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Orbital order out of spin disorder: how to measure the orbital gap

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Abstract. The interplay between spin and orbital degrees of freedom in the Mott–Hubbard insulator is studied by considering an orbitally degenerate superexchange model. We argue that orbital order and the orbital excitation gap in this model are generated through the order-from-disorder mechanism known previously from frustrated spin models. We propose that the orbital gap should show up indirectly in the dynamical spin structure factor; it can therefore be measured using the conventional inelastic neutron scattering method.

1. Introduction

The recent renaissance in the study of transition metal oxides has emphasized the important role being played by the orbital (pseudo-) degeneracy inherent to perovskite lattices. First of all, the type of spin structure and the character of spin excitations crucially depend on the orientation of the occupied orbitals [1–3]. Second, the excitations in the orbital sector get coupled to the other degrees of freedom (electronic, lattice, spin) and might therefore strongly modify their excitation spectra. It has also been suggested recently [4, 5] that low-energy orbital fluctuations are responsible for the highly correlated metallic state of ferromagnetic manganites.

Apparently, orbital order and orbital fluctuations deserve for careful theoretical and experimental study. However, the orbital excitation itself is spinless and chargeless and can therefore be detected only indirectly due to its coupling to other types of excitation. For instance, Ishihara *et al* [6] have recently discussed the possibility of detecting orbital excitations by means of the anomalous x-ray scattering method. In this paper we propose the idea of detecting the orbital excitation in a conventional inelastic neutron scattering experiment. That is, due to the inherent coupling of orbitals to the spin, a magnon and a single-orbital wave can be excited by neutrons. Below, we illustrate this idea by considering an antiferromagnetic model possessing an e_g orbital degeneracy.

2. Spin–orbital model

Physically realistic spin–orbital models [3, 7] are usually rather complicated. To make the discussion more transparent we consider a simplified version of the Kugel–Khomskii

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model [3, 8, 9]:

$$H = \sum_{\langle ij \rangle} \left[4(\vec{S}_i \cdot \vec{S}_j)\left(\tau_i^\alpha - \frac{1}{2}\right)\left(\tau_j^\alpha - \frac{1}{2}\right) + \left(\tau_i^\alpha + \frac{1}{2}\right)\left(\tau_j^\alpha + \frac{1}{2}\right) - 1 \right] \qquad (1)$$

describing the superexchange process in a Mott–Hubbard insulator with e_g degeneracy, where the Hund's splitting in the intermediate state has been neglected. Here, $\vec{S}_i$ is the spin-1/2 operator, while the operators τ_i^α act in the orbital subspace with basis vectors $(1, 0)$, $(0, 1)$ corresponding to the e_g orbital states $(x^2 - y^2) \sim |x\rangle$ and $e_g(3z^2 - r^2) \sim |z\rangle$, 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:

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

where σ^z and σ^x are Pauli matrices. The physical meaning of the τ-operators is to describe the fluctuations of exchange bonds due to the orbital dynamics. On the cubic lattice, equation (1) can be rewritten in the equivalent form:

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

where

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

The main feature of this model is the strong interplay between spin and orbital degrees of freedom which is suggested by the very form of Hamiltonian (3). In fact, the Kugel–Khomskii model contains rather nontrivial physics: the classical Néel state in equation (3) ($\langle \vec{S}_i \cdot \vec{S}_j \rangle = -1/4$) is infinitely degenerate due to the presence of the orbital sector; this extra degeneracy must be lifted by some mechanism. According to a recent proposal by Feiner, Oleś, and Zaanen [8, 9], this orbital-frustration problem is probably solved by the formation of a RVB-type spin-singlet ground state. Technically, their suggestion is based on the observation that spin–orbit coupling results in a new, composite excitation (simultaneous spin and orbital flip). This mode is found to be soft in certain directions in the momentum space, leading to one-dimensional fluctuations, thus completely destroying the magnetic order. In contrast, Khaliullin and Oudovenko [10] have found the orbital-flip excitation to be gapped. The authors hence concluded that the orbitally ordered quasi-one-dimensional quantum Néel state is the proper low-temperature fixed point of model (3).

The purpose of our paper is twofold. First, we re-examine the solution of Feiner *et al* [8, 9] and show that the soft spin–orbital mode does indeed acquire a finite gap through Villain's order-from-disorder mechanism [11]. Basically, this gap is determined by the orbital excitation energy. Second, we calculate the spectral weight of the composite spin–orbital excitation in the dynamical spin structure factor, suggesting the possibility of measuring the orbital excitation gap indirectly in a conventional neutron scattering experiment.

3. Excitation spectrum

To study the model (1) we follow the same scheme and method as were used by Feiner *et al* [8, 9]. Namely we start with the assumption of long-range staggered spin order and an uniform z-type orbital order (see figure 1).

Next we calculate the transverse spin-fluctuation spectrum by using the equation-of-motion method, and calculate the quantum corrections to the magnetic order parameter. We are motivated to use this method here for the following reason: employing a conventional

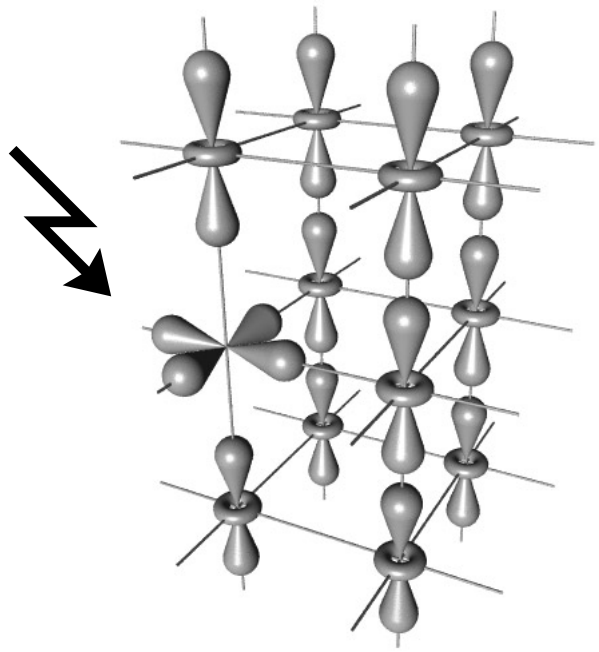

Figure 1. $|3z^2-r^2\rangle$ orbital order which leads to weakly coupled AF spin chains ($J_c = 4$, $J_\perp = 1/4$). As discussed in reference [10], this type of orbital ordering provides the largest energy gain due to quantum spin fluctuations. An orbital flip (indicated by an arrow) modulates the strength of the neighbouring exchange bonds, breaking the c-chain. In the classical Néel state, orbital excitations cost no energy. However, strong quasi-one-dimensional spin fluctuations stabilize this structure by opening the orbital gap through the order-from-disorder mechanism.

diagrammatic technique, it was argued in reference [10] that the orbital degeneracy of the classical state is removed by quantum spin fluctuations which generate orbital order and spontaneously break the cubic symmetry. The interesting question is then whether and how this order-from-disorder phenomenon manifests itself in the equation-of-motion *ansatz* of references [8, 9].

The quantity to be calculated is the Green's function $G_{i,i'} = \langle\langle S_i^+ | S_{i'}^- \rangle\rangle$, from which the dynamical spin structure factor as well as the reduction of the spin order parameter can be deduced. It obeys the following equation of motion:

$$\omega G_{i,i'} = \langle [S_i^+, S_{i'}^-] \rangle + \langle\langle [S_i^+, H] | S_{i'}^- \rangle\rangle. \tag{4}$$

Within a conventional spin-wave approximation, this equation reads

$$(\omega - \lambda m_i) G_{i,i'} = \delta_{i,i'} + 2m_i \sum_{j_c} G_{j,i'} + \frac{1}{8} m_i \sum_{j_\perp} G_{j,i'} + \frac{\sqrt{3}}{8} m_i \sum_{j_\perp} \alpha_{ij} D_{j,i'}. \tag{5}$$

The following notation is introduced here: $m_i = 1$ if site i belongs to sublattice A with up-spin orientation and $m_i = -1$ otherwise. The summations over j_c and $j_\perp$ are over nearest neighbours of site i in the c-chain and perpendicular directions, respectively. The factor $\lambda = J_c + 2J_\perp = 9/2$ and $\alpha_{ij} = |R_{ij}^x| - |R_{ij}^y|$. Distinct from a Heisenberg model, a new Green's function $D_{j,i'} = \langle\langle K_{jj}^x | S_{i'}^- \rangle\rangle$ appears in the above equation. The operator $K_{jj'}^x = \sigma_j^x S_{j'}^+$ describes a simultaneous orbital–spin excitation which couples to a single spin flip. Following references [8,9] we discarded the term $K_{j \neq j'}$ in equation (5), neglecting the correlation between orbital and spin excitations from different chains.

The dynamics of the composite excitations $K^{\pm}_{jj} = \sigma^{\pm}_j S^{+}_j$ is described by the equation

$$\left(\omega + 3m_i \mp \frac{3}{2}\right) K^{\pm}_{jj} = -\frac{3}{16}(m_i \pm 1) \sum_{l_\perp} \left(K^{x}_{ll} + \frac{\alpha_{jl}}{\sqrt{3}} S^{+}_l \right) + L^{\pm}_j \tag{6}$$

where the last term is

$$L^{\pm}_j = \sum_{l_c} \left\{ 2K^{\pm}_{jl}\, \delta S^{z}_j - 2K^{\pm}_{jj}\, \delta S^{z}_l - (m_i \mp 1) K^{\pm}_{jl} \right\}. \tag{7}$$

Here, site l is the nearest neighbour of site j, and δS^{z}_j is the fluctuating part of the spin operator S^{z}_j. The $L^{\pm}_j$-term in equations (6) and (7) accounts for the correlations within the c-chain. If one neglects this term (as Feiner *et al* [8, 9] did), one obtains from equations (5) and (6) a soft mode mentioned above, which results finally in a breakdown of the spin-ordered state initially assumed. We do not accept this approximation. It is the intra-chain quantum fluctuations represented by the $L^{\pm}_j$-term that play a crucial role by making the orbital excitations $\sigma^{\pm}$ (and consequently $\sigma^{\pm} S^{+}$) acquire a finite-mass gap. The underlying physics can easily be observed in the following way: we write the equations of motion for the operators in equation (7) treating the bond (jl) exactly while considering the remaining bonds of the c-chain in a static Néel approximation. This results in

$$[\omega - (m_i \pm 3)]\, K^{\pm}_{jl}\, \delta S^{z}_j = (1 \pm m_i)(K^{\pm}_{jj} \pm K^{\pm}_{jj}\, \delta S^{z}_l) \tag{8a}$$

$$(\omega \mp 2) K^{\pm}_{jj}\, \delta S^{z}_l = (1 \mp m_i)(K^{\pm}_{jl} \pm K^{\pm}_{jl}\, \delta S^{z}_j) \tag{8b}$$

$$[\omega - (3m_i \pm 1)]\, K^{\pm}_{jl} = 2K^{\pm}_{jj}\, \delta S^{z}_l - 2K^{\pm}_{jl}\, \delta S^{z}_j + (m_i \pm 1) K^{\pm}_{jj}. \tag{8c}$$

Many terms in these equations in fact drop out due to $|m_i| = 1$. Equations (8*a*)–(8*c*) together with equation (7) then lead to a very simple result—namely,

$$L^{+}_{j\in \mathrm{B}} = \frac{8}{\omega - 4} K^{+}_{jj} \qquad L^{-}_{j\in \mathrm{A}} = \frac{8}{\omega + 4} K^{-}_{jj} \tag{9}$$

and $L^{-}_{j\in \mathrm{B}} = 0$, $L^{+}_{j\in \mathrm{A}} = 0$. Now equation (6) can be written as follows (note $K^{x} = K^{+} + K^{-}$):

$$\left(\omega + \frac{3}{2} m_i\right) K^{x}_{jj} = -\frac{3}{8} m_i \sum_{l_\perp} \left(K^{x}_{ll} + \frac{\alpha_{jl}}{\sqrt{3}} S^{+}_l \right) + \Sigma_j K^{x}_{jj}. \tag{10}$$

The function $\Sigma_j = 8/(4m_j + \omega)$ is interpreted as a self-energy correction to the composite excitation $\sigma^{x}_j S^{+}_j$ due to quantum fluctuations about the Néel state. We think that the large-ω behaviour of Σ is not quite reliable since short-time fluctuations of the environment of the bond (jl) were neglected in the above crude derivation. We therefore take the low-energy limit, $\Sigma_j \approx 2m_j - \frac{1}{2}\omega$, which is of main interest. Physically, the self-energy is mainly due to the orbital-flip excitation (see figure 1) which acquires a finite energy as strong correlations within the c-chains are explicitly taken into account.

We now use the notation $G_s = G_{\mathrm{AA}}$, $D_s = D_{\mathrm{AA}}$ if the index $s = 1$, and $G_s = G_{\mathrm{BA}}$, $D_s = D_{\mathrm{BA}}$ if $s = -1$. In the momentum space, equations (5) and (10) lead to

$$(s\omega - \lambda) G_s - \lambda \gamma_k G_{-s} + V_k D_{-s} = \delta_{s,1} \tag{11a}$$

$$[s\omega - \tau - \Sigma(s\omega)]\, D_s - \tau \gamma_\perp D_{-s} + V_k G_{-s} = 0. \tag{11b}$$

Here,

$$\Sigma(\omega) = 2 - \omega/2 \qquad \tau = 3/2$$

$$\gamma_k = (8c_z + \gamma_\perp)/9 \qquad \gamma_\perp = (c_x + c_y)/2$$

$$V_k = \sqrt{3}(c_x - c_y)/2 \qquad c_\alpha = \cos k_\alpha.$$

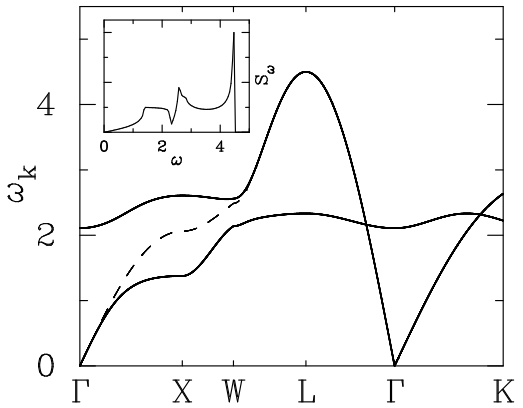

Figure 2. The transverse spin-excitation spectrum along the direction Γ–X$(\pi, 0, 0)$–W$(\pi, \pi/2, 0)$–L$(\pi/2, \pi/2, \pi/2)$–Γ–K$(3\pi/4, 3\pi/4, 0)$ in the Brillouin zone. To the right from L, the spin–orbit coupling vanishes and excitations are of either pure spin-flip or of simultaneous spin–orbital-flip character. Solid lines: present theory. Dashed line: spin-wave dispersion calculated with $J_c = 4$, $J_\perp = 1/4$ and neglecting orbital fluctuations. Inset: the on-site dynamic structure factor; the pseudogap seen about $\omega \approx 2$ is the manifestation of the orbital gap.

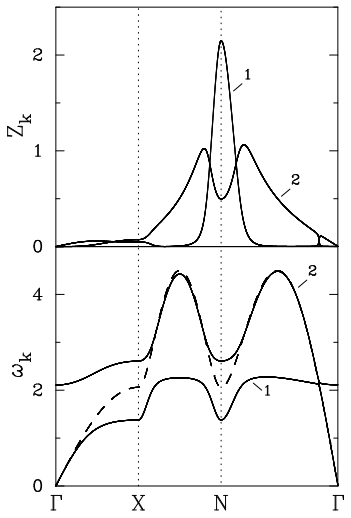

Figure 3. The transverse spin-excitation spectrum (lower panel) and the corresponding spectral weights in the dynamical structure factor (upper panel) along the Γ–X$(\pi, 0, 0)$–N$(\pi, 0, \pi)$–Γ direction. Dashed line: the spin-wave spectrum calculated neglecting spin–orbit coupling.

Equations (11*a*) and (11*b*) can easily be solved to give two branches $\omega_1(k)$ and $\omega_2(k)$ for the spin excitations and their relative weights in a dynamic spin structure factor. The results are shown in figures 2 and 3 for certain directions in the momentum space.

It should be noticed first that there is only one gapless mode at the Γ point. This is a plausible result since the original model has a continuous symmetry only in the spin subspace; the second branch related to the breaking of the discrete orbital symmetry is expected to have a finite gap. Our observation of only one gapless excitation in the spin response is consistent with reference [10], but deviates strongly from the finding of Feiner *et al* [8] of two gapless modes. It has recently been suggested [9] that the reason for the disagreement between the results of reference [8] and reference [10] could be found in the violation of the commutation relations for composite spin–orbital operators in reference [10]. However, the present paper employs the same method and the same commutation relations as Feiner *et al* [8, 9], but still arrives at the

same conclusions as in reference [10]. This clearly rules out the above reasoning. The present paper shows that the real origin of the controversy is in fact rather different: the quantum deviations from the Néel state are strong and must therefore be properly taken into account in the calculation of the orbital and composite excitations. In the approach employed here, the effects of quantum fluctuations are represented by the last term in equation (6), which has been discarded by Feiner *et al* [8,9]. Alternatively, the self-consistent diagrammatic method of reference [10] accounts for the underlying spin fluctuations through the deviations of the mean value of nearest-neighbour spin products from that of the classical Néel state. We conclude that the soft mode discussed in references [8, 9] is an artefact of the approximation which neglects the crucial effect of quantum spin fluctuations on the composite excitation $\sigma^x S^+$. With regard to the low-energy–momentum limit, model (1) behaves as a conventional Heisenberg system. However, the underlying anisotropic orbital ordering and the presence of orbital fluctuations strongly enhance quantum effects and reduce the order parameter. We find $\langle S^z \rangle = 0.23$ which is consistent with reference [10].

Second, at the magnetic zone boundaries the spin–orbit coupling results in quite visible deviations of the main magnon branch from that of an anisotropic Heisenberg model (see figures 2 and 3). This effect, which is due to the modulation of exchange bonds by orbital fluctuations, is expected to be a generic feature of spin–orbital models. In the present model, the zone-boundary effect is related to the particular momentum dependence of the coupling constant V_k in equations (11*a*) and (11*b*). Experimentally, an anomalous zone-boundary magnon softening has been observed in ferromagnetic manganites [12]. We have recently suggested [13] this effect to originate in the modulation of double-exchange bonds by orbital fluctuations.

Due to the finite hybridization of the excitations S^+ and $\sigma^x S^+$ (note that orbital pseudospin is not a conserved quantity), a conventional inelastic neutron scattering experiment might provide information on orbital excitation energies as well. In figure 3, we plot the relative spectral weight of two modes in the dynamical structure factor. A strong mixture of single-spin S^+ and $\sigma^x S^+$ excitations and the 'level repulsion' effect take place at energies where these excitations meet each other. In the present model, this results in an additional peak which has substantial weight at certain momenta.

One comment is necessary at this point: the present approach (as well as the one of references [8, 9]) implicitly assumes the formation of a bound state of orbital and spin excitations (a tightly bound composite excitation). However, it might well be possible that the bound state decays into an independent orbital wave and magnons. Instead of a well defined additional peak, one should then expect a softening and damping of magnons due to the coupling to the orbital–magnon excitation continuum. In fact, the latter picture is described in reference [10], which treats the spin–orbit coupling perturbatively. In some low-dimensional spin–orbital models the bound state might exist [14]. The problem of a bound state versus a particle–hole continuum in a Kugel–Khomskii model remains open at present. With regard to the low-energy behaviour, however, both approaches—the present one as well as that of reference [10]—are fully consistent with each other since the essence of the model—that is, the orbital-order-out-of-spin-disorder mechanism—is equally taken into account.

4. Conclusions

To summarize, we have studied a toy model providing a strong interplay between spin and orbital excitations. In the general context of transition metal oxides, this work suggests that the orbital fluctuations lead to a softening and damping of the zone-boundary magnons, and may even result in an additional structure in the spin-response function of these compounds.

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