Hole spectral functions of LaMnO$_3$

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By use of the orbital $t$-$J$ model, we calculate the photoemission spectra of LaMnO$_3$ using the exact diagonalization technique, and interpret our numerics quite well in the orbital-polaron scenario where the scattering between holes and orbital excitations is treated within the self-consistent Born approximation. The quasiparticle bandwidth is found to be of the order of $J$ and $t$ in the purely Coulombic and Jahn-Teller phononic model, respectively. We suggest that angle-resolved photoemission spectroscopy experiments allow one to distinguish between the orbital-polaron scenario and the Jahn-Teller polaron scenario.

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Orbital Physics is a key concept for recent intensive studies on transition-metal oxides [1], especially on manganese oxides with perovskite structure $R_1-x$A$_2$MnO$_3$ ($R = $ La, Pr, Nd, Sm and $A = $ Ca, Sr, Ba) due to the discovery of colossal magnetoresistance in this class of materials. Mn$^{3+}$ in RMnO$_3$ has four $d$ electrons of which three are put into the $t_{2g}$ orbitals and form an $S = 3/2$ localized spin, and the mobile one occupies one of the $e_g$ orbitals ($d_{x^2-y^2}$ and $d_{z^2-r^2}$). This $e_g$ orbital degree of freedom may cause new phenomena through strong coupling with charge, spin, and lattice dynamics [2]. For example, alternating orbital order was observed in the ferromagnetic (FM) planes of LaMnO$_3$ [3] and uniform $x^2-y^2$ orbital order in $A$-type AF Pr$_{1/2}$Sr$_{1/2}$MnO$_3$ [4], while orbital liquid was proposed to exist in metallic FM La$_{1-x}$Sr$_x$MnO$_3$ ($x > 0.2$) [5]. These discoveries attract attention to the orbital correlation, dynamics and order-disorder transition. An essential first step is to understand the motion of a hole in an orbital ordered system such as the FM planes of LaMnO$_3$. We address it in this Letter.

Orbital ordering in LaMnO$_3$ can be induced by either the Jahn-Teller (JT) lattice distortion originating from the degeneracy of the $e_g$ orbitals $\{2\}$ or the intra-atomic Coulomb interaction in the $e_g$ orbitals $\{3\}$. In the latter mechanism, the Coulomb interaction eliminates doubly occupied sites and results in the orbital superexchange interaction $\{4\}$. The experimental counterpart of this problem is angle-resolved photoemission spectroscopy (ARPES) measurements. ARPES experiments have not yet been reported in this doping regime. Thus, we perform here computational experiments to explore unbiased information. Then, we satisfactorily interpret the outcome of such experiments in an orbital-polaron picture where the scattering between holes and orbital excitations is treated within the self-consistent Born approximation. Furthermore, we point out that although the JT lattice distortion leads to a large gap in the orbital excitation spectrum and thus resists the formation of the orbital-polaron, the hole can move almost freely through the orbital-flip process. The quasiparticle bandwidth is of the order of $J$ ($t$) in the purely Coulombic (Jahn-Teller phononic) model, respectively. Our results indicate that ARPES may provide a possible approach to distinguish between the Coulombic scenario and the Jahn-Teller scenario.

The orbital $t$-$J$ model is [10,11,12]

$$H = -\sum_{\langle ij\rangle \langle ab \rangle} \langle \mathbf{i} \mathbf{j} \rangle d^\dagger_{\mathbf{i} a} \tilde{d}_{\mathbf{j} b} + \text{H.c.} + \frac{J}{2} \sum_{\langle ij \rangle \langle k l \rangle} \left[ T_{ij}^a T_{lj}^b + 3 T_{ij}^a T_{lj}^c + \sqrt{3}(T_{ij}^a T_{lj}^d + T_{ij}^d T_{lj}^a) \right],$$

where $\tilde{d}^\dagger_{\mathbf{i} a} = d^\dagger_{\mathbf{i} a} (1 - n_{\mathbf{i} a})$ is the constrained fermion operator for the $e_g$ electron at orbital $a$. $T_{ij}^a = (\tilde{d}^\dagger_{\mathbf{i} a} \tilde{d}_{\mathbf{j} a} - \tilde{d}^\dagger_{\mathbf{j} a} \tilde{d}_{\mathbf{i} a})/2$ and $T_{ij}^c = (\tilde{d}^\dagger_{\mathbf{i} a} \tilde{d}_{\mathbf{j} c} + \tilde{d}^\dagger_{\mathbf{j} c} \tilde{d}_{\mathbf{i} a})/2$ are orbital-pseudospin operators with $| \uparrow \rangle = d_{x^2-y^2}$ and $| \downarrow \rangle = d_{z^2-r^2}$. The anisotropic transfer matrix elements are $t_{ij}^\uparrow = 3t/4$, $t_{ij}^\downarrow = t/4$, and $t_{ij}^{\uparrow \downarrow} = t_{ij}^{\downarrow \uparrow} = \mp \sqrt{3}t/4$, here the $\mp$ sign distinguishes hopping along the $x$ and $y$ directions. The orbital superexchange interaction $J = t^2/(U' - J')$ with $U$ ($J'$) being the interorbital Coulomb (exchange) integral [12,13]. In LaMnO$_3$, the realistic parameters are estimated from photoemission experiments [14] to be $t \sim 0.72$ eV, $U \sim 5$ eV, $J' \sim 2$ eV, thus $J \approx 0.24t$.

The photoemission spectrum (PES) $(\tilde{d}^\dagger_{\mathbf{i} a} \tilde{d}_{\mathbf{k} a}/\omega)$ is calculated by using the standard Lanczos algorithm [12]. There are two bands and we find $(\tilde{d}^\dagger_{\mathbf{k} 1} \tilde{d}_{\mathbf{k} 1}/\omega) = \ldots$
a new orbital basis, we perform a uniform rotation of orbitals by 90° about the \( T^y \) axis: \( \vec{d}_{i\uparrow} \rightarrow (\vec{d}_{i\uparrow} - \vec{d}_{i\downarrow})/\sqrt{2} \), \( \vec{d}_{i\downarrow} \rightarrow (\vec{d}_{i\uparrow} + \vec{d}_{i\downarrow})/\sqrt{2} \), and thus \( T_i^z \rightarrow -T_i^x \). This is the interorbital hopping in the rotated basis and causes the bare hole dispersion as shown below. Orbital excitations can be described within the linear spin-wave theory by defining \( T_i^+ = \vec{d}_{i\uparrow} \), \( T_i^- = \vec{d}_{i\downarrow} \), \( T_i^\mp = \vec{d}_{i\uparrow} \pm \frac{1}{\sqrt{2}} \vec{d}_{i\downarrow} \) on the sublattices, and \( T_i^z = \vec{f}_{ij} \). We introduce the orbital Neél state as the vacuum state, we define holon operators \( \vec{t}_i \) similar to those in the spin-polaron scenario \[5\] so that \( \vec{d}_{i\uparrow} = \vec{f}_i \), \( \vec{d}_{i\downarrow} = \vec{f}_i \) on the sublattices, and \( \vec{d}_{ij} = \vec{f}_j \) on the \( ij \) bond.

Introducing the new fermion operators \( \{ f^\dagger_k, h_k \} \) in the momentum space \( f_k = (f_k + h_k)/\sqrt{2} \), \( f_k = (f_k - h_k)/\sqrt{2} \), we arrive at an effective orbital-polaron Hamiltonian

\[
H_{\text{eff}} = \sum_i \varepsilon_k (f_i^\dagger f_i - h_i^\dagger h_i) + \sum_{q} \left( \omega^\alpha_{q\alpha} \alpha^\dagger_q \alpha_q + \omega^\beta_{q\beta} \beta^\dagger_q \beta_q \right) + \sum_{q} \left( f_i^\dagger f_i - h_i^\dagger h_i \right) (\gamma^\alpha_{q\alpha} \alpha_q + \gamma^\beta_{q\beta} \beta_q + \text{H.c.}) (2)
\]

where the bare hole dispersion is \( \varepsilon_k = -t\gamma_k \). The holon-orbital-wave coupling functions are \( \gamma^\alpha_{q\alpha} = \frac{2\pi}{\sqrt{N}} (\gamma_n v_n^\alpha + \gamma_{-n} v_n^\alpha) \), \( \gamma^\beta_{q\beta} = \frac{2\pi}{\sqrt{N}} (\eta_n v_n^\beta + \eta_{-n} v_n^\beta) \), \( \rho^\alpha_{q\alpha} = \frac{2\pi}{\sqrt{N}} (\rho_n v_n^\alpha + \rho_{-n} v_n^\alpha) \), and \( \omega^\alpha_{q\alpha} = \frac{2\pi}{\sqrt{N}} (\omega_n v_n^\alpha + \omega_{-n} v_n^\alpha) \).

To get the physical insight of the above numerics, we perform an analytical calculation of the QP spectral functions in an orbital-polaron scenario. The \( J \) term induces the orbital Neél state, i.e., the alternation of orthogonal orbitals in the ground state. Here we analyze our problem to that of a hole moving in the AF spin background, an essential problem in the field of high-temperature superconductivity \[1\]. The latter can be accurately understood in the spin-polaron scenario where holes are described as spinless fermions (holons) and spins as hard-core bosons. The scattering between holes and spin-wave excitations is treated within the self-consistent Born approximation (SCBA) \[1\]. In the SCBA, higher order spin-wave excitations are also neglected, consistent with our ED results. Hence, we expect that our numerics may be interpreted in a similar way.

In LaMnO\(_3\), alternately occupying orbitals on the two sublattices are oriented in the FM planes: \( \{| \uparrow \uparrow \downarrow - | \downarrow \downarrow \uparrow \}/\sqrt{2} \) and \( \{| \uparrow \downarrow \downarrow \uparrow \}/\sqrt{2} \). This state is different from the alternating orbital states \( 3x^2 - r^2 \) and \( 3y^2 - r^2 \), which have been naively expected \[1\]. To obtain the Neél configuration \( |T^x_i \uparrow i_{i+1} T^x_i \uparrow \cdots \rangle = \downarrow \uparrow \downarrow \uparrow \cdots \) in
\[ \varepsilon_k + \Sigma_f(k, E_f^k) \] (see Fig. 2) and the spectral weights
\[ Z(k) = \left[ 1 - \frac{\delta \Sigma(k, \omega)}{\partial \omega} \right]^{-1} \omega = E_k \] (see Table I). All of the SCBA results are in good agreement with the ED results, especially with those for the truncated Hamiltonian as expected. Therefore, the orbital-polaron scenario may provide a valuable scheme for further works on the orbital dynamics.

In the rest of this Letter, we discuss the Jahn-Teller effect on the quasiparticle band. Recently, Bala and Oleš suggested that a large gap in the orbital excitation spectrum induced by the JT lattice distortion might lead to a strong confinement of holes in lightly doped LaMnO\(_3\) insulators [13]. This implies that the hole quasiparticle bandwidth is more narrow in the presence of JT phonons. We include the JT effect in the same way as Bala and Oleš but we get a different result. The Jahn-Teller interaction considered here is [14]

\[ H_{JT} = -2E_{JT}(\phi, \delta_x, \delta_z, u)(\sum_{i \in A} T^i_{\phi} - \sum_{j \in B} T^j_{\phi}). \] (6)

Here \( T^i_{\phi} \) refers to the rotated basis. \( E_{JT}(\phi, \delta_x, \delta_z, u) = \lambda[(\delta_x - \delta_z)^2 \sin 2\phi - 2\sqrt{3}u \cos 2\phi] \) acts as a fictitious “magnetic field” in which \( \phi \) is the tilting angle of pseudospins, and \( \delta_x, \delta_z, u \) characterize lattice distortions (in units of the lattice constant): \( \delta_x, \delta_z \) — uniform deformation along the \( x \) and \( y \) (\( z \)) directions, and \( u \) — oxygen ionic displacement along Mn-O-Mn bond in the \( xy \) plane [19]. The distorted lattice energy per Mn ion is

\[ E_l(\delta_x, \delta_z, u) = K_1(\frac{1}{2}\delta_x^2 + 2u^2 + \frac{1}{2}\delta_z^2) + K_2(\delta_x^2 + \frac{1}{2}\delta_z^2), \]

where \( K_1(\delta_x) \) is the nearest-neighbor Mn-O (Mn-Mn) spring constant. To estimate the JT effect on the quasiparticle behavior, we may consider the case of \( \phi = 0 \) (i.e., no static distortions due to a tetragonal field) without loss of generality. In the classical ground state at \( \phi = 0 \), \( E_{JT} = -3\lambda^2/K_1 \). For the realistic parameters of LaMnO\(_3\); the spring constant \( K_1 = 200 \) eV and the JT interaction parameter \( \lambda \geq 6 \) eV [18, 16], \( E_{JT} = -0.54 \) eV.

In order to calculate \( E_f^k \) in the presence of JT lattice distortion, all we have to modify the above derivation is to make the following replacement in [3] and [8]: \( A_q \rightarrow 3J - 2E_{JT} = 3J + 6\lambda^2/K_1 \). The JT interaction adds an Ising-like component to the excitations and induces a large gap in the orbital excitation spectrum. Thus, the JT effect stabilizes the orbital ordering.

The SCBA results on the 20 \( \times \) 20 lattice are summarized in Table I. The large gap in the orbital excitation spectrum induced by the JT lattice distortion does not weaken the QP bandwidth. Instead, at \( \lambda = 6 \) eV the QP bandwidth reaches 75 percent of the width (2\( t \)) of the bare hole dispersion \( \varepsilon_k \). Although the large gap in the orbital excitation spectrum resists the formation of the orbital polaron, the hole can move almost freely via the orbital-flip process in the rotated orbital basis. For \( A_q \gg t \) because of large \( J \) or large \( \lambda \), Eq. (9) can be solved analytically in perturbation theory. Most of the spectral weight, \( 1 - O(t^2/A_q^2) \), appears in the quasiparticle part of \( G_f(k, \omega) \) which behaves indeed a bare dispersion \( E_f^k \simeq \varepsilon_k - \sum_q (\frac{\rho_{kq}^\delta}{|\rho_{kq}^\delta|^2})/\omega^2 \simeq \varepsilon_k - O(t^2/A_q) \). Fig. 4 shows the QP bandwidth \( W \) as a function of \( J \). Without the JT interaction (\( \lambda = 0 \) eV), \( W \simeq 2.2J \) and the range \( J \) in the range of 0.01 \( \leq J \leq 0.4 \), and approaches to the width of the bare hole dispersion (2\( t \)) for large \( J \). On the other hand, with strong JT interaction (\( \lambda = 6 \) eV), \( W \) is of the order of \( t \) in the whole range of \( J \). These results reminisce those for one hole moving in the spin \( t-t'-J \) model (the \( t-t'-J_z \) model with large \( J_z \)), respectively, where the transfer to next nearest neighbors (\( t' \)) provides the bare hole dispersion [11, 14]. Therefore, the QP bandwidth for realistic \( J \) is of the order of \( t \) (\( J \)) in the presence (absence) of strong Jahn-Teller interaction. Our results can be tested by future ARPES experiments.

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[19] When a hole is present on site \( i \), it attracts the surrounding oxygen ions equally, giving rise to a breathing distortion energy. Since the breathing mode does not distinguish between the \( e_g \) orbitals, it can be safely neglected in the present study.

TABLE I. Bandwidth \( W \) and spectral weights \( Z(k) \) as a function of the Jahn-Teller interaction parameter \( \lambda \), \( J = 0.3t \).

| \( \lambda \) (eV) | \( W/t \) | \( Z(0,0) \) | \( Z(\pi/2, \pi/2) \) | \( Z(\pi, \pi) \) | \( Z(\pi, 0) \) |
|------------------|-----------|----------------|----------------|----------------|----------------|
| 0                | 0.65210   | 0.65989        | 0.37446        | 0.09774        | 0.39068        |
| 4                | 1.12691   | 0.79211        | 0.60401        | 0.30239        | 0.60701        |
| 6                | 1.49980   | 0.87222        | 0.77294        | 0.57996        | 0.77022        |
| 8                | 1.74565   | 0.92421        | 0.88066        | 0.80063        | 0.87943        |

FIG. 1. The PES \( \langle \tilde{d}^\dagger_{kx} \tilde{d}_{kx} \rangle_{\omega} \) for the orbital \( t-J \) model on the \( 4 \times 4 \) cluster using the ED technique for the full Hamiltonian (solid lines) and the truncated Hamiltonian (dashed lines). Here \( J = 0.3t \).
FIG. 2. Hole quasiparticle dispersion for the orbital $t$-$J$ model with $J = 0.3t$: The ED results for the full Hamiltonian (solid squares) and the truncated Hamiltonian (solid circles); The SCBA results on the $4 \times 4$ cluster (open circles) and the $20 \times 20$ one (open squares). Solid lines are the fits using $E_k^f = a_0 + a_1 \gamma_k + a_2 \cos k_x \cos k_y + a_3 \gamma_k$.

FIG. 3. The PES $A_f(k, \omega)$ for the orbital $t$-$J$ model on the $4 \times 4$ cluster calculated within the SCBA. Here $J = 0.3t$.

FIG. 4. The quasiparticle bandwidth $W$ as a function of $J$ at different Jahn-Teller interaction parameters: $\lambda = 0$ (solid squares) and $\lambda = 6$ eV (open circles). Calculations are performed on the $16 \times 16$ cluster within the SCBA.