Spin and orbital excitations in undoped manganites

R. Y. Gu
National Laboratory of Solid State Microstructures, Nanjing University, Nanjing 210093, People's Republic of China and Department of Physics, The University of Hong Kong, Pokfulam, Hong Kong, People's Republic of China

Shun-Qing Shen and Z. D. Wang[a)]
Department of Physics, The University of Hong Kong, Pokfulam, Hong Kong, People's Republic of China

D. Y. Xing
National Laboratory of Solid State Microstructures, Nanjing University, Nanjing 210093, People's Republic of China and National Center for Theoretical Sciences, P.O. Box 2-131, Taiwan 300

(Received 20 March 2000; accepted for publication 11 August 2000)

We develop a theory for spin and orbital excitations in undoped manganites to account for the spin and orbital orderings observed experimentally. It is found that the anisotropy of the magnetic structure is closely related to the orbital ordering, and the Jahn-Teller effect stabilizes the orbital ordering. The phase diagram and the low-energy excitation spectra for both spin and orbital orderings are obtained. The calculated critical temperatures can be quantitatively comparable to the experimental data. © *2000 American Institute of Physics.* [S0021-8979(00)04622-3]

I. INTRODUCTION

$LaMnO_3$ is the parent compound of colossal magnetoresistance manganites, and has been studied both experimentally and theoretically. The compound is an insulator with layered antiferromagnetic (A-type AF) spin ordering and an orbital ordering of e_g electrons, accompanied by a Jahn-Teller (JT) lattice distortion.[1] Murakami *et al.*[2] have recently succeeded in detecting the orbital ordering in $LaMnO_3$ by using resonant x-ray scattering techniques with the incident photon energy tuned near the Mn K-absorption edge. The orbital order parameter decreases above the Neel temperature $T_N \sim 140$ K and persists until $T_O \sim 780$ K. Theoretically, the problem of orbital degeneracy in a *d*-electron system was pioneered by Kugel and Khomskii[3] in the 1970's and investigated extensively in recent years.[4–12] Meanwhile, the lattice distortion, from the band structure calculation, is believed to play an important role in the undoped manganites.[13,14]

The electron configuration of Mn^{3+} in $LaMnO_3$ is $t_{2g}^3 e_g^1$. The three t_{2g} electrons are localized, while the twofold degenerate orbitals of the e_g electron are strongly hybridized, with its spin aligning parallelly to that of the t_{2g} electrons due to the Hund's coupling. The strong on-site Coulomb interaction prohibits the double occupancy of e_g electrons at one site, and leads to an effective low-energy Hamiltonian in which the spin and orbital degrees of freedom are interrelated. In this article, starting from the effective Hamiltonian of the spin and orbital interactions, as well as the JT coupling between the e_g electrons and the lattice distortion, we investigate the interplay among the spin, orbit and the lattice distortion. We present the phase diagram as functions of interaction parameters, and obtain the low-energy excitations of the system in different phases. It is found that special properties of the orbital operators can result in an anisotropy of the magnetic structure and an energy gap of the orbital excitations. We also estimate the critical temperatures for spin and orbital orderings as well as their dependence on the JT coupling. The calculated results are comparable to the experimental measurements.

II. EFFECTIVE HAMILTONIAN

The effective spin and orbital interactions are derived by the projection perturbation method up to the second order[6,11,12]

$$H_e^{\mathrm{eff}} = J_1 \sum_{ij} (\mathbf{S}_i \cdot \mathbf{S}_j - S^2) n_i^\alpha n_j^\alpha + J_2 \sum_{ij} (\mathbf{S}_i \cdot \mathbf{S}_j - S^2) n_i^\alpha n_j^{\bar{\alpha}} - J_3 \sum_{ij} [\mathbf{S}_i \cdot \mathbf{S}_j + S(S+1)] n_i^\alpha n_j^{\bar{\alpha}}, \quad (1)$$

where $\mathbf{S}_i$ is the spin operator of $S=2$. The three terms describe three processes with different intermediate states, with $J_1 = t^2/[2(U+3J_H/2)S^2]$, $J_2 = t^2/[(U'+3J/2+3J_H/2)S(2S+1)]$, and $J_3 = t^2/[(U'-J/2)S(2S+1)]$.[11] Here t is the hopping integral, U (U') is the intra- (inter-) orbital Coulomb interaction, J and J_H are the Hund's coupling between the e_g electrons and between the e_g and the t_{2g} electrons, respectively. The terms $n_i^\alpha = d_{i\alpha}^\dagger d_{i\alpha}$ and $n_i^{\bar{\alpha}} = d_{i\bar{\alpha}}^\dagger d_{i\bar{\alpha}}$ ($\alpha = x,y,z$) are the particle number operators of e_g electron in orbit states $|\alpha\rangle = \cos(\varphi_\alpha/2)|z\rangle + \sin(\varphi_\alpha/2)|\bar{z}\rangle$ and $|\bar{\alpha}\rangle = -\sin(\varphi_\alpha/2)|z\rangle + \cos(\varphi_\alpha/2)|\bar{z}\rangle$, respectively, with orbital states $|z\rangle \propto (3z^2-r^2)/\sqrt{3}$ and $|\bar{z}\rangle \propto x^2-y^2$. The term φ_α depends on the direction of the (ij) bond by $\varphi_x = -2\pi/3$, $\varphi_y = 2\pi/3$, and $\varphi_z = 0$, respectively, for bond (ij) parallel to the x, y and z directions. The introduced $d_{i\alpha}^\dagger, d_{i\alpha}$ and $d_{i\bar{\alpha}}^\dagger, d_{i\bar{\alpha}}$

[a)]Author to whom correspondence should be addressed: electronic mail: zwang@hkucc.hku.hk

are operators in the orbital space, with $d_{i\alpha}^{\dagger}|0\rangle=|\alpha\rangle$, $d_{i\bar{\alpha}}^{\dagger}|0\rangle=|\bar{\alpha}\rangle$, they should satisfy the constraint $n_i^{\alpha}+n_i^{\bar{\alpha}}=1$.

The JT interaction may be expressed as[15]

$$H_{\rm JT}=-g\sum_{i\gamma\gamma'} d_{i\gamma}^{\dagger}\mathbf{T}_{\gamma\gamma'}\cdot\mathbf{Q}_i d_{i\gamma'}+\frac{K}{2}\sum_i \mathbf{Q}_i^2, \tag{2}$$

where $\mathbf{T}=(T_x,T_z)$ are the Pauli matrices in the orbital space with $\gamma\ (\gamma')=z$ or $\bar{z}$, and g is the coupling between the e_g electrons and the local JT lattice distortion $\mathbf{Q}_i=Q_i(\sin\phi_i,\cos\phi_i)$. In principle, JT distortions are global as they physically correspond to movement of share oxygen atoms. Here to simplify our calculation we have adopted an independent local distortion approximation. To take partly the effect of collective nature of JT distortions into account, as well as that of the local anharmonic oscillation and the higher order coupling, a preferable direction ϕ_i of the JT distortion at each site will be selected according to the experimental observation, which implies that the lattice distortions at different sites are not really independent. It seems that the main effect of the coupled distortions renormalized the model parameters in Eq. (2).

Experimental measurement on $LaMnO_3$ indicates that the critical temperature of the orbital ordering, T_O, is much higher than that of the magnetic ordering, T_N.[2] As a result, the spin and orbital degrees of freedom, which are coupled to each other in Hamiltonian (1), may be separately treated by the Hartree-Fock mean-field approach. The total Hamiltonian is reduced to $H_{\rm MF}=H_S+H_O+E_0$, where H_S and H_O are the decoupled spin and orbital Hamiltonians, respectively, and E_0 is an energy constant. The spin Hamiltonian H_S is given by

$$H_S=\sum_{\langle ij\rangle}\tilde{J}_{ij}\mathbf{S}_i\cdot\mathbf{S}_j \tag{3}$$

with the effective spin coupling depending on the orbital configuration of the two neighboring sites by

$$\tilde{J}_{ij}=\tfrac{1}{2}J_1\langle(1+m_i^{\alpha})(1+m_j^{\alpha})\rangle + \tfrac{1}{2}(J_2-J_3)\langle 1-m_i^{\alpha}m_j^{\alpha}\rangle+\tilde{J}_{\rm AF}, \tag{4}$$

where $m_i^{\alpha}=n_i^{\alpha}-n_i^{\bar{\alpha}}$ are the orbital operators, and the $\tilde{J}_{\rm AF}$ term comes from the magnetic superexchange between the nearest neighboring local spins. It is worth pointing out here that the orbital operators introduced above have unusual operator algebra, being quite different from that of the spin operators. It can be shown that they satisfy the following relations:

$$(m_i^{\alpha})^2=1; \tag{5a}$$

$$m_i^x+m_i^y+m_i^z=0; \tag{5b}$$

$$[m_i^x,m_i^y]=[m_i^y,m_i^z]=[m_i^z,m_i^x]=\sqrt{3}(d_{i\bar{z}}^{\dagger}d_{iz}-d_{iz}^{\dagger}d_{i\bar{z}}). \tag{5c}$$

The orbital Hamiltonian H_O can be written as

$$H_O=\sum_{\langle ij\rangle}u_{ij}m_i^{\alpha}m_j^{\alpha}-\sum_{\langle ij\rangle}h_{ij}m_i^{\alpha}+\frac{K}{2}\sum_i Q_i^2 -g\sum_i Q_i\left(m_i^z\cos\phi_i+\frac{1}{\sqrt{3}}(m_i^y-m_i^x)\sin\phi_i\right), \tag{6}$$

where the effective orbital coupling u_{ij} depends on the spin configuration of the two neighboring sites by

$$u_{ij}=\tfrac{1}{2}(J_1-J_2+J_3)\langle\mathbf{S}_i\cdot\mathbf{S}_j\rangle+\tfrac{1}{2}(J_3-J_1+J_2)S^2+\tfrac{1}{2}J_3S$$

and $h_{ij}=-1/2J_1\langle\mathbf{S}_i\cdot\mathbf{S}_j-S^2\rangle$. All these coupling parameters $\tilde{J}_{ij}$, u_{ij} and h_{ij} in H_S and H_O are determined not only by the spin and orbital configurations of the nearest neighboring sites i and j, but also by the direction of the (ij) bond. For short, we denote them by $\tilde{J}_{\alpha}$, u_{α} and h_{α} thereafter. If there are two symmetric directions in the system, e.g., x and y direction, one has $\tilde{J}_x=\tilde{J}_y$, $u_x=u_y$, and $h_x=h_y$.

III. SPIN AND ORBITAL EXCITATIONS

The spin Hamiltonian H_S is an anisotropic Heisenberg Hamiltonian with SU(2) symmetry. At low temperatures, the spin configuration along the α direction is determined by the sign of J_{α}. Dividing the system into two sublattices A and B according to their spin alignments, and performing the well-known Holsten-Primakoff (HP) transformation in the linear spin wave theory, up to the quadratic terms, we diagonalize H_S as

$$H_S=\sum_{\mathbf{k}}\left[\omega_{\mathbf{k}}(\psi_{\mathbf{k}}^{\dagger}\psi_{\mathbf{k}}+\chi_{\mathbf{k}}^{\dagger}\chi_{\mathbf{k}}+1)-2S(S+1)W\right]. \tag{7}$$

Here $\psi_{\mathbf{k}}$ and $\chi_{\mathbf{k}}$ are the quasiparticle operators of the spin wave excitations with $\mathbf{k}$ the wave vectors of one sublattice. The quasiparticle spectrum is given by $\omega_{\mathbf{k}}=\sqrt{(W+P_{\mathbf{k}}^-)^2-(P_{\mathbf{k}}^+)^2}$, with $P_{\mathbf{k}}^{\mp}=2S\Sigma_{\alpha}\tilde{J}_{\alpha}\cos k_{\alpha}\Theta(\mp\tilde{J}_{\alpha})$, and $W=2S\Sigma_{\alpha}|\tilde{J}_{\alpha}|$, in which Θ is the unit step function.

From the obtained spin-wave spectrum, the magnitude of the average spin per site in one sublattice at low temperatures is

$$\langle S_{\rm sub}\rangle=S-\frac{1}{2}\int\frac{d^3k}{(2\pi)^3}\left(\frac{J_1+P_{\mathbf{k}}^-}{\omega_{\mathbf{k}}}\coth\frac{\beta\omega_{\mathbf{k}}}{2}-1\right), \tag{8}$$

where β is the inversed temperature and N is the number of sites in one sublattice.

The orbital Hamiltonian H_O looks quite like H_S, where the orbital operator may be regarded as an isospin operator. But the absence of the SU(2) symmetry in H_O and the abnormal algebra of orbital operators Eqs. (5a)–(5c) make the orbital operators quite different from the spin operators. For example, orbital F-type arrangement is not an eigenstate of H_O, and in the case of orbital AF configuration, on orbital sublattice $\bar{A}$ or $\bar{B}$ there are only several preferable orbital alignments at which the ground-state energy of the system reaches its minimum, unlike in an AF spin system where all the spin orientations on a sublattice are energy degenerate. In this case, the orbital state at site i can be generally expressed as $|i\rangle=\cos(\theta_{\sigma}/2)|z\rangle+\sin(\theta_{\sigma}/2)|\bar{z}\rangle$ with $\sigma=+$ for $i\in\bar{A}$ and

$-$ for $\overline{B}$, respectively. From the symmetry of $h_x = h_y$ and relation $m_{ix}+m_{iy}+m_{iz}=0$, the second term on the right-hand side of Eq. (6) can be rewritten in a more intuitive form

$$H_z = -\epsilon_z \sum_i m_i^z \tag{9}$$

with $\epsilon_z = h_z - h_x$. This anisotropic Hamiltonian arises from anisotropy of electronic hopping integrals in orbital space as well as unusual algebra of orbital operators. Both u_α and h_α are anisotropic and depend on the spin configurations along the α direction, as shown in their expressions below Eq. (6). Since $J_1 - J_2 + J_3$ is always positive, we have $u_z < u_x$ and $\epsilon_z > 0$ for the A-type AF spin configuration; $u_z > u_x$ and $\epsilon_z < 0$ for the C-type AF one; and $u_z = u_x$ and $\epsilon_z = 0$ for the ferromagnetic (F) one. The static JT distortions $\mathbf{Q}_i$ are approximately treated as classical variables and assumed to be different in the two sublattices, i.e., $Q_i \equiv Q_\sigma$ and $\phi_i \equiv \phi_\sigma$ with $\sigma = +\ (-)$ for $i \in \overline{A}\ (\overline{B})$. From x-ray diffraction experiments, it has been confirmed that the MnO_6 octahedron is elongated along the *x* or *y* direction, and the octahedrons are alternatively aligned in the *x*-*y* plane,[16] which corresponds to $\phi_+ = 2\pi/3$ and $\phi_- = -2\pi/3$ in the present formula. Similar to the treatment of the spin degrees of freedom, we perform the HP transformation for localized orbital operators by replacing[8]

$$\begin{aligned} &d_{i\downarrow}^{\sigma\dagger} d_{i\uparrow}^{\sigma} \to a_{i\sigma}^{\dagger}(1 - a_{i\sigma}^{\dagger} a_{i\sigma})^{1/2}, \\ &d_{i\uparrow}^{\sigma\dagger} d_{i\downarrow}^{\sigma} \to (1 - a_{i\sigma}^{\dagger} a_{i\sigma})^{1/2} a_{i\sigma}, \\ &d_{i\downarrow}^{\sigma\dagger} d_{i\downarrow}^{\sigma} \to a_{i\sigma}^{\dagger} a_{i\sigma}, \quad d_{i\uparrow}^{\sigma\dagger} d_{i\uparrow}^{\sigma} \to 1 - a_{i\sigma}^{\dagger} a_{i\sigma}, \end{aligned} \tag{10}$$

where $d_{i\uparrow}^{\sigma} = \cos(\theta_\sigma/2) d_{iz} + \sin(\theta_\sigma/2) d_{i\bar{z}}$, $d_{i\downarrow}^{\sigma} = -\sin(\theta_\sigma/2) d_{iz} + \cos(\theta_\sigma/2) d_{i\bar{z}}$. To the lowest order, H_O can be diagonalized as

$$H_O = \sum_{\mathbf{k}\sigma} \epsilon_{\mathbf{k}\sigma} \xi_{\mathbf{k}\sigma}^{\dagger} \xi_{\mathbf{k}\sigma} + \frac{1}{2} \sum_{\mathbf{k}\sigma} (\epsilon_{\mathbf{k}\sigma} - P_\sigma) + E_C. \tag{11}$$

Here $\xi_{\mathbf{k}\sigma}^{\dagger}$ and $\xi_{\mathbf{k}\sigma}$ are the quasiparticle operators of the orbital excitations, the second term stands for the quantum fluctuation energy, where

$$\begin{aligned} P_\sigma = &-\sum_\alpha 4u_\alpha \cos\theta_+^\alpha \cos\theta_-^\alpha + 2\epsilon_z \cos\theta_\sigma \\ &+ \frac{2g^2}{K} \cos^2(\theta_\sigma - \phi_\sigma) \end{aligned}$$

with $\theta_\sigma^\alpha = \theta_\sigma - \varphi_\alpha$, and E_C is the classical ground-state energy. The expression for E_C depends on the orbital configuration. For both G- and C-type AF configurations, it is given by

$$\begin{aligned} E_C/N = &\sum_\alpha u_\alpha \cos\theta_+^\alpha \cos\theta_-^\alpha \\ &- \frac{1}{2} \sum_\sigma \left[\epsilon_z \cos\theta_\sigma + g Q_\sigma \cos(\theta_\sigma - \phi_\sigma) - \frac{K}{2} Q_\sigma^2 \right] \end{aligned} \tag{12}$$

with *N* the number of the sites. In principle, θ_σ and Q_σ in Eq. (11) should be determined by minimizing the total ground- state energy of the system. In the present case, the quantum fluctuations in H_S and H_O are small, and so the ground-state energy can be approximately replaced by E_C. It is found that besides the same ground-state energy E_C, there is the same excitation spectrum for the C- and G-type AF orbital configurations, yielding

$$\epsilon_{\mathbf{k}\sigma} = \sqrt{\tfrac{1}{2}\{P_+^2 + P_-^2 + \sigma[(P_+^2 - P_-^2)^2 + 16 P_+ P_- C_{\mathbf{k}}^2]^{1/2}\}}, \tag{13}$$

where $C_{\mathbf{k}} = \Sigma_\alpha 2u_\alpha \sin\theta_+^\alpha \sin\theta_-^\alpha \cos k_\alpha$. This degeneracy of C- and G-type AF orbital configurations agrees with Mizokawa and Fujimori's result.[17] Independent of the magnetic structure, such a degeneracy suggests the possibility of a mixed C- and G-type AF orbital configuration in the system, i.e., neighboring orbital states along the *z* direction may be either ''parallel'' or ''antiparallel.'' In the absence of the Coulomb interactions,[9] a C-type AF orbital structure may have lower energy. The energy of the C-type AF orbital ordering can also be lowered by including an additional hopping term which might be from the tilting of the MnO_6 octahedron.[18]

The JT coupling plays an important role in determining the orbital ordering. In the absence of the JT coupling and in the small limit of ϵ_z, the e_g electrons may occupy two antiparallel states in the two sublattices: $(|z\rangle \pm |\bar{z}\rangle)/\sqrt{2}$ ($\theta_+ = -\theta_- = \pi/2$) for $u_z < u_x$; $|z\rangle$ and $|\bar{z}\rangle$ ($\theta_+ = 0, \theta_- = -\pi$) for $u_z > u_x$. Such symmetric antiparallel states will be broken by the uniform crystal field appearing in Eq. (9). Furthermore, the JT distortions also lead to an effective anisotropic crystal field acting on the two sublattices. To distinguish it from the uniform crystal field ϵ_z, we call it the JT field. The JT field, whose strength increases with the coupling constant *g*, tends to align the orbital states in the two sublattices towards $|y\rangle$ ($\theta_+ = 2\pi/3$) and $|x\rangle$ ($\theta_- = -2\pi/3$), respectively.

The orbital ordering is described by the average value of operators m_i^α. From the orbital spectrum, it can be shown that

$$\langle m_\sigma^\alpha \rangle = M_\sigma \cos\theta_\sigma^\alpha \tag{14}$$

with $\sigma = +\ (-)$ for $i \in \overline{A}\ (\overline{B})$, where

$$\begin{aligned} M_\sigma = 1 - &\sum_{\sigma'} \int \frac{d^3k}{(2\pi)^3} \frac{2P_{\bar{\sigma}} C_{\mathbf{k}}^2}{\epsilon_{\mathbf{k}\sigma'}[4P_+ P_- C_{\mathbf{k}}^2 + (P_{\sigma}^2 - \epsilon_{\mathbf{k}\sigma'}^2)^2]} \\ &\times \left(\frac{2(P_\sigma^2 + \epsilon_{\mathbf{k}\sigma'}^2)}{e^{\beta\epsilon_{\mathbf{k}\sigma'}} - 1} + (P_\sigma - \epsilon_{\mathbf{k}\sigma'})^2 \right). \end{aligned} \tag{15}$$

The second term on the right-hand side of Eq. (15) comes from the quantum and thermal fluctuations. To keep a good approximation, this term must be small at low temperatures.

IV. RESULTS AND DISCUSSIONS

We now discuss the ground state of the system. First, it is impossible to realize an isotropic orbital ordering. Since $m_i^x + m_i^y + m_i^z = 0$, if $\langle m_i^x \rangle = \langle m_i^y \rangle = \langle m_i^z \rangle$, they must be equal to zero and there is not any orbital ordering. From Eq. (4), it then follows that the anisotropy in $\langle m_i^\alpha \rangle$ will lead to aniso-

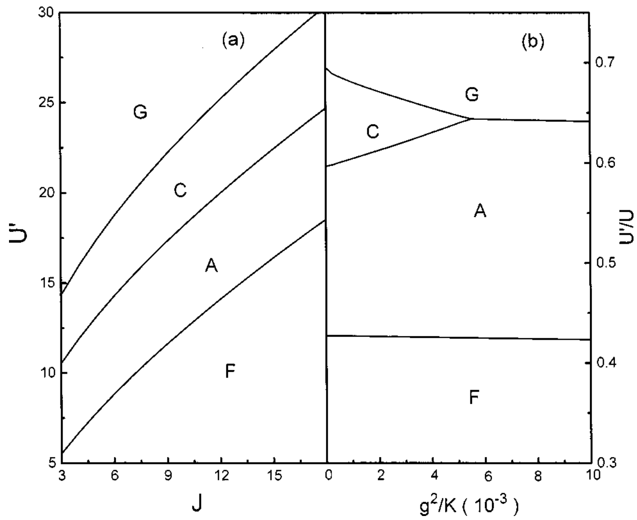

FIG. 1. Phase diagram at zero temperature in the absence (a) and presence (b) of the JT field. The parameters used are $\tilde{J}_{\rm AF}=0.006$ and $J_H=4/3$ with t the unit of energy. The relation $U=U'+2J$ has been used (see Ref. 21) and $U=20$ fixed in (b).

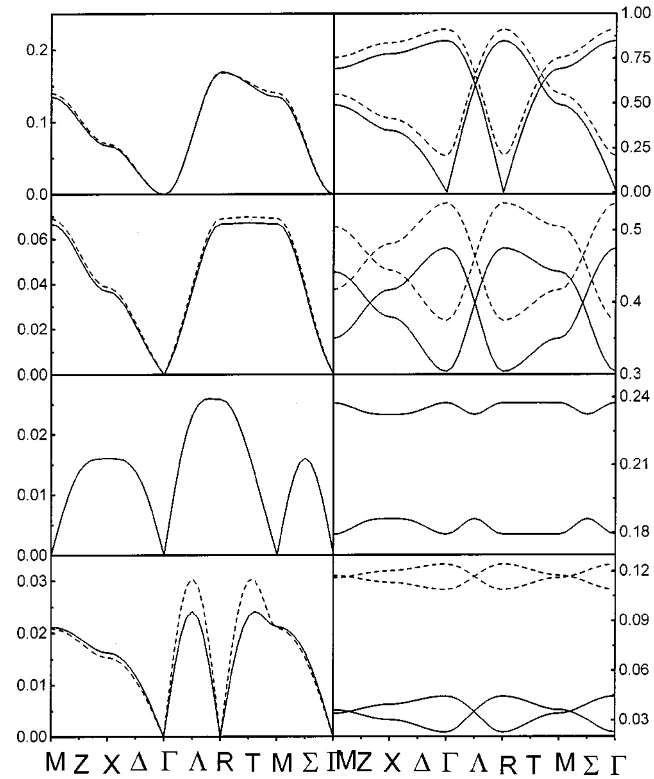

FIG. 2. Spin and orbital excitation spectra shown in the left and right columns, respectively, with $g=0$ (solid lines) and $g^2/K=0.04$ (dashed lines). Here $U'=8$, 10, 12 and 14 from top to bottom, corresponding to the F and A-, C-, G-type AF spin configurations. The other parameters are the same as those in Fig. 1(b).

tropic $\tilde{J}_\alpha$. At zero temperature, $M_\sigma=1$ and $\langle m_\sigma^\alpha\rangle=\cos\theta_\sigma^\alpha$ if the quantum fluctuation in Eq. (14) is neglected. Taking into account the symmetry requirement of $\langle m_i^x+m_j^x\rangle=\langle m_i^y+m_j^y\rangle$, we get two possible relations: (I) $\theta_+ + \theta_- = 0$ or (II) $\theta_+ - \theta_- = \pi$. As the quantum fluctuation is taken into account, relation (I) remains unchanged, while relation (II) is satisfied only approximately. In both cases, we have $\tilde{J}_x=\tilde{J}_y \neq \tilde{J}_z$ from Eq. (4), provided the small quantum fluctuations are neglected. Since the magnetic structure at zero temperature is determined by the sign of $\tilde{J}_\alpha$, the same sign of $\tilde{J}_\alpha$, regardless of anisotropic magnitude of them, will lead to an F or G-type AF spin configuration, while different signs of $\tilde{J}_x$ and $\tilde{J}_z$ will result in an A- or C-type spin configuration. Our calculations show that the ground-state magnetic structure is very sensitive to the on-site Coulomb interactions. Even though the magnetic superexchange $\tilde{J}_{\rm AF}$ is fixed and the JT coupling is absent $(g=0)$, an evolution of spin configuration in the order of $F\rightarrow A\rightarrow C\rightarrow G$ can be obtained with increasing the Coulomb interactions, as shown in Fig. 1(a). It is found that spin configurations A and G satisfy relation (I), and spin configuration C satisfies relation (II). Figure 1(b) shows that an increasing JT coupling narrows gradually the C-type AF region. This is because the JT coupling tends to align the orbital states along $|x\rangle$ and $|y\rangle$, and so raises the effective ferromagnetic coupling in the *x*-*y* plane and the AF coupling in the *z* direction, making the C-type AF spin configuration unstable.

We next discuss the orbital excitation spectra. Owing to the absence of SU(2) symmetry in the orbital Hamiltonian, an orbital excitation spectrum usually has an energy gap. As shown in Fig. 2, for A-, C- and G-type AF spin configurations, there is always an energy gap in the orbital spectrum, regardless whether or not the JT coupling is taken into account. However, if the JT coupling is absent, gapless orbital spectra may appear for the F spin configuration, as shown in the right top panel of Fig. 2. Furthermore, if relation (II) is satisfied, the orbital spectrum has a two-dimensional form: $\epsilon_{\mathbf{k}\sigma}=6u_x\sqrt{1+\sigma(\cos k_x+\cos k_y)/2}$. For such a two-dimensional system, quantum and thermal fluctuations, characterized by the second term of M_σ in Eq. (15), will completely destroy long-range orbital ordering at finite temperatures,[19] resulting in an orbital-liquid state similar to that obtained by Ishihara, Yamanaka, and Naguosa[20] The orbital excitation gap can be widened by the JT field acting on the orbital states. It is very similar to an anisotropic magnetic crystal field on the spin states in an AF Heisenberg Hamiltonian. Quantum fluctuations are greatly suppressed by this JT field, making the orbital ordering stable. For comparison, the spin excitation spectra are also plotted in the left-column panels, in which all the spin excitations are gapless due to the SU(2) symmetry of H_S and the JT coupling has little significant effect on them.

At finite temperatures, $\langle S_{\rm sub}\rangle$ in Eq. (8) and M_σ in Eq. (14) serve as the spin and orbital order parameters, respectively. Both of them decrease with increasing the temperature, and $\langle S_{\rm sub}\rangle$ (M_σ) vanishes as the temperature is increased beyond the critical temperature T_N (T_O). One may evaluate $\langle S_{\rm sub}\rangle$ and M_σ from Eqs. (8) and (15). In our calculation, parameters J_1, J_2 and J_3 are taken from the Racah parameters[21] and $t=0.41$ eV.[12] The system is found to have an A-type AF spin configuration at low temperatures. In Fig. 3 we plot the variation of T_N and T_O as functions of the strength of the JT coupling. Both T_N and T_O increase with the JT coupling, but there are different physical origins. The

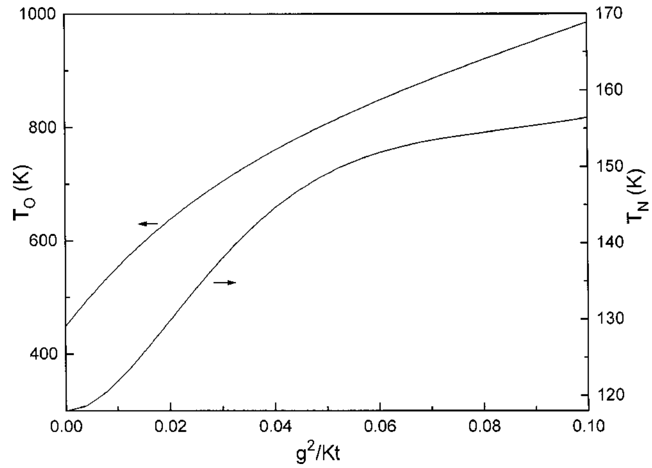

FIG. 3. Critical temperatures T_N and T_O as functions of g^2/Kt.

increase of T_N is attributed to an enhancement of the effective magnetic coupling $\tilde{J}_x$ and $\tilde{J}_y$ caused by the JT field. On the other hand, the increase of T_O stems from the fact that a stronger JT field will widen the energy gap of the orbital excitation spectrum, and so a higher temperature is required to excite orbital quasiparticles to break the long-range orbital ordering. According to experimental data and theoretical analysis, g is of the same order of magnitude as t and K is greater than g by a factor of 10–100,[22,23] so that g^2/K is the order of $0.01t \sim 0.1t$. According to Fig. 3, to fit with $T_O = 780$ K measured by the experiment, the strength of JT coupling should be $g^2/K = 0.045t$, at which the calculated $T_N = 146$ K is very close to the experimental value of $T_N = 140$ K. The present calculation may overestimate the critical temperatures due to neglecting the frequency-softened effect for the excitation spectrum at high temperatures, and so the required strength of JT coupling may be greater than the evaluated magnitude.

In summary, we have studied the excitation spectra of the spin and orbital degrees of freedom in undoped manganites, with the JT coupling and the Coulomb interactions taken into account. It is found that the observed A-type AF spin configuration has an electronic mechanism, resulting from anisotropic properties of the orbital operators. The JT coupling can considerably stabilize the magnetic ordering and in particular the orbital ordering at finite temperatures. Self-consistent calculations give T_N and T_O quantitatively comparable to the experimental data.

ACKNOWLEDGMENTS

This work was supported by a CRCG grant at the University of Hong Kong and a RGC grant of Hong Kong (HKU7113/00P). The work was also supported by a grant for the State Key Program for Basic Research of China and the National Natural Science Foundation of China.

[1] J. B. Goodenough, Phys. Rev. **100**, 564 (1955).
[2] Y. Murakami *et al.*, Phys. Rev. Lett. **81**, 582 (1998).
[3] K. I. Kugel and D. I. Khomskii, ZhETE **64**, 1429 (1973).
[4] S. Ishihara, J. Inoue, and S. Maekawa, Physica C **263**, 130 (1996).
[5] A. Moreo, S. Yunoki, and E. Dagotto, Science **283**, 2034 (1999).
[6] R. Shiina, T. Nishitani, and H. Shiba, J. Phys. Soc. Jpn. **66**, 3159 (1997).
[7] R. Maezono, S. Ishihara, and N. Nagaosa, Phys. Rev. B **58**, 11583 (1998).
[8] J. van den Brink, P. Horsch, F. Mack, and A. M. Oleś, Phys. Rev. B **59**, 6795 (1999).
[9] T. Hotta, S. Yunoki, M. Mayr, and E. Dagotto, cond-mat/9907034 (1999).
[10] A. J. Millis, Phys. Rev. B **55**, 6405 (1996).
[11] S. Q. Shen and Z. D. Wang, Phys. Rev. B **59**, 3291 (1999); **58**, R8877 (1998); **61**, 9532 (2000); S. Q. Shen, R. Y. Gu, Q. H. Wang, Z. D. Wang, and X. C. Xie, *ibid.* **62**, 5829 (2000); R. Y. Gu, Z. D. Wang, S. Q. Shen, and D. Y. Xing, *ibid.* **61**, 1211 (2000); X. J. Fan, S. Q. Shen, Z. D. Wang, X. G. Li, and Q. H. Wang, *ibid.* **62**, 3869 (2000).
[12] L. F. Feiner and A. M. Oleś, Phys. Rev. B **59**, 3295 (1999).
[13] W. E. Pickett and D. J. Singh, Phys. Rev. B **53**, 1146 (1996).
[14] I. Solovyev, N. Hamada, and K. Terakura, Phys. Rev. Lett. **76**, 4825 (1996); I. Solovyev, N. Hamada, and K. Terakura, Phys. Rev. B **53**, 7158 (1996).
[15] J. Kanamori, J. Appl. Phys. **31**, 14S (1960).
[16] G. Matsumoto, J. Phys. Soc. Jpn. **29**, 606 (1970).
[17] T. Mizokawa and A. Fujimori, Phys. Rev. B **51**, 12880 (1995); **54**, 5368 (1996).
[18] T. Mizokawa, D. I. Khomskii, and G. A. Sawatzky, Phys. Rev. B **60**, 7309 (1999).
[19] P. C. Hohenberg, Phys. Rev. **158**, 153 (1967).
[20] S. Ishihara, M. Yamanaka, and N. Nagaosa, Phys. Rev. B **56**, 686 (1997).
[21] U, U' and J are connected through three Racah parameters by $U = A + 4B + 3C$, $U' = A - B + C$ and $J = 5B/2 + C$. Taking $A \approx 4.7$ eV [Ref. 12], $B = 0.107$ and $C = 0.477$ eV [A. E. Bocquet *et al.*, Phys. Rev. B **46**, 3771 (1992)], one has $U \approx 6.6$, $U' = 5.1$ and $J = 0.745$ eV. $J_H \approx 1.2$ eV is estimated from $La_{1-x}Sr_xMnO_3$ at $x = 0.175$. [M. Imada *et al.*, Rev. Mod. Phys. **70**, 1039 (1998)].
[22] Y. Okimoto, A. Fujimori, and Y. Tokura, Phys. Rev. Lett. **75**, 109 (1995); Phys. Rev. B **55**, 4206 (1997).
[23] A. J. Millis, Phys. Rev. B **53**, 8434 (1996); Y. Motome and M. Imada, J. Phys. Soc. Jpn. **68**, 16 (1999).