Spin and orbital excitation spectrum in the Kugel-Khomskii model

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We discuss spin and orbital ordering in the twofold orbital degenerate superexchange model in three dimensions relevant to perovskite transition metal oxides. We focus on the particular point on the classical phase diagram where orbital degeneracy is lifted by quantum effects exclusively. Dispersion and damping of the spin and orbital excitations are calculated at this point taking into account their mutual interaction. Interaction corrections to the mean-field order parameters are found to be small. We conclude that quasi-one-dimensional Néel spin order accompanied by the uniform $d_{3z^2-r^2}$-type orbital ordering is stable against quantum fluctuations. [S0163-1829(97)50646-1]

It is well known that the orbital (quasi)degeneracy of $3d$ states in transition metal oxides plays an important role in their magnetic and lattice properties. An orbital ordering driven by exchange interactions and/or by Jahn-Teller effect occurs at low temperature resulting in a rich variety of magnetic structures (for a review see, e.g., Ref. 1). On the other hand, little is known on dynamical aspects of the coupling between spin and orbital degrees of freedom in these systems, i.e., on (a) what the spectrum is of low-energy orbital excitations, (b) how orbital excitations are coupled to the spin sector, and (c) how this coupling affects magnetic order parameter and spin waves. In the present paper we address these questions by considering the superexchange model with twofold orbital degeneracy, which corresponds to the d^9 Mott-Hubbard insulator on a cubic lattice.

To be specific, we consider the following Hamiltonian derived by Kugel and Khomskii,[1] and studied recently by Feiner *et al.*:[2]

$$H=\frac{t^2}{U}\sum_{\langle ij\rangle}\,[4(\vec{S}_i\vec{S}_j)(\tau_i^\alpha-\tfrac{1}{2})(\tau_j^\alpha-\tfrac{1}{2}) +(\tau_i^\alpha+\tfrac{1}{2})(\tau_j^\alpha+\tfrac{1}{2})-1]. \quad (1)$$

In Eq. (1) we follow notations used in Ref. 2: t is the hopping between $e_g(3z^2-r^2)$ orbitals along the c axis, $\vec{S}_i$ is the spin-1/2 operator. Operators τ_i^α act in the orbital subspace with basic vectors $\binom{1}{0}$, $\binom{0}{1}$ corresponding to the $e_g(x^2-y^2)\sim|x\rangle$ and $e_g(3z^2-r^2)\sim|z\rangle$ orbital states, respectively. The structure of τ_i^α depends on the index α which specifies the orientation of the bond $\langle ij\rangle$ relative to the cubic axes a,b,c:

$$\tau_i^{a(b)}=\tfrac{1}{4}(-\sigma_i^z\pm\sqrt{3}\sigma_i^x),\quad \tau_i^c=\tfrac{1}{2}\sigma_i^z, \quad (2)$$

where σ^z and σ^x are Pauli matrices.

It is rather easy to see that the classical Néel state [i.e., $\langle\vec{S}_i\vec{S}_j\rangle=-1/4$ in Eq. (1)] is infinitely degenerate: orbitals at each site may be rotated independently. Feiner *et al.*[2] have suggested that local orbital fluctuations associated with this degeneracy strongly affect the spin sector when quantum fluctuations around the Néel state are included, and drive the system into a disordered spin-liquid state even in three dimensions.[3] Our results presented below do not support this interesting scenario. We have investigated spin and orbital orderings, and their excitations in the model defined by Eqs. (1) and (2). Our main findings are (i) $|z\rangle$-type orbital ordering favoring quasi-one-dimensional spin order is the most promising candidate for the ground state. (ii) Orbital excitations have a gap generated by quantum effects. This gap controls well the fluctuations around the mean-field solution. (iii) Spin-orbit coupling does indeed act to decrease the staggered moment, but this effect is not enough to destroy the long-range order in a cubic lattice.

To begin with, we use the condition $\Sigma_{\langle i,j\rangle}\tau_i^\alpha=0$ following from Eq. (2), and represent Eq. (1) in a more transparent way:

$$H=-3+\sum_{\langle i,j\rangle}\hat{J}_\alpha^{ij}(\vec{S}_i\vec{S}_j+\tfrac{1}{4}), \quad (3)$$

$$\hat{J}_\alpha^{ij}=4\tau_i^\alpha\tau_j^\alpha-2(\tau_i^\alpha+\tau_j^\alpha)+1. \quad (4)$$

The first term in Eq. (3) represents the classical Néel energy (in units of t^2/U), which we drop hereafter. From the above Hamiltonian the key feature of the Kugel-Khomskii model is evident: The exchange ''constant'' has in fact an internal operator structure accounting for the orbital dynamics, and its expectation value strongly depends on the orientation of orbitals. It follows from Eqs. (3) and (4) that the strength of the intersite orbital coupling (hence the energy gain due to the orbital ordering) is proportional to the deviation of spins from the Néel state, i.e., to the value of $\langle\vec{S}_i\vec{S}_j+\tfrac{1}{4}\rangle$. This acts to reduce the effective dimensionality of the spin system: Orbitals are arranged in such a way that makes the exchange coupling strongly nonuniform thus enhancing spin fluctuations as much as possible. In low-dimensional models, a similar consideration suggests that the orbital ordering may lead to the spin-liquid state.[4] The z-type

TABLE I. Néel order parameter $\langle S^z\rangle$ and some other expectation values (see text for notations) calculated in the self-consistent mean-field approximation ($V=0$), and corrected by including fluctuation effects ($V\neq 0$).

	$\langle S^z\rangle$	$\langle J_\perp\rangle/\langle J_c\rangle$	$\langle \vec{S}_i\vec{S}_j\rangle_c$	$\langle \vec{S}_i\vec{S}_j\rangle_\perp$	E_{mf}	E_0
$V=0$	0.226	0.052	−0.417	−0.122	−0.609	−0.609
$V\neq 0$	0.191	0.072	−0.421	−0.103	−0.564	−0.690

ordering of orbitals in the model (1) is suggested by this picture. Indeed, the expectation value of exchange coupling (4) between z orbitals is $J_c=4$ along the c axis, and it is only small in the (ab) plane: $J_\perp=1/4$. Exchange energy is mainly accumulated in c chains and can be approximated as $J_c\langle \vec{S}_i\vec{S}_j+\frac{1}{4}\rangle_c+2J_\perp\langle \vec{S}_i\vec{S}_j+\frac{1}{4}\rangle_\perp\simeq -0.65$ per site (using $\langle \vec{S}_i\vec{S}_j\rangle_c=1/4-\ln 2$ for one dimension[5] (1D) and assuming $\langle \vec{S}_i\vec{S}_j\rangle_\perp\sim 0$). On the other hand, x-type ordering results in the easy plane magnetic structure ($J_{a,b}=9/4, J_c=0$) with a much smaller energy gain $\simeq -0.38$.

Our strategy is to study the Hamiltonian (3) within the following scheme. (i) We rewrite (3) in the form $H=H_{sp}+H_{orb}+H_{int}$. Here the first two terms describe spin and orbital sectors in the mean-field level:

$$H_{sp}=\sum_{\langle i,j\rangle}\langle \hat{J}^{ij}_\alpha\rangle(\vec{S}_i\vec{S}_j+\tfrac{1}{4}), \tag{5}$$

$$H_{orb}=\sum_{\langle i,j\rangle}\langle \vec{S}_i\vec{S}_j+\tfrac{1}{4}\rangle\delta(\hat{J}^{ij}_\alpha), \tag{6}$$

where $\delta A=A-\langle A\rangle$. The crucial importance is the stability of the mean-field state against fluctuations generated by dynamical coupling between spin and orbital excitations. This coupling is represented by

$$H_{int}=\sum_{\langle i,j\rangle}\delta(\hat{J}^{ij}_\alpha)\delta(\vec{S}_i\vec{S}_j). \tag{7}$$

(ii) We assume the antiferromagnetic spin order and uniform z- or x-type ordering of orbitals. Then we employ spin (orbital) wave representation for $\vec{S}_i$ ($\vec{\sigma}_i$) operators. (iii) We calculate spin-orbit interaction corrections to the excitation spectrum and to the order parameters. Since latter quantities enter in coupling constants in Eqs. (5) and (6), all steps have to be done in the self-consistent way.

Consider z orbital order which results in a highly anisotropic quasi-1D magnetic structure. We discuss first mean-field results, which follow from Eqs. (5) and (6). Spin and orbital wave energies are given by $\omega_{1k}=J_1\sqrt{1-\gamma_{1k}^2}$, and $\omega_{2k}=J_2\sqrt{1+2\gamma_{2k}}$, respectively. Here $J_1=(J_c+2J_\perp)$, and

$$J_c=\langle 1-\sigma_i^z-\sigma_j^z+\sigma_i^z\sigma_j^z\rangle_c, \tag{8}$$

$$J_\perp=\langle 1+\tfrac{1}{2}\sigma_i^z+\tfrac{1}{2}\sigma_j^z+\tfrac{1}{4}\sigma_i^z\sigma_j^z+\tfrac{3}{4}\sigma_i^x\sigma_j^x\rangle_\perp .$$

The orbital stiffness is controlled by $J_2=-8(\kappa_c-\frac{1}{4}\kappa_\perp)$, with $\kappa_\alpha=\langle \vec{S}_i\vec{S}_j+\frac{1}{4}\rangle_\alpha$. Momentum dependencies of ω_{nk} (index $n=1,2$) are determined by the functions $\gamma_{1k}=(J_c\cos k_z+2J_\perp\gamma_k)/J_1$, $\gamma_{2k}=-\frac{3}{2}[\kappa/(4-\kappa)]\gamma_k$, where

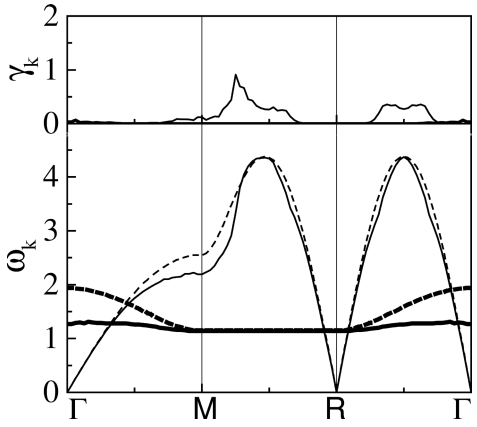

FIG. 1. Dispersion ω_k and damping γ_k of the spin and orbital waves along the direction $\Gamma\to M(\pi,\pi,0)\to R(\pi,\pi,\pi)\to\Gamma$ in the Brillouin zone, calculated including fluctuation effects (solid lines), and in the mean-field approximation (dashed lines). Thin (thick) lines correspond to the spin (orbital) excitation. γ_k for orbital waves is almost indistinguishable from the zero line.

$\kappa=\kappa_\perp/\kappa_c$, and $\gamma_k=\frac{1}{2}(\cos k_x+\cos k_y)$. We calculate all expectation values within linear spin (orbital) wave theory, with only one exception, the interchain spin correlator which we approximate as $\langle \vec{S}_i\vec{S}_j\rangle_\perp=\langle \vec{S}_i^z\rangle\langle \vec{S}_j^z\rangle+\langle S_i^+S_j^-\rangle$.[6]

Self-consistent mean-field calculations show that the orbital pseudospin is almost saturated (the mixture of $|x\rangle$ state is about 1% only). Coupling between chains $J_\perp$ is weak (see Table I) but sufficient to produce quite large magnon dispersion in the (ab) plane (see thin dashed lines in Figs. 1 and 2). Orbital excitations are gapfull, since the orbital ordering is not associated with the breaking of any continuous symmetry. Of a similar spirit, a mean-field picture was recently discussed by Ishihara *et al.*[7] in the context of their spin-orbital model for manganites. Quantitatively, we find that the orbital gap is smaller than the spin-wave bandwidth. The softness of the orbital excitations is related to the fact that the orbital degeneracy in the model (1) can be lifted only due to quantum effects in the spin sector.

Now, what happens when we switch on the coupling between spin and orbital excitations? The latter is represented by H_{int} [Eq. (7)], which in terms of spin (β_k) and orbital (φ_k) wave excitations reads as

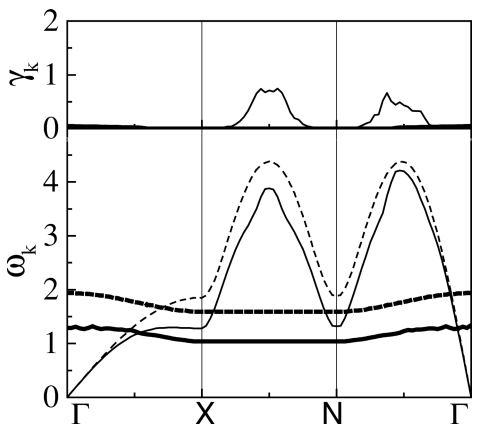

FIG. 2. The same as Fig. 1 but along the $\Gamma\to X(\pi,0,0)\to N(\pi,0,\pi)\to\Gamma$ direction.

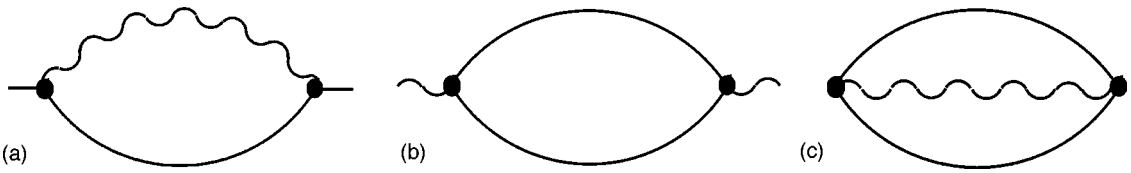

FIG. 3. Spin-orbit interaction corrections to the spin (a) and orbital excitations (b) and to the ground-state energy (c). Lines (wavy lines) represent spin (orbital) waves.

$$H_{int}=-V\sum_{kp}\{f_{k,p}\beta_k^+\beta_p \tag{9}$$

$$+\tfrac{1}{2}g_{k,p}(\beta_k^+\beta_{-p}^+ +\beta_{-k}\beta_p)\}(\varphi_q$$

$$+\varphi_{-q}^+),$$

where the lowest-order (three magnon) terms only are kept. Here $V=\sqrt{3}/2$, and $q=k-p$. The matrix elements are

$$f_{k,p}=(u_{2q}+v_{2q})[\eta_q M_{k,p}+(\eta_k+\eta_p)N_{k,p}], \tag{10}$$

$$g_{k,p}=(u_{2q}+v_{2q})[\eta_q N_{k,p}+(\eta_k+\eta_p)M_{k,p}],$$

$$M_{k,p}=(u_{1k}u_{1p}+v_{1k}v_{1p}),\quad N_{k,p}=(u_{1k}v_{1p}+v_{1k}u_{1p}),$$

with $\eta_k=(\cos k_x-\cos k_y)/2$. The Bogoliubov transformation coefficients in the spin subspace are given by $u_{1k}=\{(s+1)/2\}^{1/2}$, $v_{1k}=-\{(s-1)/2\}^{1/2}\mathrm{sgn}\gamma_{1k}$, and $s=(1-\gamma_{1k}^2)^{-1/2}$. The factor $(u_{2k}+v_{2k})=(1+2\gamma_{2k})^{-1/4}$ in Eq. (10) is due to Bogoliubov transformation in the orbital sector.

Physically, the interaction (9) accounts for the process when spin exchange is accompanied by the simultaneous orbital transition $|z\rangle\leftrightarrow|x\rangle$, thus enhancing the x orbital component in the ground state. Spin-orbital coupling leads to the conventional 2×2 matrix bosonic Green's function in both subspaces, with diagonal (G) and nondiagonal (F) components given by

$$G_{\omega,k}=[(i\omega-A_{\omega,k})+(\omega_k+S_{\omega,k})]/\ \mathrm{Det}, \tag{11}$$

$$F_{\omega,k}=-\Sigma_{\omega,k}^{(a)}/\ \mathrm{Det},$$

$$\mathrm{Det}=(i\omega-A_{\omega,k})^2$$

$$-(\omega_k+S_{\omega,k}-\Sigma_{\omega,k}^{(a)})(\omega_k+S_{\omega,k}$$

$$+\Sigma_{\omega,k}^{(a)}).$$

Here $A_{\omega,k}$ and $S_{\omega,k}$ represent the antisymmetric and symmetric (with respect to the Matsubara frequency $i\omega$) components of the diagonal self-energy $\Sigma_{\omega,k}^{(n)}$, respectively, while $\Sigma_{\omega,k}^{(a)}$ is a nondiagonal element of the self-energy matrix. It is implied that all quantities in Eq. (11) carry the subspace index n as well, and $n=1$ (2) stands for spin (orbital) waves. We calculate self-energies from the lowest-order diagrams shown in Fig. 3.

In spin subspace we find (at zero temperature)

$$A_{\omega,k}=V^2\sum_p\ (f_{k,p}^2-g_{k,p}^2)\frac{\omega}{(\omega+i\delta)^2-\varepsilon_{k,p}^2}, \tag{12}$$

$$S_{\omega,k}\pm\Sigma_{\omega,k}^{(a)}=V^2\sum_p\ (f_{k,p}\pm g_{k,p})^2\frac{\varepsilon_{k,p}}{(\omega+i\delta)^2-\varepsilon_{k,p}^2}.$$

Here $\varepsilon_{k,p}=(\omega_{1p}+\omega_{2q})$, $q=k-p$, and

$$(f^2-g^2)_{k,p}=[\eta_q^2-(\eta_k+\eta_p)^2]x_{2q}, \tag{13}$$

$$(f\pm g)_{k,p}^2=(\eta_k+\eta_p\pm\eta_q)^2(x_{1k}x_{1p})^{\pm1}x_{2q},$$

where $x_{1k}=[(1-\gamma_{1k})/(1+\gamma_{1k})]^{1/2}$, $x_{2k}=(1+2\gamma_{2k})^{-1/2}$.

In the orbital sector one finds that $A_{\omega,k}=0$, and

$$S_{\omega,k}=\Sigma_{\omega,k}^{(a)}=2V^2\sum_p\ g_{k+p,p}^2\frac{\tilde{\varepsilon}_{k,p}}{(\omega+i\delta)^2-\tilde{\varepsilon}_{k,p}^2}, \tag{14}$$

where $\tilde{\varepsilon}_{k,p}=(\omega_{1p}+\omega_{1,k+p})$, and $g_{k+p,p}^2$ can be found from Eq. (13). We recall that the ''bare'' energies ω_{nk} in Eqs. (11)–(14) are also affected by the interaction, due to the renormalization of parameters J_n and γ_{nk}.

Results of self-consistent calculations by including interaction corrections are presented in Table I and Figs. 1 and 2. Dynamical spin-orbit coupling results in the following: (i) It enhances quantum fluctuations in both subspaces, thus reducing the staggered moment and increasing the weight of the x orbital component (which is about 6%). The latter effect is also reflected in a larger value of the ratio $J_\perp/J_c$. (ii) Spin and orbital excitations are both softened, which is more pronounced in the $k_y=0$ plane (and in equivalent ones), see Fig. 2. The orbital gap still remains well defined. A spin-Peierls-like instability is absent, because of the vanishing matrix elements in Eq. (9) for momenta along z [note $\eta(0,0,q_z)=0$], and because of the finite interchain coupling. (iii) Spin waves get a finite damping. Orbital waves are almost undamped since the density of spin states inside the orbital gap is small. (iv) Joint spin-orbital fluctuations significantly lower the ground-state energy (see Table I). The latter is given by $E_0=E_{mf}+\langle H_{int}\rangle$, where the interaction correction to the mean-field result, calculated from the last diagram in Fig. 3, is

$$\langle H_{int}\rangle=-V^2\sum_{k,p}\ g_{k,p}^2/(\omega_{1k}+\omega_{1p}+\omega_{2,k-p}). \tag{15}$$

The exchange energy gain $E_0=-0.69$ per site is found, which is close to our above estimation from physical considerations. Summarizing, interaction effects do not qualitatively change predictions of the self-consistent mean-field theory, which seems to work quite reasonably. This is an important observation, giving some credit to the mean-field ansatz in studying more complicated spin-orbital models. Of

course, the latter fails when the orbital gap is softened close to the phase boundaries between different orbitally ordered states, and the dynamical spin-orbit coupling becomes of crucial importance.

Considering x-type ordering, we found it to be unstable against fluctuations. It turns out that orbital excitations around this mean-field state are gapless at the Γ point, $\omega_{2q} \sim q$. In addition, the spin-orbit interaction vertex remains finite at $q=0$, since the orbital pseudospin is not a conserved quantity, and orbital waves cannot be considered as Goldstone modes. All these lead to the divergencies in perturbation theory indicating that an x-type ordered state is not an appropriate one, as we already mentioned above. This result is consistent with Ref. 2.

In summary, we have studied the spin-orbital coupling problem in the specific model, where this coupling is particularly important because of infinite degeneracy of the classical Néel state in this model. The problem of the orbital frustration pointed out in Ref. 2 is actually removed by reducing the effective dimensionality of the spin system. Quantum spin fluctuations then generate an orbital excitation gap through the spin-orbit coupling mechanism. Orbital degeneracy in the model (1) should manifest itself in a strong reduction of the Néel temperature, by favoring soft quasi-1D spin structure. This is consistent with a basic idea of Feiner *et al.*[2] that orbital degeneracy, in general, acts to enhance quantum spin fluctuations. In contrast to Ref. 2 we find, however, that this effect is not strong enough to destroy the Néel order. Melting of the long-range magnetic order by orbital fluctuations suggested in Ref. 2 does not occur in a cubic perovskite system, for a simple reason: A certain (model-dependent) orbital ordering always results in the three-dimensional (albeit very anisotropic) network of exchange interactions among spins. *Three-dimensionality* of the spin sector and existence of the *orbital gap* are important factors stabilizing the Néel order. We believe that the orbital gap is a robust property of Mott-Hubbard insulators, which is related to the fact that the underlying symmetry in orbital subspace is only a discrete one. In a metallic state, doped holes can drastically change the situation, by inducing low-energy orbital fluctuations.[8] A study of the orbital melting in the Kugel-Khomskii model, driven by hole doping, deserves further work.

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