Single hole motion in LaMnO$_3$

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We study single hole motion in LaMnO$_3$ using the classical approximation for JT lattice distortions, a modified Lang-Firsov approximation for dynamical breathing-mode phonons, and the self-consistent Born approximation (verified by exact diagonalization) for hole-orbital-excitation scattering. We show that in the realistic parameter space for LaMnO$_3$, quantum effects of electron-phonon interaction are small. The quasiparticle bandwidth $W \approx 2.2J$ in the purely orbital $t$-$J$ model. It is strikingly broadened to be of order $t$ by strong static Jahn-Teller lattice distortions even when the polaronic band narrowing is taken into account.

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LaMnO$_3$ is a typical parent compound of a class of colossal magnetoresistance materials $\text{R}_1-x\text{A}_x\text{MnO}_3$ ($\text{R} = \text{La}, \text{Pr}, \text{Nd}, \text{Sm}$ and $\text{A} = \text{Ca}, \text{Sr}, \text{Ba}$). As temperature decreases, it undergoes a structural phase transition at $T_c \approx 750$ K from cubic to tetragonal because of Jahn-Teller (JT) lattice distortions of its MnO$_6$ octahedra. Below $T_N \approx 140$ K, it is an $A$-type antiferromagnetic (AF) insulator. Staggered orbital ordering was recently observed in the ferromagnetic planes of LaMnO$_3$ [4], which can be driven by either the intra-atomic Coulomb interaction in the $e_g$ orbitals [3] or the cooperative JT splitting of the degenerate $e_g$ orbitals [4]. When electrons are removed from the $e_g$ orbitals upon doping, charge fluctuations as well as spin, orbital defects are introduced into the system. LaMnO$_3$ with one hole is one of the simplest real systems containing charge, spin, orbital, and lattice dynamics. The clear understanding of single hole motion in it is an essential step to the full understanding of abundant dopant-induced phase transition phenomena observed in this class of materials [1].

The Coulomb interaction ($\approx 7$ eV) in the $e_g$ orbitals eliminates doubly occupied sites and results in a two-dimensional anisotropic orbital $t$-$J$ model [5]. There exist off-diagonal transfer matrix elements, thus in principle a hole can hop without disturbing the staggered orbital background. If the orbitals were frozen, a hole would move freely and disperse with bandwidth $2t$, as described by the large-$U$ LDA+$U$ band calculation [6]. However, the low-energy physics may be controlled by two polarization effects: orbital polarization and lattice polarization. First, the motion of a hole will distort the orbital order via its diagonal transfer matrix elements; this will induce a strong polarized cloud of orbital excitations in the vicinity of the hole. The resultant quasiparticle (QP) is of orbital-polaron type with bandwidth $\propto J$ and a shift of spectral weight from the coherent to the incoherent part of the hole spectrum [7]. Second, when a hole is present, it attracts the surrounding oxygen ions equally, giving rise to a breathing distortion energy. In the single hole problem, charge fluctuations exactly locate at where the hole is and thus accompany the hole propagation. One thus needs to take into account dynamical breathing-mode phonons. Beyond a critical hole-phonon coupling strength, the hole is self-trapped in an “anti-JT” small polaron state via the polaronic band narrowing effect [8]. Therefore, the QP behavior is sensitive to model parameters. In this Letter, we present a systematic study on single hole motion in LaMnO$_3$ in a variety of parameter regions of physical interest.

The total Hamiltonian consider here is $H = H_{t-J} + H_{br} + H_{JT}$ [8], where

$$H_{t-J} = - \sum_{\langle ij \rangle \alpha \beta} (t_{ij}^\alpha \bar{d}_{ia}^\dagger \bar{d}_{jb} + \text{H.c.})$$

$$+ \frac{J}{2} \sum_{\langle ij \rangle \parallel} \{ T_{ij}^x T_{ij}^x + 3T_{ij}^y T_{ij}^y \mp \sqrt{3}(T_{ij}^x T_{ij}^z + T_{ij}^y T_{ij}^z) \},$$

$$H_{br} = - \sqrt{E_{br} \omega_0} \sum_i n_i^b (a_i^\dagger + a_i) + \omega_0 \sum_i a_i^\dagger a_i,$$

$$H_{JT} = 2 \lambda \sum_i (Q_{3i} T_{i}^z + Q_{3i}^\dagger T_{i}^z)^2 + \frac{1}{2} K \sum_i (Q_{3i}^2 + Q_{3i}^\dagger)^2,$$

where $\bar{d}_{ia}^\dagger = d_{ia}^\dagger (1 - n_i)$ is the constrained fermion operator for the $e_g$ electron at orbital $a$. $T_{ij}^x = (\bar{d}_{i\uparrow}^\dagger \bar{d}_{j\uparrow} - \bar{d}_{i\downarrow}^\dagger \bar{d}_{j\downarrow} + \bar{d}_{i\downarrow} \bar{d}_{j\uparrow})/2$ and $T_{ij}^z = (\bar{d}_{i\uparrow}^\dagger \bar{d}_{j\downarrow} - \bar{d}_{i\downarrow}^\dagger \bar{d}_{j\uparrow} + \bar{d}_{i\downarrow} \bar{d}_{j\uparrow})/2$ are orbital-pseudospin operators with $| \uparrow \rangle = d_{x^2-y^2}^\dagger$ and $| \downarrow \rangle = d_{3z^2-r^2}^\dagger$. The anisotropic transfer matrix elements are $t_{ij}^\dagger = 3t/4$, $t_{ij}^\perp = t$, and $t_{ij}^\parallel = t$. The $\mp$ sign distinguishes hopping along the $x$ and $y$ directions. The orbital superexchange $J$-induced orbital ordering reproduces the experimental observation: $(| \uparrow \rangle + | \downarrow \rangle)/\sqrt{2}$ and $(| \uparrow \rangle - | \downarrow \rangle)/\sqrt{2}$ in the $A$ and $B$ sublattices respectively [10]. In $H_{br}$, $n_i^b = 1 - \sum_a \bar{d}_{ia}^\dagger \bar{d}_{ia}$ is the hole number operator. $a_i^\dagger$s are the breathing-mode phonon operators with frequency $\omega_0$. $E_{br}$ is the hole-phonon coupling strength. In $H_{JT}$ [8], $Q_{3i}$ and $Q_{3i}^\dagger$ are, respectively, JT distortions for the $3z^2-r^2$ and $x^2-y^2$ modes satisfying $Q_{3i} = q \cos \psi_i$ and $Q_{3i}^\dagger = q \sin \psi_i$ with $\psi_{i\in A} = - \psi_{i\in B}$ [11]. For simplicity, $\psi_{i\in A}$ is set to be $\pi/2$ so that the orbital ordering driven by the cooperative JT effect also agrees with the experiment.

We employ the slave-fermion formalism to cope with
the constraint of no doubly occupancy [13]. First, we perform a uniform rotation of orbitals by 90° about the $T^y$ axis: $d_{1t} \rightarrow (\bar{c}_{1t} - \bar{c}_{1u})/\sqrt{2}$, $d_{3z} \rightarrow (\bar{c}_{1t} + \bar{c}_{1u})/\sqrt{2}$, $T^z_i \rightarrow -\bar{T}^z_i$, $T^x_i \rightarrow \bar{T}^x_i$ in order to obtain the Néel configuration $\ldots T^z_i T^{x}_{i+1} T^z_{i+2} T^{x}_{i+3} \ldots = \ldots \downarrow \uparrow \downarrow \uparrow \ldots$. Then, considering the Néel state as the vacuum state, we define holon (spinless fermion) operators $h_1$ so that $\bar{c}_{1t} = h^\dagger_1 h_1$ on the ↓ sublattice and $\bar{c}_{1u} = h^\dagger_1 h_1$ on the ↑ sublattice. Here $b_1 = \bar{T}^z_i$ on the ↓ sublattice and $\bar{T}^z_i$ on the ↑ sublattice are hard-core boson operators. Such a treatment is in spirit similar to that done by Schmitt-Rink et al. for one hole motion in a quantum antiferromagnet [13, 17].

Following Röder, Zang, and Bishop [12], we treat dynamical phonons within the modified Lang-Firsov approximation using a canonical transformation $\mathcal{H} = U^\dagger H U = e^{-S_1(\Delta)} e^{-\frac{1}{2} S_2(\tau)}$, where $S_1(\Delta) = \Delta / \sqrt{2 E_{\text{br}} \omega_0} \sum_i (\alpha_i^2 - \alpha_i)$ with $\Delta$ measuring static breathing-mode distortions and $S_2(\tau) = -\tau \sqrt{E_{\text{br}} / \omega_0} \sum_i h^\dagger_i h_i (\alpha_i^2 - \alpha_i)$ with $\tau$ measuring the degree of the polaron effect. Averaging $\mathcal{H}$ over the phonon vacuum, we arrive at an effective orbital-lattice-polaron Hamiltonian $\mathcal{H} = E_0(\tau) + H_{\text{eff}}$

$$H_{\text{eff}} = \sum_{k} \varepsilon_k(\tau) h^\dagger_k h_k + \sum_{q} \omega_q \beta^\dagger_q \beta_q$$

$$+ \sum_{kq} h^\dagger_k h_{k-q} (M_{kq} \beta^\dagger_q + N_{kq} \beta_q + Q) + \text{H.c.} \quad (2)$$

where $Q = (\pi, \pi)$. $E_0(\tau) = N \varepsilon_k[1 - \varepsilon_k(1 - \tau) \Delta + \Delta^2/4 E_{\text{br}} - E_{\text{br}} (2\tau - \tau^2)]$ with $x = 1/N$ being the hole concentration and $\Delta = 2E_{\text{br}}(1 - \tau)x$. The hole-orbital-wave coupling functions are $M_{kq} = \frac{2}{\sqrt{N}} (\gamma_k \alpha_q + \gamma_q \alpha_k)$, $N_{kq} = \frac{-\sqrt{2}}{\sqrt{N}} (\gamma_k \beta_k + \gamma_q \beta_q)$, with $\omega_q = \left((A_q + B_q)/\omega + 1/2\right)^{1/2}$ and $x = -\text{sgn}(B_q) \left((A_q + B_q)/\omega - 1/2\right)^{1/2}$.

Here short notations are $\gamma_k = (c_{kx} + c_{ky})/2$, $\gamma_q = (c_{kx} - c_{ky})/2$, $A_q = 3J + E_{\text{JT}}$, and $B_q = J_q/2$. The $\beta_q$’s are orbital wave operators, $h^\dagger_q = u_q \beta^\dagger_q + v_q \beta^\dagger_q$, with dispersion $\omega_q = \sqrt{A_q (A_q + 2B_q)}$. The bare hole dispersion is $\varepsilon_k(\tau) = -\xi(\tau) \gamma_k$ with $\xi(\tau) = \exp(-E_{\text{br}} \tau^2/\omega)$ being the polaronic band narrowing. $\xi(\tau)$ is determined by the following equation [12],

$$E_{\text{br}} = -\omega_0 \ln \xi \left[ 1 - \frac{1}{(1 + x \omega_0)} \frac{\partial E_{\text{min}}(\xi)}{\partial \xi} \right]^2, \quad (3)$$

where $E_{\text{min}}(\xi)$ is the minimum of the QP dispersion $E_k \equiv \varepsilon_k + \text{Re}\Sigma(k, E_k)$ with $\Sigma(k, \omega)$ being the holon self-energy. Note that in this effective Hamiltonian, the JT effect acts as $E_{\text{JT}} = 2\Delta^2/\bar{K}$ in $A_q$.

We calculate the holon Green’s function $G(k, \omega) = [\omega - \varepsilon_k - \Sigma(k, \omega)]^{-1}$ treating the hole-orbital-wave coupling within the self-consistent Born approximation (SCBA) [13, 17]. The self-energy is thus of the form

$$\Sigma(k, \omega) = \sum_q |M_{kq}^2 G(k - q, \omega - \omega_q)|$$

$$+ |N_{kq}^2 G(k - q, \omega - \omega_q + Q)|. \quad (4)$$

We test the applicability of the SCBA by numerically diagonalizing $H_{t-J}$ on 16- and 18-site clusters. In addition, since in the SCBA the AP orbital background is treated within linear orbital-wave theory, the mixed terms $\propto T^z_i T^z_{i+1} T^z_{i-1}$ in $H_{t-J}$, which contribute only in higher order orbital-wave theory, are neglected in the SCBA. It is thus interesting to numerically diagonalize a “truncated” Hamiltonian without these terms [13]. As shown in Fig. 1 and Fig. 2, we find that all of the SCBA
results are in good agreement with the exact diagonalization (ED) results, especially with those for the truncated Hamiltonian as expected.

The physically relevant parameter space for LaMnO$_3$ are: $t \sim 0.72$ eV (energy unit) and $0.1 \leq J \leq 0.3$ are estimated from photoemission experiments [18]. $\omega_0 = 0.1$ is taken from Raman data [13]. $E_{JT} = 1.5$ and $E_{br}$ is of order $E_{JT}$ [14,15]. Numerical studies found that the $A$-type spin and $C$-type orbital structures were stabilized in this region of parameter space [3]. Below let us discuss the QP properties in different parameter regions of interest. All calculations are performed on the $16 \times 16$ lattice unless noted.

First, we consider the single hole motion in the pure orbital $t$-$J$ model (i.e. $E_{br} = E_{JT} = 0$). As displayed in Fig. 1, at any $k$, there is a well-defined quasiparticle pole (i.e., zero orbital-wave) at the low energy side which is well separated from a broad, incoherent, multiple-orbital-wave background extending to the full free-electron bandwidth. The bottom and the top of the quasiparticle (QP) band locate at $(0,0)$ and $(\pi,\pi)$, respectively. In Fig. 2, the spectrum of the orbital polaron is flat at large momenta, which leads to a strongly distorted density of states with a massive peak at the top of the QP band. In the realistic range of $0.01 \leq J \leq 0.5$, the QP bandwidth $W \simeq 2.2J$ scales with $J$ (see Fig. 3). This new low energy scale is quite similar to that in the cuprate $t$-$J$ model where the spin is conserved during hole hopping [14,15]. In the latter, the staggered spin background is disturbed by hole propagation, leading to a vanishing bare hole dispersion $\varepsilon_k$, and thereafter restored by Heisenberg spin flipping, forming a QP band with width $\sim 2J$ (the characteristic energy of spin waves). In the present case of $\varepsilon_k = -t |k|$, the energy scale of $W \simeq 2.2J$ can be understood in the following way: For small $J$, orbital excitations are easily stimulated by incoherent hole motion and accompany the hole propagation. Thus the hole spectral weights $Z(k) = (1 - \delta \Sigma(k,\omega)/\delta \omega)^{-1}$ are strongly reduced by such a cloud of polarized orbital waves. The reduced weights by incoherent hole motion can be approximately obtained by neglecting $\varepsilon_k$ in the calculation, referred to as $Z_0(k)$. Then one can naively express the QP dispersion as $Z_0(k)\varepsilon_k$. In the range of $0.01 \leq J \leq 0.5$, $Z_0(k) \simeq 1.3J/t$. Therefore, $W$ scales with $J$.

Second, we consider the case of strong static JT distortions. The contribution of static JT interaction is adding an Ising-like component to the excitations and inducing a large gap in the orbital excitation spectrum. Thus, the JT effect stabilizes the orbital ordering. As a result, even for small $J$, orbital excitations are difficultly stimulated. In fact, for $A_q \gg \xi(\tau)t$ due to either large $E_{JT}$ or large $E_{br}$, Eq. (3) can be solved analytically in perturbation theory: $E_k \simeq -\xi(\tau)t\gamma_k - O(\xi^2t^2/A_q)$. Thus, the QP band is narrowed mainly by lattice polarons instead of orbital polarons. The QP spectral weight is $\xi(\tau) - O(\xi^2t^2/A_q^2)$ approximately. The QP bandwidth scales with $t$. As illustrated in Fig. 3, $W \geq 1.5t$ is a slowly changing function of $J$ at $E_{JT} = 1.5$. Fig. 4(a) shows $W$ and $\xi(\tau)$ as a function of $E_{br}$ at $E_{JT} = 1.5$ and $\omega_0 = 0.1$; they behave similarly in general, indicating that lattice polarization overwhelms orbital polarization in a wide range of $E_{br}$. For not too large $E_{br}$, the small deviation of $W/2t$ from $\xi(\tau)$ is attributed to the remnant hole-orbital-wave coupling. $W$ decreases exponentially with $E_{br}$ for $E_{br} > 12.5$. This is the Lang-Firsov band narrowing. However, for the realistic value of $E_{br} < 5$, $W > 1.2t$. Therefore, in the presence of strong static JT distortions, the QP bandwidth is strikingly broadened in comparison with $W \simeq 2.2J$ obtained in the purely orbital $t$-$J$ model even when the quantum effect of polaronic band narrowing is taken into account.

Third, it is interesting to examine the case of no static JT distortions (i.e., $E_{JT} = 0$), which is relevant to the experimental fact that the static JT distortion rapidly disappears around $x \sim 0.1$ in La$_{1-x}$Sr$_x$MnO$_3$ [24]. For not too large $E_{br}$, there is mix of polarization of phonons.

![FIG. 3. QP bandwidth $W$ as a function of $J$ at $E_{br} = 0$.](image)

![FIG. 4. The variation of bandwidth $W$ and the polaronic narrowing $\xi(\tau)$ with $E_{br}$ at $J = 0.3$ for (a) $\omega = 0.1$ and (b) $\omega = 0.5$.](image)
and orbital-waves. Fig. 4(a) shows the QP bandwidth $W$ as a function of $E_{br}$ at $\omega_0 = 0.1$. For $E_{JT} = 0$, $W/2t$ remains almost unchanged ($\sim 1.1J$) in the range of $0 \leq E_{br} \leq 10$ and obviously deviates from the polaronic band narrowing $\xi(\tau)$ up to $E_{br} \simeq 17$. This implies that the QP is of orbital-polaron type for $E_{br} < 10$ and is of polaron type for $E_{br} > 17$ as well as a mix of the two types for $10 < E_{br} < 17$. Therefore, for realistic value of $E_{br} < 5$, the electron-phonon interaction effect on the QP band can be neglected. The hole-orbital-wave scattering dominates the formation of the QP.

Finally, it should be made clear that the small value of $\omega_0 = 0.1$ (which is however the highest value of phonon frequency observed in Raman spectra [12]) underlies the unimportance of quantum effects of electron-phonon interaction on the QP band. Note that the lattice polaron effect is the most pronounced in the adiabatic limit ($\omega_0 \rightarrow \infty$), where $\gamma \rightarrow 1$, and is negligible in the adiabatic limit ($\omega_0 \rightarrow 0$), where $\gamma \rightarrow 0$. At $\omega_0 = 0.1$, $\tau = 0.1$, the system is close to adiabatic limit. Fig. 4(b) shows the same quantities as Fig 4(a) but at $\omega_0 = 0.5$ which is chosen in Ref. [12]. The quantum effects of hole-phonon interaction are improved remarkably. For example, the critical values of $E_{br}$ at which different QP types occur are 3 times as smaller as those at $\omega_0 = 0.1$.

Summarizing, we present a systematic study on single hole motion in LaMnO$_3$. We show that in the realistic parameter space for LaMnO$_3$, quantum effects of hole-phonon interaction are small. In the purely orbital $t$-$J$ model, the QP bandwidth $W$ scales with $J$. Considering the hole-phonon coupling, $W$, which however scales with $t$, is strikingly broadened by strong static JT lattice distortions even when the polaronic band narrowing is taken into account. We predict that orbital polarization is pronounced for $x > 0.1$ where static JT distortions disappear. Our results can be tested by future angle-resolved photoemission spectroscopy (ARPES) experiments.

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Note added.—Very recently, two papers concerning ARPES of LaMnO$_3$ have appeared. Perebeinos and Allen [10] addressed the Frank-Condon broadening effect driven by electron-phonon interaction on the bare hole dispersion in the JT-ordered ground state. Brink, Horsch, and Oleś (BHO) [2] calculated the single-hole spectral functions for the purely orbital $t$-$J$ model using the SCBA and discussed the crystal-field effect on the QP band. Their pieces of work are in a sense complementary to ours. The present work takes into account both of orbital polarization and lattice polarization, giving rise to a different, yet more realistic picture from BHO’s.