due to the hole hopping and the strong Jahn–Teller electron–phonon coupling is limited to a range of a few lattice constants. This fact suggests that the small-polaron picture is suitable for the CMR materials. Through a canonical transformation, $H' = \mathrm{e}^{-S} H \mathrm{e}^{S}$, one could eliminate the term linear in phonon degrees of freedom in equation (1). By choosing

$$S = \sum_i \sqrt{\alpha/\hbar\omega}\, n_{i\alpha}\beta_\alpha (b_i^\dagger - b_i)$$

an effectively attractive electron–electron interaction is introduced, and the Hamiltonian equation (1) can be rewritten as

$$H' = \sum_{ia\sigma} \epsilon_{i\sigma}^a d_{ia\sigma}^\dagger d_{ia\sigma} + \sum_{\langle ij\rangle ab\sigma} t^{ab} d_{ia\sigma}^\dagger d_{jb\sigma} \hat{X}_{ia}^\dagger \hat{X}_{jb} - \sum_{ia,b} \alpha n_{ia}\beta_a n_{ib}\beta_b + \sum_i \hbar\omega_i b_i^\dagger b_i + H_{e-m} \tag{5}$$

where $\epsilon_{i\sigma}^a = \epsilon_d^a - \mu - \sigma\mu_B B$, and $\hat{X}_{ia}^\dagger = \exp[\sqrt{\alpha/\hbar\omega}\beta_a(b_i - b_i^\dagger)]$. In the present studies we are only interested in the ground state of the CP, so it is reasonable to assume that the mobile electrons stay in the lower level of the Jahn–Teller splittings, and we will drop the indices a and b in equation (5). The term $-\sum_{ia,b}\alpha n_{ia}\beta_a n_{ib}\beta_b$ is reduced to a renormalization shift of the bare energy level by $-\alpha$, and the energy shift of the electrons from the electron–phonon interaction, α, is absorbed into ϵ_d^a.

We choose a set of basis functions for constructing the variational ground-state wavefunction of the CP at $T = 0$ K:

$$|\psi_i(S_0^z)\rangle = \sqrt{\frac{S + S_{0i}^z + 1}{2S+1}}\left[d_{i\uparrow}^\dagger + \frac{d_{i\downarrow}^\dagger S_i^\dagger}{S + S_{0i}^z + 1}\right]\prod_{j=1}^{N} |S_j S_j^z\rangle |0\rangle_{e-ph} \tag{6}$$

where $\prod_{j=1}^N |S_j S_j^z\rangle$ denotes the ferromagnetic background and $|0\rangle_{e-ph}$ represents the electron and phonon vacuum state. S_{0i}^z denotes the mean value of the z-component of the local spin

at $\boldsymbol{R}_i$. This basis set is constructed from the wavefunction of the vacuum state of the phonon, $|0\rangle_{ph}$, and that of the magnetic polaron (reference [2]):

$$|\chi\rangle = \sqrt{\frac{S+S_{0i}^z}{2S+1}}[d_{i\downarrow}^\dagger - d_{i\uparrow}^\dagger S_i^-/(S+S_{0i}^z)]\prod_{j=1}^{N}|J_j J_j^z\rangle|0\rangle_e.$$

If we let $\alpha \to 0$ or $J_H \to 0$ in (5), the problem reduces to that of the magnetic polaron or the phononic polaron, respectively.

The ground-state wavefunction of the CP can be constructed in terms of the basis set, $|\psi_i(S_0^z)\rangle$, through a linear combination:

$$|G\rangle = \sum_{i S_0^z} c_i(S_0^z)|\psi_i(S_0^z)\rangle. \tag{7}$$

Here $c_i(S_0^z)$ is the variational coefficient of the ground-state wavefunction, and S_0^z represents all spin variables. Acting with the Hamiltonian H' on $|G\rangle$ gives the ground-state energy of the CP: $E_g\langle G|G\rangle = \langle G|H'|G\rangle$. Minimizing the above expression with respect to the coefficients $c_i(S_0^z)$, one gets

$$\begin{aligned} E_g c_i = {} & -J_H S c_i + t\mathrm{e}^{-p/2}\sum_\delta \frac{\sqrt{(S+S_{0i}^z+1)(S+S_{0i+\delta}^z+1)}}{2S+1}c_{i+\delta} \\ & + \frac{S+S_{0i+\delta}^z+1}{2S+1}\left[\epsilon_\uparrow + \epsilon_\downarrow \frac{S-S_{0i}^z}{S+S_{0i+\delta}^z+1}\right]c_i \\ & + \frac{S-S_{0i}^z}{2S+1}\left[-g\mu_B B - 2A\sum_\delta S_{0i+\delta}^z\right]. \end{aligned} \tag{8}$$

After Fourier transformation, the nontrivial solution of the coefficients $c_i(S_i^z)$ gives rise to the ground-state energy spectrum of the CP:

$$\begin{aligned} E_g(k) = {} & \epsilon_d - J_H S + zt\mathrm{e}^{-p/2}\gamma(k)\frac{\sqrt{(S+S^z+1)(S+S_\delta^z+1)}}{2S+1} \\ & - \frac{2S}{2S+1}\left[\mu_B B + \frac{g}{2S}\mu_B B + A\sum_\delta S_\delta^z\right] \end{aligned} \tag{9}$$

where $p = \alpha/\hbar\omega$ is the relative strength of the electron–phonon interaction, z is the partition number, and $\gamma(k)$ the structure factor. The physical meaning of each term in the above expression is obvious: the first two terms, $\epsilon_d - J_H S$, come from the intra-atomic interaction, and the third term describes the renormalized energy dispersion. The electron–phonon interaction and the spin–electron coupling narrow the bare energy spectrum of the electrons, $zt\gamma(k)$, by a factor of $\exp(-p/2)$ and a factor of $\sqrt{(S+S_0^z+1)(S+S_{0\delta}^z+1)}/(2S+1)$, respectively. However one notices that at the absolute zero of temperature, the system is ferromagnetically ordered and there is no spin excitation, $S^z = S$, so

$$\frac{\sqrt{(S+S_{0i}^z+1)(S+S_{0i+\delta}^z+1)}}{2S+1} = 1.$$

Therefore the CP behaves as a phononic polaron at zero temperature. This point rests in the fact that at zero temperature, there is no spin deviation in ferromagnets, so the electron is only accompanied by a dynamically distorted lattice field. However, the zero-temperature CP differs from the Jahn–Teller polaron in the intra-atomic Hund coupling (the second term in equation (9)) and the effective Zeeman energy of the CP (the last term in equation (9)).

It can be seen that the formation of the CP lowers the energy of the system significantly, so it is more favourable as regards energy.

The presence of strong Jahn–Teller electron–phonon coupling in ferromagnetic perovskite manganites has been confirmed by several experiments [15–18]. By using neutron powder diffraction data [15], it has been shown that the static distortion of oxygen around manganese resulting from Jahn–Teller electron–phonon coupling is about 0.12 Å in $La_{1-x}Ca_xMnO_3$ ($x \approx 0.2$). Direct support for the presence of electron–phonon coupling came from the oxygen isotope experiments performed by Zhao *et al* [23]. It is believed that the ferromagnetic coupling between manganese spins arises from the double-exchange interaction, mediated through oxygen atoms, so the motion of oxygen atoms will affect the double-exchange strength, and hence the ferromagnetic coupling, by a factor of $e^{-p(1/2+\langle n_{ph}\rangle)}$ (here $\langle n_{ph}\rangle$ is the mean phonon number at finite temperatures; see the next section). Since $p \sim 1/\omega \sim M^{1/2}$, where M is the mass of oxygen, the heavier the oxygen nucleus, the weaker the coupling. Therefore, the presence of a strong electron–phonon interaction will lead to a significant decrease of the Curie temperature. Zhao *et al* showed that the oxygen isotope exponent, $\alpha_0 = -\mathrm{d}\ln T_c/\mathrm{d}\ln M_0$, can be as high as 0.85 for $La_{0.8}Ca_{0.2}MnO_3$, suggesting the existence of strong electron–phonon interaction. Accordingly, one could estimate that the relative strength of the electron–phonon interaction, p, is about 1–2, which places the compound in the strong-coupling regime.

On the other hand, in addition to the strong electron–phonon interaction, the spin–electron interaction is also fairly strong. In the model Hamiltonian given as equation (4), the Hund coupling between the carrier and the local spin is about 5 eV, much larger than the conduction bandwidth $2zt$ (≈2.0 eV [13]). Experiments performed by Kusters *et al* [3], Von Helmolt *et al* [4], Jin *et al* [5] and other groups led to the proposal that spin polarons may dominate the electric transport. Such a viewpoint is supported by the fact that the logarithm of the conductivity of $La_{1-x}Ca_xMnO_3$ exhibits a $T^{-1/4}$-dependence on temperature [24], which suggests a typical spin-polaron character for the transport properties of $La_{1-x}Ca_xMnO_3$.

3. Properties of composite polarons at finite temperature

With the increase of the temperature, more and more phonons and spin waves are excited, whereas the increase of the external magnetic field decreases the degree of spin-wave excitation. Thus the formation and the properties of the CP can be heavily affected by the external magnetic field $\boldsymbol{B}$ and temperature T.

At finite temperatures, the thermal excitation of the phonons and spin waves becomes more and more important; their occupations obey the Bose–Einstein distribution law. We choose the following basis function for the CP at temperature T:

$$|\phi_i(S_0^z)\rangle = \sqrt{\frac{S+S_{0i}^z}{2S+1}}\left[d_{i\uparrow}^{\dagger} + \frac{d_{i\downarrow}^{\dagger}S_i^{\dagger}}{S+S_{0i}^z}\right]|0\rangle_e|n_i\rangle_{ph}|m_i\rangle_m \tag{10}$$

where $|0\rangle_e$ denotes the electron vacuum state, $|n_i\rangle_{ph}$ the thermal equilibrium phonon state, and $|m_i\rangle_m$ the magnetic excitation state at site $\boldsymbol{R}_i$ at temperature T under the magnetic field $\boldsymbol{B}$. Denoting as a_i^+ (or a_i) the creation (or the annihilation) operator of the CP, the wavefunction of the CP can be expressed as

$$|\Phi_i\rangle = \sum_i a_i|\phi_i(S_0^z)\rangle. \tag{11}$$

Expressing the Hamiltonian (5) in terms of the CP operators, we obtain

$$\tilde{H} = \sum_{i,j} a_j^{\dagger} a_i \langle \phi_j(S_0^z)|H'|\phi_i(S_0^z)\rangle. \tag{12}$$

After a tedious calculation with the use of Fourier transformation, one obtains

$$\tilde{H} = \sum_k \left[\epsilon_d - J_H S - \frac{2S}{2S+1}\left(\mu_B B + \frac{g}{2S}\mu_B B + A\sum_{\delta} S_{\delta}^z\right)\right] a_k^{\dagger} a_k + \sum_k zt\mathrm{e}^{-p(\langle n_B\rangle + 1/2)}\left[1 + \frac{1}{2S+1}\sum_q \left(\frac{2S}{2S+1}\gamma_q - 1\right)\langle m_q\rangle\right]\gamma(k) a_k^{\dagger} a_k \tag{13}$$

where $\langle n_B\rangle = 1/[\exp(\hbar\omega/k_B T) - 1]$ and $\langle m_q\rangle = 1/[\exp(\hbar\Omega_q/k_B T) - 1]$ denote the thermal equilibrium mean occupations of the phonons and of the spin waves at temperature T, respectively; here $\hbar\Omega_q$ is the energy spectrum of the spin waves in double-exchange ferromagnets, and $\hbar\Omega_q = g\mu_B B + 2zA(1-\gamma_q)$. In the calculation, the linear approximation of the Holstein–Primakoff transformation, namely $S^{\dagger} = \sqrt{2S}c$, $S^{-} = \sqrt{2S}c^{\dagger}$, $S^z = S - c^{\dagger}c$, was adopted. The average energies of the free phonons and of the free spin waves were not included.

Compare the dispersion of the CP in equation (13) with the bare energy spectrum $zt\gamma(k)$ of the electrons; the bandwidth of the carriers at finite temperature T is renormalized by the factor Z:

$$Z = \mathrm{e}^{-p(\langle n_B\rangle + 1/2)}\left[1 - \frac{1}{2S+1}\sum_q \left(1 - \frac{2S}{2S+1}\gamma(q)\right)\langle m_q\rangle\right]. \tag{14}$$

One finds that the renormalized factor Z consists of two parts: the phonon-renormalized part Z_{ph} and the spin-wave part Z_m; $Z = Z_{ph}Z_m$. Z exhibits strong temperature and magnetic field dependence. The phononic part, $Z_{ph} = \exp[-p(\langle n_B\rangle + \frac{1}{2})]$ depends only on the temperature. The increase of the electron–phonon interaction and temperature will narrow the conduction band further. The magnetic part

$$Z_m = 1 - \frac{1}{2S+1}\sum_q \left[1 - \frac{2S}{2S+1}\gamma(q)\right]\langle m_q\rangle$$

depends both on the temperature and on the magnetic field. With the increase of temperature, the occupation of the spin waves, $\langle m_q\rangle$, becomes large. Since

$$1 - \frac{2S}{2S+1}\gamma(q)$$

is always positive, the factor Z_m becomes small. Therefore, in the region where the temperature is below T_c, the bandwidth of the CP becomes narrower and narrower with the increase of the temperature, and the CP may be more easily trapped, so the mobility of the CP becomes smaller. Under a strong magnetic field, the increase of the field strength depresses the excitation of the spin waves, and reduces the number of spin waves surrounding the electron; thus the bandwidth of the CP becomes broader with increasing external magnetic field and the degree of mobility of the CP becomes greater. These properties coincide with experimental observations for the resistivity. When T approaches the Curie temperature from below, the number of spin-wave excitations reaches its maximum and the factor Z_m approaches its minimum. When $T > T_c$, the above treatment of the linear spin-wave approximation (via the Holstein–Primakoff transformation) is no longer appropriate, since there is no longer long-range ferromagnetic order, and one has to deal with spin

operators rather than spin-wave operators. Recent experimental observations [25] showed that collective spin excitations also exist above the Curie temperature, which suggested that the composite polaron can persist for temperatures above T_c.

An interesting property of the CP is its transport properties in the presence of a magnetic field $\boldsymbol{B}$ at finite temperature T, which is closely related to the transport properties of lanthanum manganite, especially the CMR effect. It is found that the magnetoresistance behaviour of lanthanum manganites can be understood qualitatively within the present theory of composite polarons. The electric resistivity can be expressed as

$$\rho = 1/(ne\mu) \qquad \mu = e\tau/m^*$$

where μ is the mobility of the CP, and m^* is the effective mass which is proportional to the inverse of the bandwidth. According to equations (13) and (14), one can determine the mobility through the effective mass, $\mu \propto 1/m^* \propto Z_m Z_{ph}$. At temperatures below the Curie temperature T_c, there exist both spin waves and Jahn–Teller phonon excitations. As the temperature rises, more and more spin waves and phonons are excited, and the mobility of the CP becomes smaller. Consequently, the resistivity increases. The electric conductivity decreases with the increase of temperature, and reaches its maximum near the Curie point. Above T_c, the long-range magnetic ordering disappears, and only the Jahn–Teller phononic polaron plays a role. Like for usual polarons, the bonding strength of the distorted field and the electron decreases with the increase of the temperature, so the resistance declines with the increase of T in the high-temperature region. Due to the strong polaron self-trapping effect and its thermal activation, one would expect a great change in the resistance with the increase of temperature, and that the effect of the magnetic field in reducing the degree of the local spin fluctuation will reduce the bonding strength of the composite polaron, and hence decrease the resistivity. From the preceding discussions, it is found that the narrowing of the electronic bandwidth is a fundamental process controlling the transport properties of conduction electrons in ferromagnetic narrow-band metallic manganites, a point supported by recent experiment [26].

One could estimate the magnetoresistance change through the mobility of the CP. The mobility variation arises from two sources, the effective mass and the scattering lifetime:

$$\mu/\mu_0 = (m/m^*)\tau/\tau_0. \tag{15}$$

Here μ_0 and τ_0 denote the mobility and the scattering lifetime of free carriers. For typical electron–phonon coupling, $p \approx 1$, and the ferromagnetic coupling, $4zA = 30$ meV [20], when the temperature ranges from 50 to 250 K, the variation of the contribution of the effective mass to the mobility of the composite polaron decreases by about one order of magnitude, or $m/m^* \sim 0.1$. The effect of the magnetic field is not so significant as that of the temperature. At $T = 250$ K, the mobility contribution from the effective mass increases by a factor of two when the magnetic field increases from zero to 8 T. The reason for this is that the magnetic field only affects Z_m, while the temperature affects both Z_{ph} and Z_m. In fact, the huge change of the resistivity of the CP with the magnetic field and temperature may arise from the variation of the scattering lifetime, which can change by several orders of magnitude and drive a metal–insulator-like transition in doped manganites. Detailed studies will be published elsewhere.

4. Conclusion

To summarize, a theory of the composite polaron is developed. It is suggested that the composite polaron can be realized in lanthanum manganites, and contribute significantly to the unusual ground-state, thermodynamic, and transport properties of manganites.

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