Orbitons in ferromagnetic insulating manganites

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Abstract

Starting from the classical ground state with orbital order, we investigate the orbital excitations (orbitons) and quantum corrections in the model for insulating ferromagnetic LaMnO$_3$ using linear spin-wave theory. It is found that the effective dimensionality of the model is reduced, and the quantum corrections are smaller than in the respective Heisenberg model. A gap in the orbiton spectrum opens away from the orbital degeneracy, but disappears again at the point where the crystal-field splitting compensates the superexchange and the occupied orbitals become identical, but the quantum corrections are finite and follow from the lack of orbital conservation. © 1999 Elsevier Science B.V. All rights reserved.

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Orbital degeneracy of d electrons in transition metal oxides leads to low-energy effective models with both spin and orbital degrees of freedom [1,2], which pose an interesting problem, even without doping. In the ground state orbital interactions lead to an orbital ordering that is interrelated to the magnetic ordering, and the elementary excitations have both spin and orbital character [3,5]. Here we consider a uniform ferromagnetic (FM) phase with one e_g electron per site. In a large magnetic field the spin operators can be integrated out. Such a state could possibly be realized in LaMnO$_3$ at large magnetic field and serves as a reference state to consider hole propagation in doped manganites. The effective Hamiltonian with orbital antiferromagnetic (AF) super-exchange is [2,4],

$$H_J = \tfrac{1}{2}J \sum_{\langle ij \rangle \parallel} [T_i^z T_j^z + 3T_i^x T_j^x \pm \sqrt{3}(T_i^x T_j^z + T_i^z T_j^x)]$$

$$+ 2J \sum_{\langle ij \rangle \perp} T_i^z T_j^z - E_z \sum_i T_i^z, \qquad (1)$$

where T_i^x and T_i^z are orbital operators with identical commutation relations to spin operators, and the mixed term has the opposite signs for the x and y-direction and the constant energy $-J/2$ per bond is neglected. The bonds labelled as $\langle ij \rangle \parallel (\langle ij \rangle \perp)$ connect neighboring sites within (a, b) planes (along the c-axis). The crystal-field E_z removes the degeneracy of e_g orbitals.

To study the classical ground state of the system we introduce two rotations in orbital space and minimize the energy with respect to the rotation angle. For $E_z = 0$ the ground state is characterized by the two-sublattice AF order of occupied orbitals. We make a uniform rotation of orbitals by an angle θ at each site so that the relative angle between the orbitals is invariant under the rotation. One finds that the classical energy is $-9J/4$ per site and does not depend on the rotation angle. In linear spin-wave theory the orbiton dispersion depends on θ and has two branches,

$$\omega_k^{\pm}(\theta) = 3J[1 \pm \tfrac{1}{6}(\sqrt{3}\cos 2\theta - \sin 2\theta)^2 \gamma_x(\boldsymbol{k})$$

$$\pm \tfrac{1}{6}(\sqrt{3}\cos 2\theta + \sin 2\theta)^2 \gamma_y(\boldsymbol{k})$$

$$\pm \tfrac{2}{3}\sin^2 2\theta \gamma_x(\boldsymbol{k})]^{1/2}, \qquad (2)$$

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with $\gamma_\alpha(\mathbf{k}) = \cos k_\alpha$, $\alpha = x, y, z$, respectively. The excitations are gapless at the Γ point, independent of the rotation angle. A priori, such a gapless mode is not expected, as Hamiltonian (1) does not have a continuous symmetry. However, the classical states are rotationally invariant and the cubic symmetry of the model is restored only on the quantum level. The zero-point energy ΔE and quantum corrections ΔT^z to the order-parameter are invariant only under rotations by an angle $\theta = n\pi/6$ (Fig. 1). Note that the quantum corrections are somewhat lower than for the three-dimensional (3D) AF Heisenberg model.

For $\theta = 0$ the dispersion is completely two-dimensional (2D), $\omega_k^\pm(0) = 3J\sqrt{1 \pm \gamma_\parallel(\mathbf{k})}$ where $\gamma_\parallel(\mathbf{k}) = \frac{1}{2}$ $(\cos k_x + \cos k_y)$. In this case the quantum corrections are largest, as this dispersion has a line of nodes as $\omega_{(0, 0, q)}^- = 0$ for any q. It is interesting to note that this fully 2D orbital-wave dispersion leads to an orbital ordered state at zero temperature only. At any finite temperature fluctuations destroy the long-range order (as in the 2D Heisenberg model). In this way, one obtains an orbital liquid at non-zero temperature. However, it might well be that this excitation develops a gap in higher-order spin-wave theory. We expect that if this gap arises, it remains small and proportional to the size of quantum fluctuations.

The ground state for $E_z \neq 0$ leads to a particular linear combination of occupied orbitals, in analogy to a spin-flop phase in the AF Heisenberg model at finite magnetic field. The tilting of the orbitals in the direction of the field E_z is described by a rotation of the orbitals on one sublattice with the angle $-\phi$ and on the other with angle ϕ, starting from the orbitals given by $\theta = \pi/4$ at $E_z = 0$. The mean-field energy for the 3D model is $E_{3D}^{MF} = -\frac{3}{4}J \cos 4\phi$ and is minimized by $\sin 2\phi = E_z/6J$. For the 2D model $E_{2D}^{MF} = -\frac{1}{4}J(2 \cos 4\phi + 1)$ and $\sin 2\phi = E_z/4J$. One finds the same energy of $-3J/4$ in the 2D and 3D model at orbital degeneracy. This shows
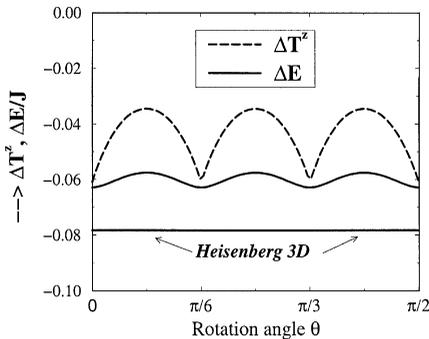
that the *orbital superexchange interactions are geometrically frustrated*. Bonds in the third dimension do not lower the mean-field energy apart from the constant term, but restore the cubic symmetry on the quantum level. For $E_z \neq 0$ the spectrum becomes gapped because the degeneracy of the classical ground state is removed and the dispersion of the orbitons shown in Fig. 2, is given by

$$\omega_k^{\phi\pm} = 3J[1 \pm \tfrac{1}{3}(2 \cos 4\phi - 1)\gamma_\parallel(\mathbf{k})$$
$$\pm \tfrac{1}{3}(\cos 4\phi + 1)\gamma_z(\mathbf{k})]^{1/2}. \qquad (3)$$

The gap as a function of E_z is shown in Fig. 3. For small orbital fields the gap first increases. For larger values the gap decreases and for the 3D system it vanishes at $E_z = 6J$. This can be understood by the analogy of an AF Ising model in a magnetic field. As the field increases, the energy for a spin-flip excitation decreases as in making an excitation the system gains energy because of the parallel
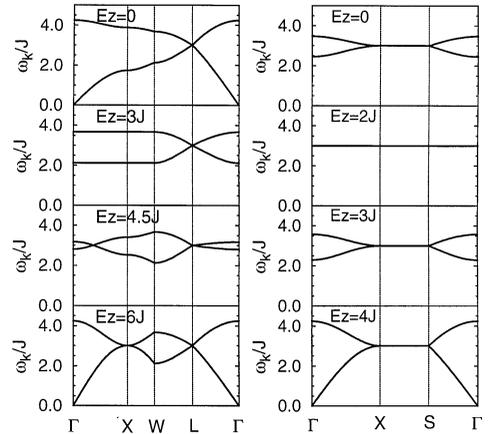

Fig. 2. Orbital wave excitations in a 3D (left) and 2D (right) model for different values of the crystal-field splitting.

Fig. 1. Quantum corrections to order parameter ΔT^z and ground state energy $\Delta E/J$ for the 3D model as functions of angle θ.
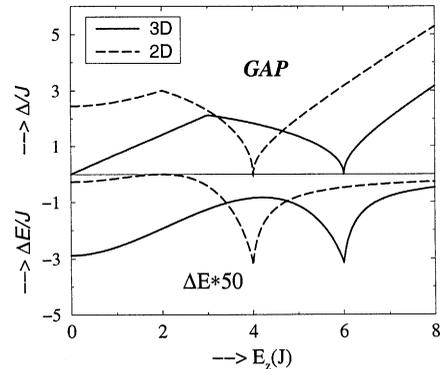

Fig. 3. Gap Δ/J in the orbiton spectrum and quantum correction to the ground state energy $\Delta E/J$ as function of E_z/J for a 3D and 2D model.

alignment of the excited spin to the field. Approaching the phase transition from AF to FM ordering, this modes becomes softer and is at zero energy exactly at the transition point. The increase of the gap for $E_z > 6J$ is therefore due to the fact that in this parameter regime the system is completely ferromagnetically aligned along the field axis, and any excitation acts against the orbital field. In Fig. 3 also the quantum corrections to the ground state energy are shown as a function of the orbital field. The presence of a gap in the excitation spectrum naturally suppresses quantum fluctuations. For the 2D system the missing interactions along the c-axis induce a gap in the excitation spectrum, which closes at a critical field, as in the 3D system.

In conclusion, we have shown that new purely orbital excitations exist in ferromagnetic systems close to orbital degeneracy. It is expected that they play an important role in the transport properties of the colossal magnetoresistance compounds.

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