J. Phys.: Condens. Matter **10** (1998) L555–L561. Printed in the UK PII: S0953-8984(98)94434-4

LETTER TO THE EDITOR

Quantum disorder versus order-out-of-disorder in the Kugel–Khomskii model

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Received 20 May 1998, in final form 29 June 1998

Abstract. The Kugel–Khomskii model, the simplest model for orbital degenerate magnetic insulators, exhibits a zero temperature degeneracy in the classical limit which could cause genuine quantum disorder. Khaliullin and Oudovenko (1997 *Phys. Rev.* B **56** R14243) suggested recently that instead a particular classical state could be stabilized by quantum fluctuations. Here we compare their approach with standard random phase approximation and show that it strongly underestimates the strength of the quantum fluctuations, shedding doubt on the survival of any classical state.

Motivated by developments in the manganites, interest in the role of orbital degeneracy in strongly correlated systems has been revived. A classic model in this context is the Kugel–Khomskii model [1], believed to be realistic [2] for $KCuF_3$ and related systems (one hole per site, degeneracy of the e_g orbitals). We recently discovered that this model poses a rather fundamental problem [3]: in the classical limit a point exists in the space of physical parameters where the ground state becomes infinitely degenerate due to a novel dynamical frustration mechanism. This classical degeneracy is lifted to the quantum level, and by analysing valence-bond type variational states we have arrived at the suggestion that the ground state for $S = 1/2$ might well be an incompressible spin fluid. In a follow up paper, Khaliullin and Oudovenko [4] suggested that the quantum fluctuations act to single out one particular classical state (the one with Néel order and $d_{3z^2-r^2}$ orbitals occupied by holes) over all others by an order-out-of disorder mechanism. The classical degeneracy is lifted by the differing strength of the fluctuations around the various classical states, but these fluctuations are not severe enough to destroy the classical Néel order completely. Their suggestion was based on a particular decoupling scheme and in this letter we will demonstrate that for rather simple reasons this decoupling scheme implies a serious underestimation of the strength of the fluctuations, shedding serious doubts on the possibility that classical order survives after all.

The Kugel–Khomskii model describes a three-dimensional (3D) cubic Mott–Hubbard insulator with a single hole/electron in e_g orbitals ($x^2-y^2 \sim |x\rangle$, $3z^2-1 \sim |z\rangle$), possessing, in the absence of virtual hoppings, orbital degeneracy in addition to the standard spin

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degeneracy. Its minimal version is given by ($J = t^2/U$ being the antiferromagnetic (AF) superexchange, with t the hopping element between $|z\rangle$ orbitals along the c-axis) [1, 3]

$$H = J \sum_{\langle ij \rangle} \left[4(\boldsymbol{S}_i \cdot \boldsymbol{S}_j) \left(\tau_i^\alpha - \tfrac{1}{2}\right) \left(\tau_j^\alpha - \tfrac{1}{2}\right) + \left(\tau_i^\alpha + \tfrac{1}{2}\right) \left(\tau_j^\alpha + \tfrac{1}{2}\right) - 1 \right] - E_z \sum_i \tau_i^c \tag{1}$$

where E_z is the energy splitting between the e_g orbitals, acting as a 'magnetic field' for the orbital pseudo-spins. It is used to investigate the system when it approaches the degeneracy point $E_z = 0$. The spin operators $\boldsymbol{S}_i$ are $S = 1/2$ spins, while the orbital degrees of freedom are described by (2×2) matrices in the pseudospin space

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

and α selects the cubic axis (a, b or c) that corresponds to the orientation of the bond $\langle ij \rangle$. The σ are Pauli matrices acting on the orbital pseudo-spins $|x\rangle = \begin{pmatrix} 1 \\ 0 \end{pmatrix}$, $|z\rangle = \begin{pmatrix} 0 \\ 1 \end{pmatrix}$. Hence, a Heisenberg model for the spins is coupled into an orbital problem. Here we ignore the (physically important) multiplet splittings due to a finite value of the atomic Hund's rule coupling (J_H), and focus on the special point E_z, $J_H \to 0$, contained in model equation (1): it is easy to see [3] that in the *classical* limit the system is dynamically frustrated and an infinite number of classical phases become degenerate at zero temperature. This degeneracy is lifted to the quantum level and one expects quantum effects to take over at this point, as well as in its direct vicinity [3], in analogy to what seems established in geometrically frustrated spin models [5]. If a disordered state would be stabilized by quantum effects, orbital degeneracy could be added to the list of mechanisms leading to a spin-liquid, such as the frustrated $J_1 - J_2$ Heisenberg antiferromagnet (HAF) [5], the bilayer HAF [6], and two-dimensional (2D) lattices with a reduced number of magnetic bonds, as realized in CaV_4O_9 [7].

Quite generally, the transverse modes [3, 4, 8] may be calculated starting from the equations of motion [9]

$$E \langle\langle \mathcal{S}_i^+ | ... \rangle\rangle = \frac{1}{2\pi} \langle [\mathcal{S}_i^+, ...] \rangle + \langle\langle [\mathcal{S}_i^+, H] | ... \rangle\rangle \tag{3}$$

$$E \langle\langle \mathcal{K}_i^+ | ... \rangle\rangle = \frac{1}{2\pi} \langle [\mathcal{K}_i^+, ...] \rangle + \langle\langle [\mathcal{K}_i^+, H] | ... \rangle\rangle \tag{4}$$

and using a generalization of the LSW theory. Here $\mathcal{S}_i^+$ is either S_i^+ or $\tilde{S}_i^+ \equiv S_i^+ \sigma_i^z$, while $\mathcal{K}_i^+$ is either $K_i^{++} \equiv S_i^+ \sigma_i^+$ or $K_i^{+-} \equiv S_i^+ \sigma_i^-$. The first pair of Green functions stands for *spin-wave* (SW) excitations, while the second pair describes mixed *spin-and-orbital-wave* (SOW) excitations. Similarly a longitudinal mode is given by

$$E \langle\langle \sigma_i^+ | ... \rangle\rangle = \frac{1}{2\pi} \langle [\sigma_i^+, ...] \rangle + \langle\langle [\sigma_i^+, H] | ... \rangle\rangle \tag{5}$$

where the Green function describes a *purely orbital* excitation. At each site the full set of local operators describing these excitations constitutes a so(4) Lie algebra. The spin-wave operators form a subalgebra, as seen from the familiar su(2) commutators together with the additional commutators

$$[S_i^+, S_i^z \sigma_i^z] = -\tilde{S}_i^+ \qquad [\tilde{S}_i^+, S_i^z \sigma_i^z] = -S_i^+ \tag{6}$$

while the same holds for the spin-and-orbital operators

$$[K_i^{+\pm}, S_i^z] = -K_i^{+\pm} \qquad [K_i^{+\pm}, K_i^{-\mp}] = 4 S_i^z \pm 2 \sigma_i^z. \tag{7}$$

However, for the calculation of the SW and SOW excitations one *also* needs commutators such as

$$[S_i^+, S_i^z\sigma_i^{\pm}] = -K_i^{+\pm} \qquad [K_i^{+\pm}, S_i^z\sigma_i^{\mp}] = -2S_i^+. \tag{8}$$

Clearly, *the SOWs cannot be separated from the SWs*, and one has to solve simultaneously equations (3) and (4).

The random-phase approximation (RPA) for spinlike operators linearizes the equations of motion by the familiar decoupling procedure [9]

$$\langle\langle \mathcal{A}_i\mathcal{B}_j|...\rangle\rangle \simeq \langle\mathcal{A}_i\rangle\langle\langle\mathcal{B}_j|...\rangle\rangle + \langle\mathcal{B}_j\rangle\langle\langle\mathcal{A}_i|...\rangle\rangle. \tag{9}$$

It is crucial that the decoupled operators $\mathcal{A}_i$ and $\mathcal{B}_j$ have *different* site indices, so that this procedure does not violate the local Lie-algebraic structure of the commutation rules (6)–(8). In the Néel-type AF phases with either $|x\rangle$ (AFxx) or $|z\rangle$ (AFzz) orbitals occupied, one now finds after Fourier transformation, and using the nonzero expectation values of S_j^z, σ_j^z and $S_j^z\sigma_j^z$ operators, two excitations (α = x, z for AFxx and AFzz, respectively)

$$[\omega_{\boldsymbol{k}}^{(n)}]^2 = \tfrac{1}{2}J^2\left(\lambda_\alpha^2 + \tau_\alpha^2 - Q_{\alpha\boldsymbol{k}}^2 - R_{\boldsymbol{k}}^2 - 2P_{\alpha\boldsymbol{k}}^2\right) \pm \tfrac{1}{2}J^2\left[(\lambda_\alpha^2 - \tau_\alpha^2)^2 - 2(\lambda_\alpha^2 - \tau_\alpha^2)(Q_{\alpha\boldsymbol{k}}^2 - R_{\boldsymbol{k}}^2)\right.$$
$$\left. - 4(\lambda_\alpha - \tau_\alpha)^2P_{\alpha\boldsymbol{k}}^2 + (Q_{\alpha\boldsymbol{k}}^2 + R_{\boldsymbol{k}}^2 + 2P_{\alpha\boldsymbol{k}}^2)^2 - 4(Q_{\alpha\boldsymbol{k}}R_{\boldsymbol{k}} - P_{\alpha\boldsymbol{k}}^2)^2\right]^{1/2}. \tag{10}$$

The orbital dependence enters the $\boldsymbol{k}$-independent field

$$\lambda_{\mathrm{x(z)}} = \tfrac{9}{2} \qquad \tau_{\mathrm{x(z)}} = \tfrac{3}{2} \pm \varepsilon_z \tag{11}$$

with $\varepsilon_z = E_z/J$, and the dispersion is given by

$$Q_{\mathrm{x}\boldsymbol{k}} = \tfrac{9}{2}\gamma_+(\boldsymbol{k}) \qquad Q_{\mathrm{z}\boldsymbol{k}} = \tfrac{1}{2}\gamma_+(\boldsymbol{k}) + 4\gamma_z(\boldsymbol{k}) \tag{12}$$

$$P_{\mathrm{x}\boldsymbol{k}} = \tfrac{3}{2}\sqrt{3}\gamma_-(\boldsymbol{k}) \qquad P_{\mathrm{z}\boldsymbol{k}} = \tfrac{1}{2}\sqrt{3}\gamma_-(\boldsymbol{k}) \tag{13}$$

$$R_{\boldsymbol{k}} = \tfrac{3}{2}\gamma_+(\boldsymbol{k}) \tag{14}$$

with $\gamma_\pm(\boldsymbol{k}) = \frac{1}{2}(\cos k_x \pm \cos k_y)$ and $\gamma_z(\boldsymbol{k}) = \cos k_z$.

The dispersions of SW and SOW are shown in figure 1. It is straightforward to verify that the SW dispersion is $9J/2$, given for the AFxx phase by the superexchange of $9J/4$ between $|x\rangle$ orbitals in the (a, b)-planes, and for the AFzz phase by strong interactions of $4J$ along the c-axis and weak superexchange of $J/4$ in the (a, b)-planes. In both phases one finds that the coupling between the modes due to the $P_{\alpha\boldsymbol{k}} \sim \gamma_-(\boldsymbol{k})$ term is strong, and the excitations have pure character only in the planes of $\gamma_-(\boldsymbol{k}) = 0$, as seen along Γ–$L(K)$ lines. In particular, this coupling increases along the Γ–X direction, and precisely compensates the dispersion due to the orbital dynamics $\sim \gamma_+(\boldsymbol{k})$. This results in a *soft mode* $\omega_{\boldsymbol{k}}^{(1)} = 0$ along the Γ–$X(Y)$ direction in both AF phases. As we have shown before [3], finite masses are found in the directions perpendicular to the soft mode lines, which gives a logarithmic divergence of the quantum correction to the order parameter, $\langle\delta S^z\rangle \sim \ln\Delta_i$, with $\Delta_i \to 0$ for $E_z \to 0$.

Khaliullin and Oudovenko [4] instead calculate a SW and a *longitudinal (i.e., purely orbital) excitation* $\langle\langle\sigma_i^+|...\rangle\rangle$ (5) first, and then include the effect of orbital fluctuations in the transverse channel (our SOW) selfconsistently in a perturbative way. The anomalous soft mode behaviour then does not occur and the AFzz phase is stable. While a selfconsistent calculation is, in principle, preferrable in an order-out-of-disorder problem, the particular approach of [4] violates the commutation relations in the Lie algebra (6)–(8), and only for this reason do the SW and SOW excitations become independent from each other. In the present RPA language it implies that composite spin-and-orbital operators, $S_i^\alpha\sigma_i^\beta$, are factorized into *independent products* of spin (S_i^α) and orbital (σ_i^β) operators separately, and

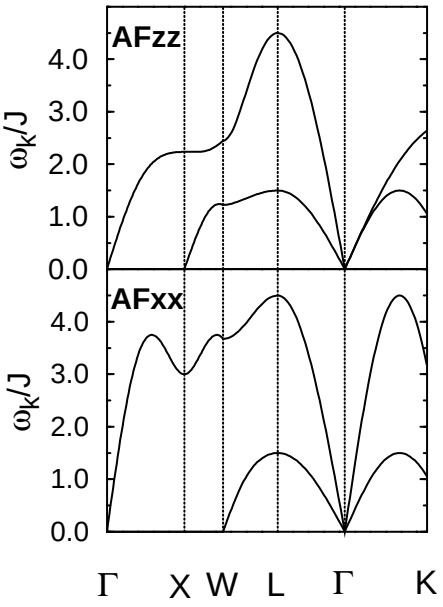

Figure 1. Transverse excitations ω_k/J for the Kugel–Khomskii model at orbital degeneracy ($E_z = 0$) within RPA for the AFzz (top) and AFxx (bottom) phases in the fcc (AFzz) Brillouin zone. Strong coupling between the (SW and SOW) modes results in a soft mode along the Γ–$X(Y)$ direction.

the commutators given by equations (6)–(8) effectively either vanish, e.g. $[S_i^+, S_i^z\sigma_i^\pm] \mapsto [S_i^+, S_i^z]\langle\sigma_i^\pm\rangle = 0$, or give a different result, e.g. $[S_i^+, S_i^z\sigma_i^z] \mapsto [S_i^+, S_i^z]\langle\sigma_i^z\rangle = -S_i^+\langle\sigma_i^z\rangle$. We call this procedure the SW+SOW scheme; it is formally equivalent to assuming $P_{\alpha k} = 0$ in equation (10). The SW modes now depend solely on the actual magnetic interactions, while the SOW modes are identical in the two phases and the soft mode behaviour is absent (figure 2). This indicates that in the approach of [4] the absence of the soft-mode behaviour is also not the consequence of selfconsistency, but rather the result of violating the commutation relations.

We further calculated the order parameter $\langle S^z\rangle$ in both AF phases including quantum corrections using a generalized RPA approach which leads to the identity

$$\langle S_i^z\rangle_{\mathrm{RPA}} = \tfrac{1}{2} - \langle S_i^- S_i^+\rangle - \tfrac{1}{2}\langle S_i^-\sigma_i^- S_i^+\sigma_i^+\rangle, \tag{15}$$

where $i \in A$, and A is the $\uparrow$-spin sublattice. The identity (15) follows from the expansion of the S_i^z operator in the so(4) algebra and replaces the su(2) relation $\langle S_i^z\rangle = \frac{1}{2} - \langle S_i^- S_i^+\rangle$, familiar from the Heisenberg model. It includes the renormalization due to *both* transverse modes in the spin-orbital model (1). Similarly the orbital occupancy is renormalized by $\langle\sigma_i^-\sigma_i^+\rangle$ fluctuations due to the longitudinal mode. The correlation functions are found from the respective Green functions [9],

$$\langle\mathcal{A}_i\mathcal{B}_i\rangle = \int_{-\infty}^{0} \mathrm{d}\omega \left(\frac{1}{N}\sum_{\boldsymbol{k}} 2\Im\langle\langle\mathcal{B}_{\boldsymbol{k}}|\mathcal{A}_{\boldsymbol{k}}\rangle\rangle_{\omega-i\epsilon}\right). \tag{16}$$

Equation (15) reproduces the result for the 2D HAF, $\langle S_i^z\rangle \simeq 0.303$, in the limit $E_z \to +\infty$, while $\langle S_i^z\rangle \simeq 0.251$ for the strongly anisotropic 3D HAF at $E_z \to -\infty$. The values of $\langle S_i^z\rangle$ are, however, strongly reduced when the degeneracy point ($E_z = 0$) is approached

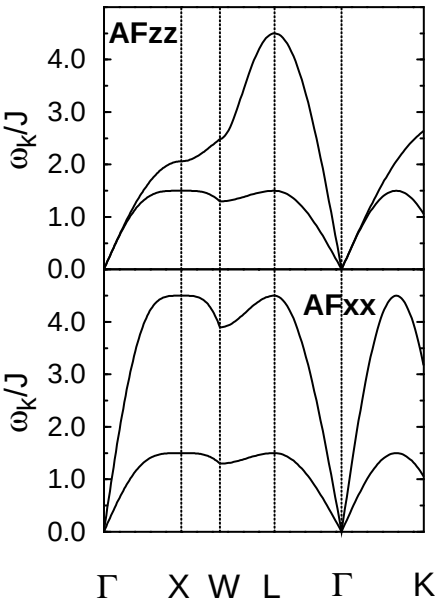

Figure 2. The same as in figure 1, but within the simplified SW+SOW scheme; the SOW dispersion is $1.5J$.

(figure 3), and the quantum corrections *overshoot* the mean-field value $\langle S_i^z \rangle_{\rm MF}$ for $-0.04 < E_z/J < 0.30$, and diverge at $E_z = 0$. In contrast, these corrections are much reduced within the SW+SOW scheme, and the divergence at E_z is removed ($\langle S_i^z \rangle \simeq 0.05$ in both phases). This is again qualitatively equivalent to the results of [4], where the renormalization of $\langle S_i^z \rangle$ due to the SOW was included only perturbatively, and a value 0.191 was found in the AFzz phase. This somewhat smaller quantum correction results from the finite gap in the orbital excitation.

Further evidence that the stability of the LRO phases is overestimated in [4] comes from energy calculations. For convenience we define the ground state energy per site as a quantum correction beyond the mean-field value

$$E = \frac{1}{N}\langle H \rangle + E_z \langle \tau_i^c \rangle + 3J. \tag{17}$$

A simple estimation at $E_z = 0$ using the Bethe ansatz solution for a disordered one-dimensional (1D) chain along the c-axis, and no magnetic correlations in the (a, b)-planes gives $E = -0.648J$ [3], while a somewhat better energy of $-0.656J$ was obtained using plaquette valence bond (PVB) states either with singlets alternating along the a- and b-axes in the (a, b)-planes (PVBA phase), or with single planes of such alternating singlets interlayered with two planes of singlets along the c-axis (PVBI phase), as explained in [3].

For the LRO phases, in spite of the divergent correction to the order parameter, an energy can still be estimated using the RPA corrections for the symmetry-broken state. Here these estimates, starting from the states with LRO, give lower energies than the above simple estimates for the VB states. This is not surprising, as it is known that improved VB wave functions that include the resonance between spin singlets lead typically to large energy gains, but are difficult to treat already in spin models [10]. First, within the generalized RPA approach one finds the largest quantum corrections in the AFxx phase (figure 4). This shows that the AFxx phase is 'more unstable' against disorder, in agreement with intuition

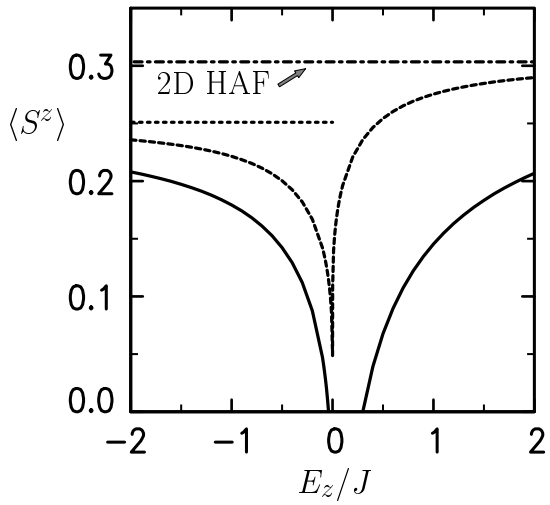

Figure 3. Order parameters $\langle S_i^z \rangle$ for AFzz (left) and AFxx (right) phases as functions of E_z/J using: the RPA (full lines) and the SW+SWO scheme (dashed lines). The horizontal lines show the limits found at $E_z/J \to -\infty$ (dashed line), and at $E_z/J \to \infty$ (2D HAF, dashed-dotted line).

and with [4]. We believe that the lowest energy $-0.896J$ obtained in the AFxx phase at $E_z = 0$ comes close to the true ground state. This is consistent with the experience with the 1D HAF, where one finds an energy of $-0.429J$ using the LSW theory, which is only 3.2% above the exact value $-0.443J$. We note that the energy obtained within the simplified SW+SOW approach is much higher, even above that of the disordered phases (PVB states). In contrast, the SW+SOW approach gives for the AFzz phase an energy somewhat lower than that of the PVB states, and our value of E differs only by $0.005J$ from that reported by Khaliullin and Oudovenko in their scheme (table 1). This indicates the qualitative similarity of these two approximations in treating the quantum fluctuations related to simultaneous spin and orbital flips (SOW excitations); in both cases the effect of such fluctuations is severely underestimated.

Table 1. Ground state energy E, in units of J, as obtained for the Kugel–Khomskii model using the full RPA and decoupled SW and SOW excitations, compared with the energy found in [4].

Method	AFzz phase	AFxx phase
RPA	−0.745	−0.896
SW+SOW	−0.685	−0.474
[4]	−0.690	—

Summarizing, the results presented in [4] are inconclusive, as their approximation violates the so(4) dynamical algebra describing the microscopic excitations. In contrast to the result of the perturbative treatment of [4], the RPA calculation yields an unstable AFzz (and AFxx) phase at orbital degeneracy, as also found in spin systems [5]. As the LSW theory performs quite well in simple spin systems with $S = 1/2$ [11], this strongly suggests that the ground state of the Kugel–Khomskii model is a spin-liquid. To our knowledge, the present case is unique in the sense that singlets arranged in a 3D valence bond solid (PVB

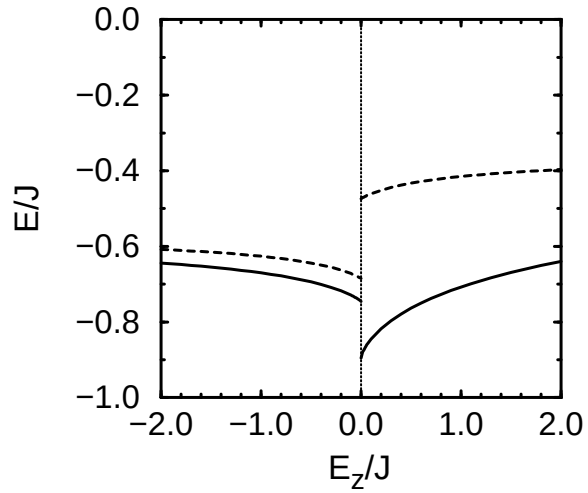

Figure 4. Ground state energies E of the AFzz (left) and AFxx (right) phases as functions of E_z/J, obtained using the RPA (full lines) and the SW+SWO scheme (dashed lines).

states) allow a lower energy than that of a classical state. However, it might well be that the final verdict on these matters has to wait for the systematic approach to the quantization of classically frustrated problems, which is still to be invented.

We acknowledge the support by the Committee of Scientific Research (KBN) of Poland, Project No. 2 P03B 175 14 (AMO), and by the Dutch Academy of Sciences (KNAW) (JZ).

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