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Unusual Symmetries in the Kugel-Khomskii Hamiltonian

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Abstract
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Comments
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Unusual Symmetries in the Kugel-Khomskii Hamiltonian

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The Kugel-Khomskii Hamiltonian for cubic titanates describes spin and orbital superexchange interactions between d^1 ions having threefold degenerate t_{2g} orbitals. Since orbitals do not couple along "inactive" axes, perpendicular to the orbital planes, the total number of electrons in $|\alpha\rangle$ orbitals in any such plane and the corresponding total spin are both conserved. A Mermin-Wagner construction shows that there is no long-range spin ordering at nonzero temperatures. Inclusion of spin-orbit coupling allows such ordering, but even then the excitation spectrum is gapless due to a continuous symmetry. Thus, the observed order and gap require more symmetry breaking terms.

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High temperature superconductivity [1] and colossal magnetoresistance [2] sparked much recent interest in the magnetic properties of transition metal oxides, particularly those with orbital degeneracy [3,4]. In many transition metal oxides, the d electrons are localized due to the large on-site Coulomb interaction U. Assuming a simple Hubbard model, with a typical nearest neighbor (NN) hopping energy t, the low-energy behavior can be described by an effective superexchange model, which involves only NN spin and orbital coupling, with energies of order $\epsilon = t^2/U$. In cubic oxide perovskites, the crystal field of the surrounding oxygen octahedra splits the d orbitals into a twofold degenerate e_g and a threefold degenerate t_{2g} manifold. In most cases, these degeneracies are further lifted by a cooperative Jahn-Teller (JT) distortion [3], and the low-energy physics is well described by an effective superexchange spin-only model [5–7]. However, some perovskites, such as $LaTiO_3$, have only a small JT distortion [8], in spite of the orbital degeneracy [9]. This distortion was not even observed in Refs. [10,11]. Since these distortions are small, several theoretical papers chose to ignore them and *assumed* cubic symmetry. The corresponding cubic model has been taken as the "minimal" model needed to explain the physics in these materials. In the present Letter, we show that, although this model is of great theoretical interest, it is insufficient to explain the experiments.

For the cubic titanates, there is one d electron in the t_{2g} degenerate manifold, which contains the wave functions $|X\rangle \equiv d_{yz}$, $|Y\rangle \equiv d_{xz}$, and $|Z\rangle \equiv d_{xy}$. The large degeneracy of the resulting ground states, which involve *both* the spin and the orbital degrees of freedom [3,4,12], may then yield rich phase diagrams, with exotic types of order, involving a strong interplay between the spin and orbital sectors (e.g., [4,10,11]), justifying the broad theoretical interest in this cubic limit. As we show, the corresponding superexchange Hamiltonian [hereafter called the cubic Kugel-Khomskii (KK) model [12]] contains several interesting hidden symmetries. In addition, our analysis shows that the KK Hamiltonian *cannot* yield some of the predictions which were claimed in the literature to follow from it. In particular, it has been suggested [13] that the KK Hamiltonian gives rise to an ordered isotropic spin phase at nonzero temperatures and that an energy gap in the spin excitations can be caused by spin-orbit interactions [14]. We use the symmetries of the KK Hamiltonian to show that both of these predictions cannot hold. The observed long-range order and finite gap [10] *must* therefore be based on more complicated Hamiltonians, which go beyond the scope of this Letter.

As can be seen from Fig. 1, cubic symmetry implies no hopping (via oxygen p states) among orbitals of type $|\alpha\rangle$ along the α axis when the direct Ti-Ti hopping is neglected. This axis has been called the "inactive axis" for α orbitals. This statement forms the basis for the remarkable symmetry properties of the KK Hamiltonian reported in this Letter. Apart from constant terms, the perturbative expansion of the Hubbard Hamiltonian with hopping of this type, to order ϵ, yields the cubic KK Hamiltonian, $\mathcal{H} = \mathcal{H}_x + \mathcal{H}_y + \mathcal{H}_z$, where

$$\mathcal{H}_\alpha = \epsilon \sum_{\langle ij\rangle \in \alpha} \sum_{\beta,\gamma \neq \alpha} \sum_{\sigma,\eta} c^\dagger_{i,\beta,\sigma} c_{i,\gamma,\eta} c^\dagger_{j,\gamma,\eta} c_{j,\beta,\sigma}, \quad (1)$$

and $\langle ij\rangle \in \alpha$ denotes a NN bond along the α axis. Here $c^\dagger_{i,\beta,\sigma}$ creates an electron at site i in a β orbital with spin

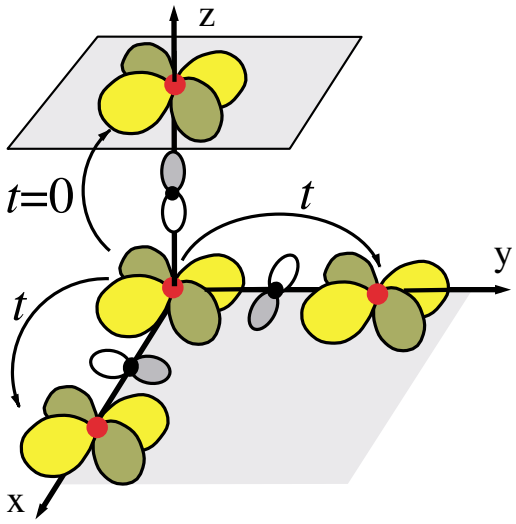

FIG. 1 (color online). A schematic view of the $|Z\rangle = d_{xy}$ orbitals and the (indirect) hopping parameter t via intermediate oxygen p orbitals. Positive (negative) regions of wave functions are represented by dark (light) lobes. Symmetry forbids indirect hopping along the z axis for an electron in the Z orbital.

σ, and one assumes that there is exactly one electron on each site, i.e., $\sum_\alpha n_{i\alpha} = 1$, with $n_{i\alpha} \equiv \sum_\sigma c^\dagger_{i,\alpha,\sigma} c_{i,\alpha,\sigma}$.

For some purposes, it is convenient to separate the spin and orbital degrees of freedom [12]. Defining the spin of an electron at site i as $\mathbf{S}_i$, one has

$$\mathcal{H}_\alpha = (\epsilon/2) \sum_{\langle ij\rangle \in \alpha} (1 + 4\mathbf{S}_i \cdot \mathbf{S}_j) J^\alpha_{ij}, \tag{2}$$

$$J^z_{ij} = n_{ix} n_{jx} + n_{iy} n_{jy} + a^\dagger_i b_i b^\dagger_j a_j + b^\dagger_i a_i a^\dagger_j b_j. \tag{3}$$

Here $a^\dagger_i$ and $b^\dagger_i$ create *spinless* electrons in orbitals $|X\rangle$ and $|Y\rangle$, respectively, and $n_{ix} = a^\dagger_i a_i$, etc.

Both Eqs. (1) and (3) imply that whenever an α-orbital electron is destroyed, an α-orbital electron is created on either the same or another site. Therefore, the total number of electrons in each orbital is a good quantum number: Any eigenfunction can be labeled by the total number of electrons *in each orbital* (i.e., N_X, N_Y, and N_Z) [15]. Furthermore, when an α-orbital electron is destroyed, it is replaced by another α-orbital electron *in the same plane perpendicular to the inactive (α) axis*. Thus, for the nth plane perpendicular to the α axis, the total number $N_{n\alpha}$ of electrons in orbital $|\alpha\rangle$ is conserved, i.e., it is a good quantum number. For example, for a cube of eight sites, the numbers N_{1X} and N_{2X}, which, respectively, are the numbers of X-orbital electrons in each of the two planes perpendicular to the x axis, are conserved, and similarly for y and z. Thus, the states of the cube can be labeled by the six quantum numbers $(N_{1X}, N_{2X}, N_{1Y}, N_{2Y}, N_{1Z}, N_{2Z})$.

Remarkably, there are more conserved quantities, associated with electron spins. Defining the spin of an electron in orbital α at site i as $\mathbf{S}_{i\alpha} \equiv \sum_{\rho\eta} c^\dagger_{i,\alpha,\rho} [\vec{\boldsymbol{\sigma}}]_{\rho\eta} c_{i,\alpha,\eta}/2$, where $\vec{\boldsymbol{\sigma}}$ represents the vector of Pauli matrices, and the total spin of all such electrons — located in an arbitrarily chosen plane n perpendicular to the inactive α axis — as $\vec{S}_{n\alpha} \equiv \sum_{i\in n} \mathbf{S}_{i\alpha}$, we next perform a uniform, but arbitrary, rotation of all the $\mathbf{S}_{i\alpha}$'s with $i \in n$: We introduce an arbitrary 2×2 unitary matrix $\mathbf{U^{(n)}}$ and write

$$c^\dagger_{i,\alpha,\sigma} = \sum_\eta U^{(n)}_{\sigma,\eta} d^\dagger_{i,\alpha,\eta}, \qquad i \in n. \tag{4}$$

Electrons in other orbitals or in other lattice planes are not affected by this transformation. Substitution of this transformation into Eq. (1) shows that it leaves $\mathcal{H}$ invariant. As a consequence of this symmetry, if one assumes long-range spin order, the spins associated with α orbitals within any given plane can be rotated at zero cost in energy, thereby destroying the supposed long-range order. It also follows that $\vec{S}_{n\alpha}$ commutes with $\mathcal{H}$, and, thus, both $|\vec{S}_{n\alpha}|^2$ and $[\vec{S}_{n\alpha}]_z$ are good quantum numbers for each value of n or α. These symmetries can also be obtained from the original Hubbard model, provided one neglects Coulomb exchange interactions.

This situation allows a *rigorous proof* of the nonexistence of long-range spin order at any nonzero temperature for $\mathcal{H}$ of Eq. (1). Following the procedure of Mermin and Wagner (MW) [16], we choose

$$\begin{aligned} C &= \hat{S}^+_\alpha(\mathbf{k}) = \sum_{\mathbf{R}} e^{-i\mathbf{k}\cdot\mathbf{R}} c^\dagger_{\mathbf{R},\alpha,\uparrow} c_{\mathbf{R},\alpha,\downarrow}, \\ A &= \hat{S}^-_\alpha(\mathbf{k}+\mathbf{K}) = \sum_{\mathbf{R}} e^{i(\mathbf{k}+\mathbf{K})\cdot\mathbf{R}} c^\dagger_{\mathbf{R},\alpha,\downarrow} c_{\mathbf{R},\alpha,\uparrow}, \end{aligned} \tag{5}$$

where $\mathbf{K}$ is the wave vector of the order we wish to discuss. Here $a\mathbf{K} = (\pi, \pi, \pi)$ is most relevant. Assuming long-range order of $\hat{S}^z_\alpha(\mathbf{K}) \equiv s_{\alpha,z}$ and a corresponding staggered field h (for spins in the α orbital), we end up with the MW-like bound

$$1 \geq 2kT |s_{\alpha,z}|^2 \frac{1}{N} \sum_{\mathbf{k}} [h s_{\alpha,z} + \hat{J}_\alpha(\mathbf{k})]^{-1}, \tag{6}$$

where $\hat{J}_\alpha(\mathbf{k}) \propto \sum_{\vec{\delta}\notin\alpha} (1 - e^{-i\mathbf{k}\cdot\vec{\delta}})$ is proportional to the $\mathbf{k}$-dependent parts in the Fourier transform of the nonzero NN spin-spin interaction (with NN vector $\vec{\delta}$) in the Hamiltonian. Since spins in orbital α couple only within planes perpendicular to the α axis, it follows that $\hat{J}_\alpha(\mathbf{k}) \propto 2\sum_{\beta\neq\alpha}[1 - \cos(k_\beta a)] \approx \sum_{\beta\neq\alpha} a^2 k^2_\beta \equiv a^2 k^2_{\perp,\alpha}$, with *no dispersion in the α direction*. For systems in $d \leq 3$ dimensions, the sum in Eq. (6) diverges as $h \to 0$, implying that $s_{\alpha,z}$ must go to zero. The conclusion is that the KK model is at its lower critical dimension $d_< = 3$ and does not support long-range spin order at $T > 0$. As we show elsewhere, a similar proof can be formulated for the original Hubbard model [17].

The same conclusion also follows from a renormalization group analysis of the model at finite T [17].

Generalizing to m orbitals and n-component spins, the spin free energy functional maps onto that of the "canonical" nm-component spin problem [18], but with a $(d-1)$-dimensional transverse gradient term,

$$F = \frac{1}{2}\sum_{\mathbf{q}\alpha}(r + q_{\perp,\alpha}^2)\hat{\mathbf{S}}_\alpha(\mathbf{q})\cdot\hat{\mathbf{S}}_\alpha(-\mathbf{q}) + \sum_{\mathbf{R}}\Big[u\sum_\alpha |\hat{\mathbf{S}}_\alpha(\mathbf{R})|^4 + v\sum_{\alpha<\beta}|\hat{\mathbf{S}}_\alpha(\mathbf{R})|^2|\hat{\mathbf{S}}_\beta(\mathbf{R})|^2\Big], \quad (7)$$

where $\hat{\mathbf{S}}_\alpha(\mathbf{q})$ is the Fourier transform of $\mathbf{S}_\alpha(\mathbf{R})$. Similar forms arise in connection with Lifshitz-like behavior [19]. This anisotropic gradient term shifts *both* the upper and the lower critical dimensions up by 1. For $3 < d < 5$ dimensions and $n > 1$ it yields decoupled n-component critical behavior. The free energy (7) also reflects the symmetry with respect to independent rotations of the spin $\mathbf{S}_\alpha$ associated with the single orbital α.

These symmetries and conservation laws are very useful in the exact numerical diagonalization of finite Ti clusters, which indeed confirms their validity. We demonstrate this for a cube of eight sites (Fig. 2). Since the Hamiltonian commutes with the total spin $\mathbf{S} = \sum_i \mathbf{S}_i$, the eigenstates can be identified by the quantum numbers S [where $\mathbf{S}^2 = S(S+1)$] and S_z. Since the energy does not depend on S_z, it suffices to study the subspace of $70 \times 3^8 = 459\,270$ states with $S_z = 0$. A numerical analysis of the low-energy spectrum of this huge sparse matrix yielded three degenerate $S = 0$ ground states, Ψ_x, Ψ_y, and Ψ_z, related by cyclic permutations. Ψ_z has $N_x = N_y = 4$. This information suffices to find the manifold containing each of these ground states. Since Ψ_z, for instance, is not degenerate within the manifold $N_x = N_y = 4$, it must also have the quantum numbers $N_{1X} = N_{2X} = 2$, $N_{1Y} = N_{2Y} = 2$, and $N_{1Z} = N_{2Z} = 0$. A nonsymmetric choice such as $N_{1X} = 3$ and $N_{2X} = 1$ would be degenerate with $N_{1X} = 1, N_{2X} = 3$. The lack of degeneracy also implies that the total spin of the $N_{n,\alpha} = 2$ electrons in orbital α in the nth plane perpendicular to the α axis must be $S_{n\alpha} = 0$. Examples of such configurations, containing dimers of α-orbital electrons in the α planes, are shown in Fig. 2. The Hamiltonian allows an exchange of an X electron with a Y electron only along the z axis (the only axis along which *both* types can hop). Starting from Fig. 2(a), and performing all such possible exchanges, creates a manifold of 16 states (three of which are shown in the figure). Two other states with the same $N_{n\alpha}$'s, but with the dimers along the z axis, form another manifold, of higher energy. Indeed, a diagonalization of the resulting 16×16 matrix reproduced the same ground state energy as found from the $459\,270 \times 459\,270$ matrix, demonstrating the power and the correctness of these symmetries. We are currently extending these numerical studies to even larger systems (such as $N = 16$ sites) to better understand the nature of the ground state in a real system.

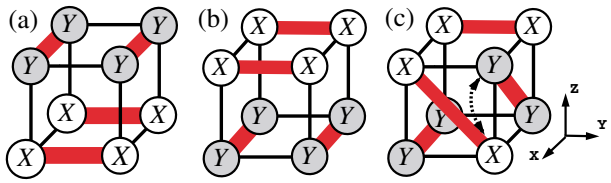

FIG. 2 (color online). Spin and orbital configurations for a cube of eight sites. The thick lines indicate singlet spin states (dimers) and the X and Y indicate the orbital states of the electrons. Configurations in (a) and (b) are the dominant ones in the ground state wave function. The less dominant configuration (c) is obtained from (b) by allowing the interchange of two (X and Y) electrons along the z axis, retaining their membership in the spin singlets (even though their positions have changed).

Since the KK Hamiltonian (1) forbids long-range spin order at $T > 0$, the existence of such order (as in $LaTiO_3$ [10]) must result from some additional mechanism. Even for cubic symmetry, such mechanisms could include the small direct Ti-Ti hopping along the inactive axis, Coulomb exchange terms in the original Hubbard model, or the spin-orbit interaction. In the real orthorhombic titanates one must also include JT distortions and oxygen octahedra rotations. A full discussion of all these effects lies beyond the present Letter. Since the present Letter concerns mainly symmetry arguments, we concentrate here on adding the spin-orbit interaction to the cubic KK Hamiltonian, where we can use such arguments to show the absence of a spin gap. Specifically, the spin-orbit Hamiltonian is

$$\mathcal{H}_{\mathrm{SO}} = \lambda \sum_{i\alpha\beta\sigma\sigma'\mu} L^\mu_{\alpha\beta} c^\dagger_{i,\alpha,\sigma}[\boldsymbol{\sigma}_\mu]_{\sigma\sigma'} c_{i,\beta,\sigma'}, \quad (8)$$

where $L^\mu_{\alpha\beta} \equiv \langle\alpha|L^\mu|\beta\rangle$ is the orbital angular momentum matrix element. Since $\mathcal{H}_{\mathrm{SO}}$ mixes orbitals, an α electron can hop via $\mathcal{H}_{\mathrm{SO}}$ to orbital β, then hop to a β orbital on a NN along the α axis, and finally use $\mathcal{H}_{\mathrm{SO}}$ to return to orbital α. This generates an effective hopping between α orbitals along the inactive α axis, invalidating the above arguments, shifting the lower critical dimension for total spin ordering back to $d_< = 2$, and restoring long-range spin order at $d = 3$. This mixing also eliminates the independent symmetries, which we found for electrons within each orbital separately. However, as discussed below, there still remain some global symmetries for the total spin. Based on the signs of the leading couplings, we assume that the total spin orders antiferromagnetically and proceed to show that the spin wave excitations in the ordered phase *must be gapless*.

Again, an exact symmetry analysis clarifies the situation. For electrons within each of the three degenerate t_{2g} orbitals discussed here, we introduce the following

canonical transformation [6] from spin to pseudospin:

$$c^{\dagger}_{i,\alpha,\sigma} = \sum_{\eta} \mathbf{U}^{(\alpha)}_{\sigma,\eta} d^{\dagger}_{i,\alpha,\eta}, \tag{9}$$

where $U^{\alpha} = \boldsymbol{\sigma}_{\alpha}$ represents a different rotation for spins in different orbitals. As discussed in Ref. [6], all the terms in both the original $\mathcal{H}$ and in $\mathcal{H}_{\mathrm{SO}}$ now contain only combined operators of the form $\sum_{\mu} d^{\dagger}_{i,\alpha,\mu} d_{i,\beta,\mu}$, with coefficients which do not depend on the pseudospin indices μ [see Eq. (6) in Ref. [6]]. These combined operators, and therefore also the full Hamiltonian, are *rotationally invariant in pseudospin space*. Said differently, the Hamiltonian is invariant with respect to a transformation on the original spins of the form $\tilde{c}^{\dagger}_{i,\alpha,\mu} = \sum_{\mu'} V^{(\alpha)}_{\mu,\mu'} c^{\dagger}_{i,\alpha,\mu'}$ with $\mathbf{V}^{(\alpha)} = \boldsymbol{\sigma}_{\alpha} \mathbf{U} \boldsymbol{\sigma}_{\alpha}$, where $\mathbf{U}$ is an arbitrary unitary matrix. Thus, the system possesses a continuous symmetry, but it is not the usual symmetry with respect to rotation of the total spin. In the antiferromagnetically ordered phase, the spin staggered moment selects an orientation, and therefore the pseudospin will also exhibit broken symmetry. Rotation of the pseudospin will give rise to a manifold of zero-energy states. This continuous symmetry guarantees that we have a (probably propagating) zero-energy hydrodynamic mode [20]. The rigorous conclusion is then that spin-orbit interaction permits the existence of long-range order at nonzero temperatures, but does not cause a gap in the elementary excitation spectrum, contrary to the assertion in Ref. [14]. Since our argument is based on symmetry considerations, it holds no matter what type of fluctuation is considered and regardless of the orbital ordering (long ranged or liquid). In analogy with results of Refs. [6,7,21], it is probable that when Coulomb exchange interactions and/or canting of the Ti–O–Ti bonds are included, spin-orbit interactions would lead to an energy gap in the excitation spectrum.

In conclusion, we uncovered several novel symmetries of the KK Hamiltonian for cubic t_{2g} systems. It is surprising that the KK Hamiltonian has been widely used in the study of interesting spin-orbital physics of transition metal oxides for a long time, yet its remarkable symmetry properties were missed until now. Using these symmetries, we rigorously showed that the KK Hamiltonian without spin-orbit interactions does not permit the development of long-range spin order in a three-dimensional cubic lattice at nonzero temperature. Inclusion of spin-orbit interactions allows the formation of long-range spin order, but the excitation spectrum is gapless.

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