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

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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 [Phys. Rev. B 56, R14 243 (1997)] 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 doubts on the survival of any classical state.

Motivated by the developments in the manganites, the interest in the role of orbital degeneracy in strongly correlated systems revived. A classic model in this context is the Kugel-Khomskii model \cite{1}, believed to be realistic \cite{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 \cite{3}: in the classical limit a point exists in the space of physical parameters where its ground state becomes infinitely degenerate, due to a novel dynamical frustration mechanism. This classical degeneracy is lifted on the quantum level, and by analyzing valence-bond type variational states we 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 \cite{4} suggested that instead the quantum fluctuations act to single out one particular classical state (the one with Néel order and $d_{x^2-y^2}$ 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 communication 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 ($x^2-y^2 \sim |x|$, $3z^2-1 \sim |z|$), possessing, in the absence of virtual hoppings, orbital degeneracy in addition to the standard spin 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) \cite{3,5},

\[
H = J \sum_{\langle ij \rangle} \left[4(\vec{S}_i \cdot \vec{S}_j)(\tau^x_i - \frac{1}{2})(\tau^x_j - \frac{1}{2})
+ (\tau^z_i + \frac{1}{2})(\tau^z_j + \frac{1}{2}) - 1 \right] - E_z \sum_i \tau^c_i,
\]

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 $\vec{S}_i$ are $S = 1/2$ spins, while the orbital degrees of freedom are described by ($2 \times 2$) matrices in the pseudospin space,

\[
\tau^a_{i}^{(b)} = \frac{1}{2}(-\sigma^z_i \pm \sqrt{3}\sigma^x_i), \quad \tau^c_i = \frac{1}{2}\sigma^z_i,
\]

and $\alpha$ selects the cubic axis ($a, b, or c$) that corresponds to the orientation of the bond ($ij$). The $\sigma$‘s are Pauli matrices acting on the orbital pseudo-spins $|x\rangle = \left(\begin{array}{c}1 \\ 0 \end{array}\right)$, $|z\rangle = \left(\begin{array}{c}0 \\ 1 \end{array}\right)$. 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 $E_z, J_H \rightarrow 0$, contained in model Eq. (1): it is easy to see \cite{3} that in the classical limit the system is dynamically frustrated and an infinity of classical phases become degenerate at zero temperature. This degeneracy is lifted on the quantum level and one expects quantum effects to take over at this point itself, as well as in its direct vicinity \cite{3}, in analogy to what seems established in geometrically frustrated spin models \cite{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) \cite{6}, the bilayer HAF \cite{7}, and two-dimensional (2D) lattices with a reduced number of magnetic bonds, as realized in CaV$_4$O$_9$ \cite{8}.
Quite generally, the transverse modes may be calculated starting from the equations of motion

\[ E(\langle \mathcal{S}_i^+ \rangle \ldots) = \frac{1}{2\pi} \langle [\mathcal{S}_i^+ \ldots] \rangle + \langle \{ [\mathcal{S}_i^+, H] ] \ldots \rangle, \tag{3} \]

\[ E(\langle \mathcal{K}_i^+ \rangle \ldots) = \frac{1}{2\pi} \langle [\mathcal{K}_i^+ \ldots] \rangle + \langle \{ [\mathcal{K}_i^+, H] ] \ldots \rangle, \tag{4} \]

and using a generalization of the LSW theory. Here \( \mathcal{S}_i^+ \) is either \( \mathcal{S}_i^+ \) or \( S_i^+ \), while \( \mathcal{K}_i^+ \) is either \( K_i^+ \) or \( K_i^\pm \). 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

\[ E(\langle \sigma_i^+ \rangle \ldots) = \frac{1}{2\pi} \langle [\sigma_i^+ \ldots] \rangle + \langle \{ [\sigma_i^+, H] ] \ldots \rangle, \tag{5} \]

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. Even though the spin-wave operators form a subalgebra, as seen from the familiar \( su(2) \) commutators together with the additional commutators

\[ [S_i^+, S_i^\pm \sigma_i^z] = -\hat{S}_i^\pm, \quad [\hat{S}_i^+, S_i^\pm \sigma_i^z] = -S_i^+, \tag{6} \]

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

\[ [K_i^\pm, S_i^z] = -K_i^\pm, \quad [K_i^\pm, K_i^\mp] = 4S_i^z \pm 2\sigma_i^z, \tag{7} \]

one needs with Hamiltonian for the calculation of the SW and SOW excitations also commutors like

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

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

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

\[ \langle A_i B_j \ldots \rangle \sim \langle A_i \rangle \langle B_j \rangle \ldots \langle B_j \rangle \langle A_i \rangle \ldots. \tag{9} \]

It is crucial that the decoupled operators \( A_i \) and \( B_j \) have different site indices, so that this procedure does not violate the local Lie-algebraic structure of the commutation rules. In the Néel-type AF phases with either \( \{ x \} \) (AFxx) or \( \{ z \} \) (AFzz) orbitals occupied, one now finds after Fourier transformation and using the nonzero expectation values of \( S_i^z, \sigma_i^z, \) and \( S_i^\pm \) operators, two excitations (\( \alpha = x, z \) for AFxx and AFzz, respectively),

\[ \omega_k = \frac{J}{2} \quad \lambda_n = \frac{9}{2} \quad \tau_n = \frac{9}{2} \pm \varepsilon_z, \tag{10} \]

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

\[ Q_{xz} = \frac{9}{2} \gamma_+ (\tilde{k}), \quad Q_{zk} = \frac{9}{2} \gamma_+ (\tilde{k}) + 4\gamma (\tilde{k}), \tag{12} \]

\[ P_{xk} = \frac{3}{2} \sqrt{3} \gamma^z (\tilde{k}), \quad P_{zk} = \frac{3}{2} \sqrt{3} \gamma^z (\tilde{k}), \tag{13} \]

\[ R_{k} = \frac{3}{2} \gamma_+ (\tilde{k}), \tag{14} \]

with \( \gamma_+ (\tilde{k}) = \frac{1}{2}(\cos k_x \pm \cos k_y) \) and \( \gamma_+ (\tilde{k}) = \cos k_z \).

The dispersions of SW and SOW are shown in Fig. It is straightforward to verify that the SW dispersion is \( 9J/2 \), given for the AFxx phase by the superexchange of \( 9J/4 \) between \( \{ x \} \) orbitals in the \( \{ a, b \} \)-planes, and for the AFzz phase by strong interactions of \( 4J \) along the \( c \)-axis and weak superexchange of \( 9J/4 \) in the \( \{ a, b \} \)-planes. In both phases one finds that the coupling between the modes due to the \( P_{xz} \sim \gamma_+ (\tilde{k}) \) term is strong, and the excitations have pure character only in the planes of \( \gamma_+ (\tilde{k}) = 0 \), as seen along \( \Gamma - L (K) \) lines. In particular, this coupling increases along the \( \Gamma - X \) direction, and precisely compensates the dispersion due to the orbital dynamics \( \sim \gamma_+ (\tilde{k}) \). This results in a soft mode \( \omega_k^{(1)} = 0 \) along the \( \Gamma - X (Y) \) direction in both AF phases. As we have shown before, 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, \( (\delta S^z) \sim \ln \Delta_i \), with \( \Delta_i \to 0 \) for \( E_z \to 0 \).

Khalilullin and Oudovenko calculate instead a SW and a longitudinal excitation \( \langle \sigma^+ \rangle \) first, and then include the effect of orbital fluctuations in the transverse channel (our SOW) in a perturbative way. Their self-consistent calculation gives a finite energy and weak dispersion of the orbital mode (in the present RPA approach the orbital excitation is found at \( \omega = 0 \)). This approach violates the commutation relations in the Lie algebra, and only for this reason the SW and SOW excitations become independent from each other. In the present RPA language it implies that composite spin-and-orbital operators, \( S_i^\alpha \sigma_i^\alpha \), are factorized into independent products of spin \( \sigma_i^\alpha \) and orbital \( \sigma_i^\alpha \) operators separately, and the commutators given by Eqs. effectively either vanish, e.g. \( [S_i^+ S_i^\pm \sigma_i^+] \to [S_i^+ S_i^\pm \sigma_i^+] \to 0 \), or give a different result, e.g. \( [S_i^+ S_i^\pm \sigma_i^+] \to [S_i^+ S_i^\pm \sigma_i^+] \to -S_i^+ \sigma_i^\pm \). We call this procedure the SW+SOW scheme; it is formally equivalent to assuming \( P_{ak} = 0 \) in Eq. (13). The SW modes depend now solely on the actual magnetic interactions, while the SOW modes are identical in the two phases and the soft mode behaviour is absent (Fig. 3). However, the SOW mode is gapless at the \( \Gamma \) point as a consequence of the rotational invariance of the classical
AF order when the occupied orbitals are rotated between $|x\rangle$ and $|z\rangle$.

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

$$\langle S_i^z \rangle_{\text{RPA}} = \frac{1}{2} - \langle S_i^- S_i^+ \rangle - \frac{1}{2} \langle S_i^- \sigma_i^z S_i^+ \sigma_i^z \rangle,$$

where $i \in A$, and $A$ is the $\uparrow$-spin sublattice. The identity (14) 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 $\langle \sigma_i^z \sigma_i^z \rangle$ fluctuations due to the longitudinal mode. The correlation functions are found from the respective Green functions (3).

$$\langle A_i B_i \rangle = \int_{-\infty}^{0} d\omega \left( \frac{1}{N} \sum_{\vec{k}} 2\Re \langle [B_{\vec{k}}^\dagger A_{\vec{k}}] \rangle_{\omega-i\epsilon} \right).$$

Eq. (15) reproduces the result for the 2D HAF, $\langle S_i^z \rangle \approx 0.303$, in the limit of $E_z \rightarrow +\infty$, while $\langle S_i^z \rangle \approx 0.251$ for the strongly anisotropic 3D HAF at $E_z \rightarrow -\infty$. The values of $\langle S_i^z \rangle$ are, however, strongly reduced when the degeneracy point ($E_z = 0$) is approached (Fig. 3), and the quantum corrections oversat the mean-field value $\langle S_i^z \rangle_{\text{MF}}$ 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 ($\langle S_i^z \rangle \approx 0.05$ in both phases). This is qualitatively equivalent to the results of Ref. 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 overstimated in Ref. 3 comes from energy calculations. For convenience we define the ground state energy per site as a quantum correction beyond the mean-field value,

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

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 $b$-axis in the $(a,b)$-planes (PVBA phase), or with single planes of such alternating singlets interlaided with two planes of singlets along the $c$-axis (PVBI phase), as explained in Ref. 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 (4). First, within the generalized RPA approach one finds the largest quantum corrections in the AFxx phase (Fig. 4). This shows that the AFx phase is ‘more unstable’ against disorder, in agreement with intuition and with Ref. 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 I]. 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.

Summarizing, the results presented in Ref. 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 Ref. 4, the RPA calculation yields an unstable AFzz (and AFxx) phase at orbital degeneracy, as also found in spin systems (3). 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 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.

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TABLE I. Ground state energy $E$, in units of $J$, as obtained for the Kugel-Khomskii model using the full RPA (RPA) and decoupled SW and SOW excitations (SW+SOW), compared with the energy found in Ref. [4].

| Method   | AFzz phase | AFxx phase |
|----------|------------|------------|
| RPA      | -0.745     | -0.896     |
| SW+SOW   | -0.685     | -0.474     |
| Ref. [4] | 0.690      | -          |

FIG. 1. Transverse excitations $\omega_k/J$ 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 $\Gamma - X(Y)$ direction.

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

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

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