

Orbital liquid in perovskite transition-metal oxides

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We study the effects of the degeneracy of the e_g orbitals in perovskite transition-metal oxides in the limit of strong repulsive electron-electron interaction. The isospin field is introduced to describe the orbital degrees of freedom and is represented by the boson with a constraint. The dispersion of this boson is flat along $(\pi/a, \pi/a, k_z)$ (a : lattice constant) and the other two equivalent directions. This enables the orbital disordered phase down to low temperatures. Some of the anomalous experiments in the low-temperature ferromagnetic phase of $\text{La}_{1-x}\text{Sr}_x\text{MnO}_3$ are interpreted in terms of this orbital liquid picture. The coupling between the orbital degrees of freedom and the Jahn-Teller lattice distortion and its implications to the optical conductivity are also discussed. [S0163-1829(97)04426-3]

I. INTRODUCTION

The colossal negative magnetoresistance observed in transition-metal oxides with perovskite structure, e.g., the double-exchange system $\text{La}_{1-x}\text{Sr}_x\text{MnO}_3$, has revived the interest in these systems.¹⁻⁴ The valence of Mn ion is Mn^{3+} for $x=0$, whose electron configuration is $(t_{2g})^3(e_g)^1$, and all the spins are aligned ferromagnetically due to the strong Hund coupling. Mn^{3+} ion also has the orbital degrees of freedom due to the degeneracy of the e_g orbitals. Because of the strong on-site Coulomb repulsion, the double occupancy of e_g orbitals is forbidden,^{5,6} and the system is a Mott insulator. Then the perturbative theory is not reliable to treat the electron-electron interaction in this system, and the strong-coupling theory is required. When one La is replaced by Sr, one hole is introduced to Mn and Mn^{3+} turns into Mn^{4+} . These doped holes, which contribute to the conduction, cannot be described in terms of the one-body theory. For a single band case, which is relevant to high- T_c cuprates, extensive studies have been focused on the doped holes to a Mott insulator. On the other hand, in the orbital-degenerate case, the nature of the mobile hole and the spin and orbital fluctuations in the doped Mott insulator has not been fully discussed up to now.⁷

Experimentally the orbital ordering has been established in the low hole-concentration region, i.e., $x \sim 0.0$,^{8,9} where the system is insulating with the A-type antiferromagnetic long-range ordering.¹⁰ As x increases the system becomes more and more conductive, and finally shows the metallic conduction below the ferromagnetic transition temperature T_c . In this metallic state ($x > 0.2$), there are several anomalous features.

(1) The optical conductivity shows a broad ‘‘Drude-like’’ band up to around 1 eV which is growing with decreasing temperature.¹¹ The integrated oscillator strength for this broad band and a narrow coherent peak changes down to the very low temperature where the ferromagnetic moment already saturates, which suggests that other degrees of freedom still remain active.

(2) The photoemission spectra shows only a small discontinuity at the Fermi energy E_F followed by a gaplike behavior.¹²⁻¹⁴ On the other hand, the specific heat is not

enhanced with $\gamma \sim 5$ mJ/K²mol.¹⁵

(3) The anisotropy of the conduction and the spin excitations, which is expected with the orbital ordering, is not observed even at low temperatures.^{15,16}

(4) There is no symptom of the Jahn-Teller distortion in neutron-scattering experiments.¹⁶ The displacements of the oxygen ions are independent of the temperature across the ferromagnetic transition temperature.

It is noted that the on-site repulsive interaction within each orbital plays no role in the ferromagnetic state because of Pauli’s exclusion principle. Then it is suggested from the above features that the orbital degrees of freedom play some roles in the low-temperature phase.

In this paper we study the fluctuation in the orbital degrees of freedom in perovskite transition-metal oxides. The mean-field and/or random-phase-approximation picture starting from the noninteracting electrons leads to the conclusion that the orbital degrees of freedom are ordered because of the strong electron-electron interaction and/or the Jahn-Teller coupling. However we present below several theoretical reasons to expect that the fluctuation is large enough to prevent the ordering although the amplitude of the short-range order is large. The orbital degrees of freedom are represented by the isospin \vec{T} , which is the quantum dynamical variable with $T=1/2$ for the e_g orbitals. We found that its fluctuation has a two-dimensional character. Based on this, we propose that the isospin is still disordered, i.e., liquid state, in the ferromagnetic state of $\text{La}_{1-x}\text{Sr}_x\text{MnO}_3$ with $x > 0.2$. We have done some numerical simulations based on this idea, and their results are at least encouraging. We also mention the coupling between the orbital and lattice degrees of freedom and also the quantum nature of the lattice distortion is relevant and the vibronic state remains down to low temperature. Its implications to the characteristic features in the optical conductivity are also discussed.

II. FORMULATION

In order to study the fluctuation in the orbital degrees of freedom, we start with the following Hamiltonian:

$$H = \epsilon_d \sum_{i,\sigma,\gamma} d_{i\gamma\sigma}^\dagger d_{i\gamma\sigma} + \sum_{\langle ij \rangle, \sigma, \gamma, \gamma'} (t_{ij}^{\gamma\gamma'} d_{i\gamma\sigma}^\dagger d_{j\gamma'\sigma} + \text{H.c.}) + H_{e-e} + \frac{1}{2} K \sum_{i,\gamma,\sigma,\sigma'} d_{i\gamma\sigma}^\dagger (\vec{\sigma})_{\sigma\sigma'} d_{i\gamma\sigma'} \cdot \vec{S}_i^{t_{2g}}. \quad (1)$$

Here it is assumed that the $2p$ orbitals of oxygen have been integrated over, and only the d orbitals of transition-metal ions are considered. The operator $d_{i\gamma\sigma}^\dagger$ creates an electron with spin $\sigma (= \uparrow, \downarrow)$ in the orbital $\gamma (= a, b)$ at site i . The transfer intensity $t_{ij}^{\gamma\gamma'}$ between the γ orbital in site i and the γ' orbital in the nearest-neighbor site j is calculated by considering the overlap integral between the Mn $3d$ and O $2p$ orbitals. We choose the up and down isospin state in the orbital space as $a = (3z^2 - r^2)$ and $b = (x^2 - y^2)$ orbitals, respectively. Then $t_{ij}^{\gamma\gamma'}$ is explicitly written by

$$t_{ij}^{\gamma\gamma'} = t_0 \begin{pmatrix} -\frac{1}{4}, & \frac{\sqrt{3}}{4} \\ \frac{\sqrt{3}}{4}, & -\frac{3}{4} \end{pmatrix}_{\gamma\gamma'}, \quad t_0 \begin{pmatrix} -\frac{1}{4}, & -\frac{\sqrt{3}}{4} \\ -\frac{\sqrt{3}}{4}, & -\frac{3}{4} \end{pmatrix}_{\gamma\gamma'}, \quad t_0 \begin{pmatrix} -1, & 0 \\ 0, & 0 \end{pmatrix}_{\gamma\gamma'}, \quad (2)$$

for $\vec{r}_j = \vec{r}_i \pm \hat{x}$, $\vec{r}_j = \vec{r}_i \pm \hat{y}$, and $\vec{r}_j = \vec{r}_i \pm \hat{z}$, respectively. The constant t_0 is positive, and depends on the distance between the d and p orbitals. The electron-electron interaction H_{e-e} is explicitly written by

$$H_{e-e} = U \sum_{i\gamma} n_{i\gamma\uparrow} n_{i\gamma\downarrow} + U' \sum_i n_{ia} n_{ib} + I \sum_{i,\sigma,\sigma'} d_{ia\sigma}^\dagger d_{ib\sigma'}^\dagger d_{ia\sigma'} d_{ib\sigma}, \quad (3)$$

where $n_{i\gamma\sigma} = d_{i\gamma\sigma}^\dagger d_{i\gamma\sigma}$ and $n_{i\gamma} = \sum_\sigma n_{i\gamma\sigma}$. The fourth-term in Eq. (1) is the Hund coupling between e_g and t_{2g} spins and $\vec{S}_i^{t_{2g}}$ is the t_{2g} spin with $S = 3/2$. The electron-electron interaction H_{e-e} in Eq. (3) is rewritten by using the spin operator for the e_g electron: $\vec{S} = \frac{1}{2} \sum_{\gamma\sigma\sigma'} d_{i\gamma\sigma}^\dagger (\vec{\sigma})_{\sigma\sigma'} d_{i\gamma\sigma'}$ and the isospin operator $\vec{T} = \frac{1}{2} \sum_{\gamma\gamma'\sigma} d_{i\gamma\sigma}^\dagger (\vec{\sigma})_{\gamma\gamma'} d_{i\gamma'\sigma}$, for the orbital degrees of freedom. First notice the identities

$$\vec{S}^2 = \frac{3}{4} n - \frac{3}{2} \sum_\gamma n_{\gamma\uparrow} n_{\gamma\downarrow} + 2 \vec{S}_a \cdot \vec{S}_b, \quad (4)$$

and

$$\vec{T}^2 = \frac{3}{4} n - 2 \vec{S}_a \cdot \vec{S}_b + n_a n_b + \frac{1}{2} \sum_\gamma n_{\gamma\uparrow} n_{\gamma\downarrow}, \quad (5)$$

where $n = n_a + n_b$. Then H_{e-e} is rewritten as

$$H_{e-e} = - \sum_i (\alpha^{(s)} \vec{S}_i^2 + \alpha^{(t)} \vec{T}_i^2 + \beta \vec{S}_{ia} \cdot \vec{S}_{ib}), \quad (6)$$

where $\alpha^{(s)} = \frac{2}{3}U + \frac{1}{3}U' - \frac{1}{6}I$, $\alpha^{(t)} = U' - \frac{1}{2}I$, and $\beta = -\frac{4}{3}(U - U' - I)$. Because (i) β is the order of I and is much smaller than $\alpha^{(s)}$ and $\alpha^{(t)}$, and (ii) the large U' forbids the simultaneous occupancy of a and b orbitals, we will neglect the $\vec{S}_{ia} \cdot \vec{S}_{ib}$ term in Eq. (6). Since $\alpha^{(s)}$ and $\alpha^{(t)}$ are both positive, the electron-electron interaction tends to induce the spin and isospin moments. By introducing two kinds of Stratonovich-Hubbard auxiliary fields, the partition function is given by

$$Z = \int \prod_i \left\{ \prod_{\sigma\gamma} [D \bar{d}_{i\sigma\gamma}(\tau) D d_{i\sigma\gamma}(\tau)] D \bar{\phi}_i^{(s)}(\tau) D \bar{\phi}_i^{(t)}(\tau) \right\} \times e^{-\int d\tau L}, \quad (7)$$

with

$$L = \sum_{i,\gamma,\sigma} \bar{d}_{i\gamma\sigma} (\partial_\tau + \epsilon_d) d_{i\gamma\sigma} + \sum_{\langle ij \rangle, \sigma, \gamma, \gamma'} (t_{ij}^{\gamma\gamma'} \bar{d}_{i\gamma\sigma} d_{j\gamma'\sigma} + \text{H.c.}) + K \sum_i \vec{S}_i \cdot \vec{S}_i^{t_{2g}} + L_{t_{2g}} - 2 \sum_i (\alpha^{(s)} \bar{\phi}_i^{(s)} \cdot \vec{S}_i + \alpha^{(t)} \bar{\phi}_i^{(t)} \cdot \vec{T}_i) + \sum_i (\alpha^{(s)} \bar{\phi}_i^{(s)2} + \alpha^{(t)} \bar{\phi}_i^{(t)2}), \quad (8)$$

where $L_{t_{2g}}$ is the Berry phase term of the t_{2g} spins. Equation (8) describes the e_g electrons moving in the background of two fluctuating fields $2\alpha^{(s)} \bar{\phi}^{(s)}$ and $2\alpha^{(t)} \bar{\phi}^{(t)}$.

We introduce at this stage the approximation which is appropriate for the strong correlated case. In this case the magnitude of the local fields $\alpha^{(s)} |\bar{\phi}^{(s)}|$ and $\alpha^{(t)} |\bar{\phi}^{(t)}|$ are much larger than the transfer intensity t_0 , and the electron is forced to be aligned in the directions of $\bar{\phi}^{(s)}$ and $\bar{\phi}^{(t)}$ at each site. Thus, it is convenient to rotate the spin and isospin axes

in each site, in order that its z axis coincides with $\bar{\phi}^{(s)}$ and $\bar{\phi}^{(t)}$. This is accomplished by the unitary matrices $U_i^{(s)}$ and $U_i^{(t)}$ for the spin and orbital spaces, respectively, which transform the fermion operator as $f_{i\gamma'\sigma'} = (U_i^{(s)})_{\sigma'\sigma} (U_i^{(t)})_{\gamma'\gamma} d_{i\gamma\sigma}$.¹⁷ In this rotating frame the fields $\bar{\phi}^{(s)}$ and $\bar{\phi}^{(t)}$ are pointing in the direction of $+z$, and accordingly the density of states for the f fermion are divided into four bands separated by the energy gaps. When the concentration of the e_g electrons is one per each transition-metal

ion, only the lowest band, which corresponds to $\sigma = \uparrow$ and $\gamma = a$, is occupied and the system becomes a Mott insulator. We keep only this lowest band, because the holes are doped to it and only this is important when the low-energy excitations are concerned. We introduce the spinless and orbital-less hole operator h_i as $h_i = f_{ia\uparrow}^\dagger$. The virtual transition processes to the higher bands cause the exchange interaction terms L_J , i.e., the so-called J term in the t - J type models.¹⁸ Then the effective Lagrangian up to the second order with respect to the electron transfer is obtained as follows:

$$L = \sum_i \bar{h}_i (\partial_\tau - \mu_h) h_i + \sum_i (1 - \bar{h}_i h_i) [(U_i^{(t)\dagger} \partial_\tau U_i^{(t)})_{aa} + (U_i^{(s)\dagger} \partial_\tau U_i^{(s)})_{\uparrow\uparrow}] + \sum_i 3(U_i^{(s)\dagger} \partial_\tau U_i^{(s)})_{\uparrow\uparrow} - \sum_{\langle ij \rangle} \bar{h}_i \tilde{t}_{ij} h_j + L_J, \quad (9)$$

where the chemical potential μ_h is determined by the condition $\langle h_i^\dagger h_i \rangle = x$. The term $3(U_i^{(s)\dagger} \partial_\tau U_i^{(s)})_{\uparrow\uparrow}$ comes from $L_{t_{2g}}$ in Eq. (8). The Berry phase terms of the original electron $d_{i\gamma\sigma}$ generates those for the rotated fermions h_i , spins, and isospins. Here we introduce the spinor boson $z_i^{(s)} = t[z_{i\uparrow}^{(s)}, z_{i\downarrow}^{(s)}]$ to represent the unitary matrix

$$U_i^{(s)} = \begin{pmatrix} z_{i\uparrow}^{(s)}, & -z_{i\downarrow}^{(s)*} \\ z_{i\downarrow}^{(s)}, & z_{i\uparrow}^{(s)*} \end{pmatrix}, \quad (10)$$

where $\vec{\phi}_i^{(s)}/|\vec{\phi}_i^{(s)}| = \sum_{\alpha\beta} z_{i\alpha}^{(s)*} (\vec{\sigma})_{\alpha\beta} z_{i\beta}^{(s)}$ and $\sum_\sigma |z_{i\sigma}^{(s)}|^2 = 1$. Correspondingly we introduce $z_i^{(t)}$ for $U_i^{(t)}$. $z_i^{(t)}$ ($l = s, t$) is written by using the angles in the spin and orbital spaces as $z_{i\uparrow}^{(l)} = \cos(\theta_i^{(l)}/2)$ and $z_{i\downarrow}^{(l)} = e^{i\phi} \sin(\theta_i^{(l)}/2)$. Then the Berry phase terms for spins are written as $(U_i^{(s)\dagger} \partial_\tau U_i^{(s)})_{\uparrow\uparrow} = \sum_\sigma z_{i\sigma}^{(s)*} \partial_\tau z_{i\sigma}^{(s)}$, and its coefficient should be regarded as the spin quantum number $2S$. Then in the undoped case ($x=0$), the quantum numbers of the spin and isospin are $S=2$ and $T=1/2$, respectively. It is expected that the quantum fluctuation is stronger for the isospin than the spin.

Now the original electron operator $d_{i\gamma\sigma}$ with the constraint of no double occupancy is expressed as $d_{i\gamma\sigma} = h_i^\dagger z_{i\gamma}^{(t)} z_{i\sigma}^{(s)}$, which is the generalization of the slave-fermion formalism to the orbital degenerate case. Then we name h_i , $z_{i\gamma}^{(t)}$, and $z_{i\sigma}^{(s)}$, holon, isospinon, and spinon, respectively. In this picture, the effective transfer intensity \tilde{t}_{ij} of the holon is given by

$$\tilde{t}_{ij} = \left(\sum_\sigma z_{i\sigma}^{(s)*} z_{j\sigma}^{(s)} \right) \left(\sum_{\gamma, \gamma'} z_{i\gamma}^{(t)*} t_{ij}^{\gamma\gamma'} z_{j\gamma'}^{(t)} \right). \quad (11)$$

It should be mentioned that the spin and orbital fluctuations cause the bond fluctuations, instead of the on-site fluctuations usually treated in the dynamical mean-field theory. In the ferromagnetic metallic states with the orbital degeneracy being lifted, the doped hole is identified as a spinless fermion. Therefore, it is reasonable to adopt the generalized slave-fermion method, where the doped hole is treated as a fermion, while the fluctuations in the spin and orbital degrees of freedom are treated as bosons. Also, we want to discuss the possibility of the disordering of the orbital degrees of

freedom. Then we employed a method which is rather good at describing the ordered state in order to avoid a bias toward the disordered state.^{19,20}

III. ORBITAL LIQUID STATES

In this section, we study nature of the orbital fluctuation in the ferromagnetic metallic states based on the effective action derived in the mean-field approximation. We show the energy dispersion for the orbital fluctuation and propose the orbital liquid state which has the significant implications to the experimental results.

We first consider the kinetic energy $-\sum_{\langle ij \rangle} \bar{h}_i \tilde{t}_{ij} h_j$ assuming that $xt_0 \gg J$ where J is the typical exchange energy and is of the order of $t_0^2/\alpha^{(s)}$ or $t_0^2/\alpha^{(t)}$. Then in the limit of strong correlation, we have the concentration region where $J/t_0 \ll x \ll 1$, which we are now interested in. The exchange terms L_J will be discussed later. We replace the fermion operators by their average value, i.e., $\langle \bar{h}_i h_i \rangle = x$, $\langle \bar{h}_i h_j \rangle = -x$, and quench the magnetic fluctuation by setting $z_{i\sigma}^{(s)} = t[1, 0]$. The constraint $\sum_\gamma |z_{i\gamma}^{(t)}|^2 = 1$ is imposed on average by introducing the chemical potential $-\lambda$. Then the effective Lagrangian of the orbital excitation $z_{i\gamma}^{(t)}$ is obtained as

$$L = \sum_{\vec{k}} xt_0 \begin{bmatrix} z_a^{(t)*}(\vec{k}) \\ z_b^{(t)*}(\vec{k}) \end{bmatrix} \times \begin{bmatrix} -\frac{1}{2}(c_x + c_y + 4c_z), & \frac{\sqrt{3}}{2}(-c_x + c_y) \\ \frac{\sqrt{3}}{2}(-c_x + c_y), & -\frac{3}{2}(c_x + c_y) \end{bmatrix} \begin{bmatrix} z_a^{(t)}(\vec{k}) \\ z_b^{(t)}(\vec{k}) \end{bmatrix} + \sum_{\gamma, \vec{k}} \lambda |z_\gamma^{(t)}(\vec{k})|^2, \quad (12)$$

with $c_x = \cos(ak_x)$, etc. The eigenvalues for each \vec{k} are given by $\epsilon_\pm^{(t)}(\vec{k}) = \lambda - xt_0 f_\pm(\vec{k})$ where

$$f_\pm(\vec{k}) = -(c_x + c_y + c_z) \pm [c_x^2 + c_y^2 + c_z^2 - c_x c_y - c_y c_z - c_z c_x]^{1/2}. \quad (13)$$

The minimum of this energy are given by the flat dispersion along the axis $(\pi/a, \pi/a, k_z)$ and the other two equivalent directions. This situation is quite in contrast to the spin case where the dispersion relation is proportional to $\cos(ak_x) + \cos(ak_y) + \cos(ak_z)$. The above feature is originated from the structure of $t_{ij}^{\gamma\gamma'}$ given in Eq. (2). In the eigenstates for the flat dispersion, $x^2 - y^2$ ($y^2 - z^2$, $z^2 - x^2$) orbitals are almost aligned in the two-dimensional layers. The electron in the $x^2 - y^2$ orbital cannot hop perpendicular to the xy plane because the overlap with the oxygen p orbital is vanishing due to the symmetry. [Only the (aa) component is nonzero in the third matrix in Eq. (2).]

This flat dispersion leads to an important consequence. The chemical potential λ is determined by $\langle \sum_\gamma z_{i\gamma}^{(t)*} z_{i\gamma}^{(t)} \rangle = (1/N) \sum_{\gamma, \vec{k}} n_B(\epsilon_\gamma^{(t)}(\vec{k})) = 1$, where n_B is the Bose distribution function. Because of this flat dispersion, the chemical potential $-\lambda$ is always negative at finite tem-

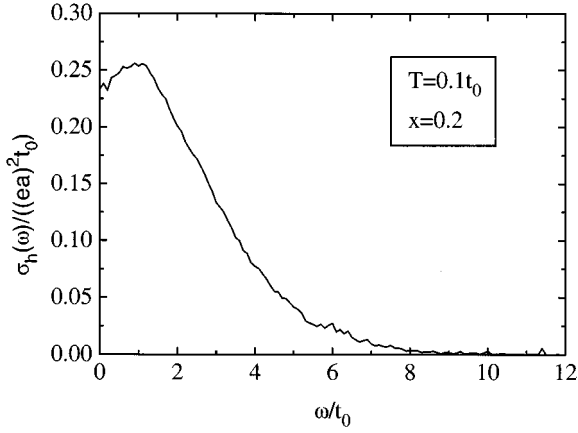


FIG. 1. The optical conductivity of the holon $\sigma_h(\omega)$ calculated in the $8 \times 8 \times 8$ system. The temperature and the hole concentration are chosen to be $T=0.1t_0$ and $x=0.2$, respectively.

perature although it becomes exponentially small below the effective Bose condensation temperature $T^* \sim xt_0$. Then the orbital long-range ordering does not occur down to low temperatures.

One may think that the physical quantities behave similarly to the orbital ordered state when the chemical potential is very small and $z^{(i)}$ is almost Bose condensed. However this is not the case because there are three branches of the low-lying fluctuations, i.e., along $(\pi/a, \pi/a, k_z)$ and two equivalent directions. In order to show it, we generate the random configuration of $z^{(i)}$ at very low temperature where the chemical potential is already very small. In this case only the static component of $z^{(i)}$ is important, which is obtained by the random numbers generated by the Gaussian function $\exp[-\epsilon_{\pm}^{(i)}(\vec{k})|z_{\pm}^{(i)}(\vec{k})|^2/T]$. Then \tilde{t}_{ij} is obtained through Eq. (11), and we diagonalize the holon Hamiltonian for that configuration of the transfer. We study the density of states and the optical conductivity $\sigma_h(\omega)$ of the holon system by averaging over 50 random samples for the cubic lattice of $8 \times 8 \times 8$. In Fig. 1, we present the results for the optical conductivity obtained by the numerical calculation. $\sigma_h(\omega)$ shows an incoherent peak, the width of which is the order of t_0 . If one looks at \tilde{t}_{ij} in each bond, it fluctuates in sign and magnitude violently, which explains the incoherent nature of $\sigma_h(\omega)$. This incoherent nature remains at lower temperature in comparison with the ferromagnetic transition temperature estimated as about xt_0 . Experimentally, the incoherent peak in $\sigma(\omega)$ observed in manganites has a broad band up to about 1 eV and it is growing with decreasing temperature in the ferromagnetic phase, where the spin degree of freedom is almost quenched. These results suggest the existence of the other degree of freedom which is still active down to the low temperature. Based on the present numerical calculation, we propose that this broad peak observed in $\sigma(\omega)$ is interpreted as the results of the fluctuation in the orbital degree of freedom. Also, we present numerical results for the density of states of the holon in Fig. 2. The density of states trails a skirt up to the region of $\omega \sim t_0$, and its global shape is far from the conventional one in the three-dimensional free-fermion system. In this hole concentration, the density of states at the Fermi level is fairly reduced due to the bond disorder. The photoemission spectra for the valence band

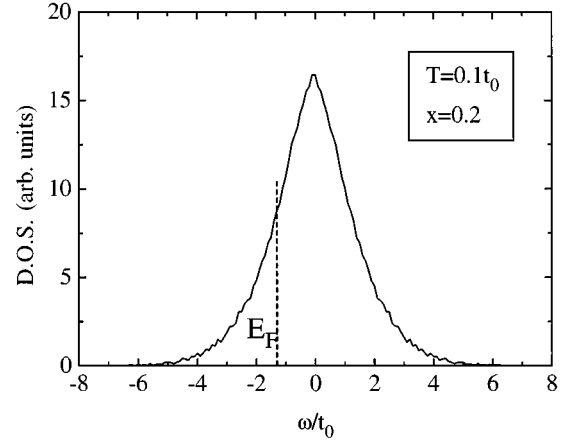


FIG. 2. The density of states of the holon $N_h(\omega)$ calculated in the $8 \times 8 \times 8$ system. The temperature and the hole concentration are chosen to be $T=0.1t_0$ and $x=0.2$, respectively. The broken line shows the Fermi level.

are, experimentally, measured in $\text{La}_{1-x}\text{Sr}_x\text{MnO}_3$, where the very low intensity near E_F and the small discontinuity at E_F are reported.^{5,12,13} These features are also consistent with the theoretical results attributed to the bond disordering in the orbital liquid states.

Here we discuss several possible effects which break the flat dispersion and/or make the orbitals long-range ordered. One possibility is the so-called J terms, i.e., L_J in Eq. (9). It is noted that the transfer matrix $t_{ij} = (t_{ij}^{\gamma\gamma'})$ does not commute with any of the Pauli matrices σ^α . Then there is no continuous rotational symmetry in the isospin space. Neglecting the terms of the order of xJ , L_J in the ferromagnetic phase has the same form as the first term in Eq. (12) with $z_a^{(i)}$ ($z_b^{(i)}$) being replaced by T_z (T_x) and xt_0 by $-t_0^2/(U'-I)$.¹⁸ Note that only T_z and T_x appear in L_J . The dispersion has the same form as $z^{(i)}$ -boson, i.e., $\epsilon_{J\pm}(\vec{k}) = -[t_0^2/(U'-I)]f_{\pm}(\vec{k})$. Then the minimum of the energy are given by the flat dispersion along the axis $(\pi/a, \pi/a, k_z)$ and the other two equivalent directions. For the lowest dispersion along $(\pi/a, \pi/a, k_z)$, $T_z=0$ and only T_x is nonzero, which competes with the fact that the kinetic energy prefers nonzero T_z . As before the ordering of the isospin is suppressed because there are three branches of the flat dispersion with the role of z 's being replaced by T 's. Applying the mean-field approximation to L_J , which contains the two-body interactions between the $z^{(i)}$ bosons, L_J does not generate a dispersion for the flat branch because flatness comes from the fact that the hopping of the x^2-y^2 electron along the z axis is forbidden. We also note that the orbital angular momentum is quenched in e_g orbitals, and the spin-orbit interaction is ineffective.

The orbital liquid, on the other hand, should be very sensitive to the anisotropy between the x , y , and z axis. In this respect, when the orbital degeneracy is lifted out in the layered materials, e.g., $\text{La}_{2-2x}\text{Sr}_{1+2x}\text{Mn}_2\text{O}_7$, the orbital liquid should be absent. Another test on our scenario is the effects of the uniaxial pressure. We expect that $\sigma(\omega)$ becomes anisotropic and becomes sharp in the more conductive plane.

IV. DISCUSSION AND CONCLUSIONS

In the previous section, we have studied the orbital fluctuation in the perovskite manganites where the two e_g orbitals are degenerate. Due to the strong electron-electron interactions, the system becomes a doped Mott insulator, and the quasi-two-dimensional nature of the orbital fluctuations prevents the orbital ordering. In this scenario, a characteristic energy of the orbital fluctuation (Ω) is given by the chemical potential of the isospinon, which is estimated as $\Omega \sim \lambda \sim \exp(-t_0 x/T)$, a very small energy scale at the low temperature. The picture of the static orbital disordering discussed in the previous section and the static approximation used in the numerical calculations are applicable only in the energy region larger than Ω . Therefore, the sharp Drude peak, experimentally observed in the actual compounds, does not appear in the numerical results presented in Fig. 1. Furthermore, the large entropy, expected from the orbital disordered states, is not consistent with the observed specific heat which is not enhanced at the low temperature. Interactions which have not been taken into account thus far can affect the nature of the orbital fluctuation and break the flatness of the dispersion. In this section, as the most probable candidate, we introduce the interaction between the orbital and Jahn-Teller (JT) distortion, which brings about a new energy scale for Ω in place of λ .

We consider the Hamiltonian,²¹ describing the coupled system between the orbital and lattice degrees of freedom, as follows:

$$H = H_{\text{ele}} + H_{\text{latt}} + H_{\text{jt}} + H_{\text{nl}}, \quad (14)$$

where H_{ele} is the Hamiltonian for the pure electronic system given in Eq. (1). We introduce the two kinds of the E_g lattice distortion modes described by Q_{il} ($l=2,3$), which are represented by the amplitude of the JT distortion r_i and its phase θ_i in the $(Q_2 - Q_3)$ plane as $Q_{i2} = r_i \sin \theta_i$ and $Q_{i3} = r_i \cos \theta_i$. H_{latt} includes the kinetic energy and the harmonic potential for these modes with the spring constant K . The third term H_{jt} represents the linear JT coupling given by

$$H_{\text{jt}} = -A \sum_i (T_{iz} Q_{i3} + T_{ix} Q_{i2}). \quad (15)$$

The last term H_{nl} is divided into the two parts: the anharmonic lattice potential and the nonlinear JT coupling as follows:

$$H_{\text{nl}} = \sum_i \{B_1(Q_{i3}^3 - 3Q_{i2}^2 Q_{i3}) + B_2[(Q_{i3}^2 - Q_{i2}^2)T_{iz} + 2Q_{i2} Q_{i3} T_{ix}]\}. \quad (16)$$

It is noted that the bandwidth of the holon is the order of xt_0 , not the order of t_0 , due to the strong electron-electron interaction. Therefore, in comparison with the noninteracting system, the JT distortion brings out the more efficacious coupling with the mobile carrier in the present case. Although magnitudes of the several energy parameters in the actual compounds are not clarified up to now, we assume, for simplicity, the strong-coupling limit where the condition $[A(A/K) \gg xt_0, B_1(A/K)^3, B_2(A/K)^2]$ is satisfied. In this limit, the phase degrees of freedom only remain and the

stable values of θ are mainly determined by H_{nl} and the kinetic term in H_{ele} . By using the phase variable, H_{nl} is rewritten as follows:

$$H_{\text{nl}} = \sum_i \cos(3\theta_i) [B_1 r_i^3 + B_2 r_i^2 (1 - h_i^\dagger h_i)]. \quad (17)$$

Because of the negative signs of B_1 and B_2 in manganites,^{8,9} this term becomes minimum at $\theta=0, \pm 2\pi/3$, that is, the $3z^2 - r^2(3y^2 - r^2, 3x^2 - r^2)$ -type orbital is in favor in H_{nl} . On the other hand, as previously pointed out, the kinetic term in H_{ele} stabilizes the $x^2 - y^2(y^2 - z^2, z^2 - x^2)$ -type orbital. With assuming the uniform value of θ , H_{ele} is rewritten as follows:

$$H_i = t_0 x^{5/3} \left\{ [1 - \cos(\theta + \pi)] \left[1 - \cos\left(\theta + \frac{\pi}{3}\right) \right] \times \left[1 - \cos\left(\theta - \frac{\pi}{3}\right) \right] \right\}^{1/3} + O(x^{7/3}), \quad (18)$$

which becomes its minimum at $\theta = \pi, \pm \pi/3$. It is noted that the potential minima introduced by the above two terms alternate in the $(Q_2 - Q_3)$ plane. With increasing the hole concentration, magnitudes of xt_0 , $B_1(A/K)^3$, and $B_2(A/K)^2$ become comparable with each other and the potential heights become shallow. Then, the quantum tunneling between the triple or sixfold potential well are expected to easily occur and govern the orbital fluctuations. The tunneling frequency gives the characteristic energy scale of the orbital fluctuation Ω , in place of λ . In this case, the effects of the dissipation on the quantum tunneling seems to be irrelevant with considering the results of the renormalization-group study in the double-well case.²² The entropy at the low temperature is expected to be quenched, because the degeneracy is lifted on every site where the singlet state is formed by the orbital-lattice coupling.

Let us consider the motion of the holon interacting with the system discussed above. When the energy of the holon ω is larger than Ω , the holon feels the static random potential at each bond due to the orbital disordering. On the other hand, in the case of $\omega < \Omega$, the holon feels the averaged potential during a lot of tunneling processes. As a result, the optical conductivity of the holon has the structure around $\omega \sim \Omega$ and shows the two-component feature. The numerical result presented in Fig. 1 corresponds to the case of $\Omega \rightarrow 0$ in the present picture. In order to demonstrate the role of the finite value of Ω , we introduce the following effective model and calculate the optical conductivity based on it. We consider the double-well potential for the lattice at each site, instead of the triple or sixfold well in the original model. Between the potential minima, the quantum tunneling occurs with the frequency Ω . The transfer intensity of the holon between the nearest-neighbor sites depends on the states of the lattice. The effective model are given as follows:

$$H = \sum_{\langle ij \rangle} (t_0 + t_1 \sigma_i^z \sigma_j^z) h_i^\dagger h_j + \text{H.c.} + \Omega \sum_i \sigma_i^x, \quad (19)$$

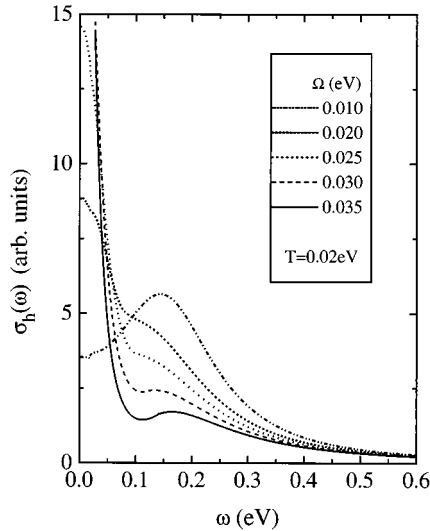


FIG. 3. The optical conductivity of the holon $\sigma_h(\omega)$ calculated by the model in Eq. (19). Ω is the tunneling frequency between the double well. Temperature is chosen to be $T=0.02$ eV.

where σ_i^l are the Pauli matrices describing the double-well potential. In Fig. 3, we present the optical conductivity spectra calculated by the memory function formalism²³ up to the order of t_1^2 . It is noted again that the numerical result in Fig. 1 corresponds to the case of $\Omega \rightarrow 0$ in the present scheme. We find the two-component feature in $\sigma_h(\omega)$, that is, the sharp Drude peak and the broad incoherent component down to $\omega \sim 4 \Omega$. With decreasing Ω , the fine structure and the sharp Drude peak are thermally smeared and the weight is transferred to the incoherent band. If we assume $\Omega \sim 0.02 - 0.03$ eV, the two-component feature in the calculated spectra are qualitatively consistent with the experimental ones in manganites, except for the small Drude weight which is still an open question.

In summary we have shown that there are reasons to expect that the orbital degrees of freedom remain disordered in the low-temperature ferromagnetic metallic state even with the well developed local orbital moments due to the strong electron-electron interaction. These are (1) the anisotropic character of the transfer integral leads to the quasi-two-dimensional dispersion for the isospinon. (2) When the JT effect is considered, there occurs competition between the energy gain in the kinetic energy and the nonlinear potential for the distortion. This competition enhances the quantum tunneling of the JT distortion and hence the vibronic state is expected to survive. This introduces the characteristic energy

scale, i.e., the tunneling frequency, which explains qualitatively the two-component feature of the optical conductivity.

Then the next question is what are the low-energy states? If the mean-field theory in the slave-fermion formalism given above is correct, it means that the electrons are in the non-Fermi-liquid state. However there is not enough experimental and theoretical evidence to believe either the non-Fermi liquid or Fermi liquid. The main purpose of this work is to study the possibility of the ordering and/or disordering of the orbital degrees of freedom. Because we obtain the disordered state within the formalism which favors the ordering, we expect this result is convincing. However knowing that the orbital degrees of freedom remain disordered, the slave-boson formalism is another candidate to describe the low-energy states. In this formalism the slave bosons are expected to be condensed easily in three dimensions, and the low-energy state is the usual Fermi liquid with the two bands crossing the Fermi energy. Actually Shiba *et al.*²⁴ took the Fermi-liquid viewpoint and calculated the optical conductivity for the noninteracting electrons. In this viewpoint the ‘‘Drude part’’ in the optical conductivity corresponds to the intraband transitions, while the ‘‘incoherent part’’ to the interband transitions. However we believe that our slave-fermion theory is applicable at least to time scales shorter than the characteristic one for the orbital fluctuations, because the electronic system feels the orbital order within that time scale. Furthermore the slave-fermion mean-field theory describes the small Drude weight without the heavy mass, because the carrier density is small (x) in this formalism. On the other hand, in the slave-boson mean-field theory with the bose condensation, the small Drude weight suggests large specific heat, which is not consistent with the experiment. In any case the fluctuations from the mean-field theory in both the slave-fermion and slave-boson formalisms need to be studied in the future.

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