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Orbital Ordering in Frustrated Jahn-Teller Systems with 90° Exchange

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We show that superexchange interactions in frustrated Jahn-Teller systems with transition metal ions connected by the 90° metal-oxygen-metal bonds (e.g., NaNiO_2 , LiNiO_2 , and ZnMn_2O_4) are much different from those in materials with the 180° bonds. In the 90°-exchange systems spins and orbitals are decoupled: the spin exchange is much weaker than the orbital one and it is ferromagnetic for all orbital states. Though the mean-field orbital ground state is strongly degenerate, quantum orbital fluctuations select particular ferro-orbital states. We explain the orbital and magnetic ordering observed in NaNiO_2 and show that LiNiO_2 is not a spin-orbital liquid.

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There is a large class of compounds containing transition metal (TM) ions with an orbital degeneracy—the so-called Jahn-Teller (JT) systems. Orbital degrees of freedom and, in particular, their ordering give rise to a very rich physics [1,2]. The interplay between orbitals and spins often leads to rather peculiar magnetic structures, turning, e.g., cubic perovskites such as KCuF_3 into quasi-one-dimensional spin-chain materials [1]. It was recently suggested that in some JT systems orbitals remain disordered at all temperatures, forming an orbital-liquid state [3–5]. The suppression of an orbital ordering was discussed for materials with a simple cubic structure, e.g., the perovskite LaTiO_3 , and was attributed to a strong anisotropy of orbital interactions and a strong coupling between orbital and spin excitations. One would expect to find an even stronger tendency to form an orbital-liquid in JT compounds with an additional “geometric frustration” [6], which is usually associated with the triangle-based lattices, such as the pyrochlore lattice of corner sharing tetrahedra formed by TM ions in spinels (e.g., ZnMn_2O_4) and the triangular lattice of low-spin Ni^{3+} ions in the layered materials LiNiO_2 and NaNiO_2 . Orbital ordering in such systems is an open issue interesting from both theoretical and practical points of view, as some of these compounds are now studied as promising materials for rechargeable batteries and their performance may be significantly affected by the JT effect.

An important difference between the geometrically frustrated oxides and perovskites is the angle between the oxygen-metal bonds connecting two neighboring TM ions. While in perovskites this angle is close to 180°, in the layered materials with the triangular lattice and spinels it is 90°. Though usually ignored, this difference has important consequences for orbital and magnetic orderings. In this Letter, we derive the Hamiltonian of the 90° exchange and show that orbitals and spins are in that case essentially decoupled (in contrast to the strong coupling in perovskites) and that orbital and magnetic orderings can be considered separately from each other. For layered materials with a triangular lattice, we find a

large number of disordered mean-field orbital ground states. This strong frustration is, however, lifted by quantum orbital fluctuations that stabilize a ferro-orbital state. Furthermore, we show that spins in the layers order ferromagnetically. Our theory explains the orbital and magnetic ordering observed in NaNiO_2 [7] and implies that LiNiO_2 , in which no long-range order is observed [8,9], is unlikely a “spin-orbital liquid.”

Orbitals in systems with one electron or hole on doubly degenerate e_g levels are conveniently described by introducing isospins (or pseudospins) \mathbf{T}_j on each TM site j , which act on the up and down states $|T^z = \pm \frac{1}{2}\rangle$ identified with, respectively, $d_{3z^2-r^2}$ and $d_{x^2-y^2}$ orbitals. Quite generally, an effective interaction between the orbitals and spins on pairs of neighboring TM sites i and j has the form [1]

$$H_{ST} = \sum_{ij} [J_S(\mathbf{S}_i\mathbf{S}_j) + J_T T_i T_j + J_{ST}(\mathbf{S}_i\mathbf{S}_j)T_i T_j], \quad (1)$$

(for simplicity we do not show here the full structure of the orbital terms). In perovskites with the 180° bonds the spin- and orbital-exchange constants J_S and J_T , as well as the coupling between the orbitals and spins J_{ST} , are all of the same order of magnitude. This makes the spin and orbital orderings dependent on each other [1]. The strong interplay between orbitals and spins is also crucial for the mechanisms of suppression of these orderings used in Refs. [4,10,11] to explain an apparent absence of both orbital and magnetic ordering in LiNiO_2 [8,9]. In particular, in the SU(4) version of the model Eq. (1) [10,11] both the spin and orbital interactions are isotropic and $J_S = J_T = \frac{1}{4}J_{ST}$. The actual situation in the 90° systems is, however, quite different: the orbital exchange is strongly anisotropic and $J_S, J_{ST} \ll J_T$.

We derive the exchange Hamiltonian using as an example the layered material NaNiO_2 , in which Ni and Na ions occupy consecutive [111] planes of the NaCl structure [see Fig. 1(a)]. In each plane the low-spin Ni^{3+} ions with one electron on the doubly degenerate e_g level form a triangular lattice. The exchange between two

nearest-neighbor Ni ions in the planes occurs via common oxygens and the angle between the two oxygen p orbitals involved in this exchange is 90° . The exchange between Ni ions from two neighboring planes (separated by a plane of Na ions) is weak and is neglected here.

The electron hopping in the z direction can occur only between the p_z oxygen and the $d_{3z^2-r^2}$ nickel orbitals. The corresponding hopping amplitude is denoted by t . All other processes are forbidden by symmetries of the oxygen p and nickel e_g orbitals. Thus, the hopping amplitude in the z direction is t for the $|T^z = +\frac{1}{2}\rangle$ nickel state, and 0 for the $|T^z = -\frac{1}{2}\rangle$ state. More generally, the hopping in the α direction ($\alpha = x, y, z$) is nonzero for only the oxygen p_α and the nickel $d_{3\alpha^2-r^2}$ orbitals. It is convenient to introduce the isospin operators $I^\alpha = \mathbf{T} \cdot \mathbf{n}^\alpha$, where \mathbf{n}^α are three unit vectors in the (T^x, T^z) plane

$$\mathbf{n}^x = -\frac{1}{2}\hat{\mathbf{z}} - \frac{\sqrt{3}}{2}\hat{\mathbf{x}}, \quad \mathbf{n}^y = -\frac{1}{2}\hat{\mathbf{z}} + \frac{\sqrt{3}}{2}\hat{\mathbf{x}}, \quad \mathbf{n}^z = \hat{\mathbf{z}}. \quad (2)$$

The operators I^x and I^y play the role of T^z for the x and y directions, i.e., they describe the occupation of, respectively, the $d_{3x^2-r^2}$ and $d_{3y^2-r^2}$ orbital. The hopping amplitude in the α direction is then t for the $|I^\alpha = +\frac{1}{2}\rangle$ state and 0 for the $|I^\alpha = -\frac{1}{2}\rangle$ state.

Figure 1(b) shows a plaquette in the $\alpha\beta$ plane ($\alpha, \beta = x, y, z$) formed by the two neighboring Ni sites 1 and 2 and the two oxygens shared by the oxygen octahedra surrounding the TM ions. The exchange Hamiltonian is obtained by expanding the energy in powers of the hopping amplitude t , which is assumed to be much smaller than the energy Δ of the electron transfer from O to Ni and the Hubbard repulsion on oxygen U_p . The exchange interaction appears in the fourth order of the expansion, and the higher-order terms are neglected here.

We first neglect also the Hund's rule coupling between the spins of the oxygen holes. In that case, the 90° exchange is independent of a spin configuration and involves only orbitals. The orbital-exchange Hamiltonian for the plaquette in the $\alpha\beta$ plane has the form

$$H_T = J_T I^{\alpha\beta} + \text{const}, \quad (3)$$

where

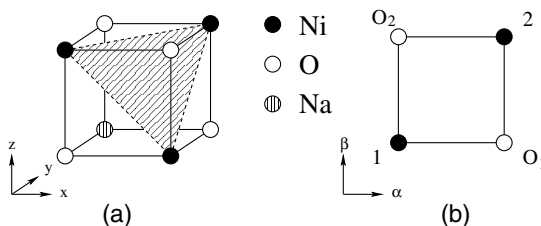


FIG. 1. The crystal structure of NaNiO₂ (a) and a plaquette in the $\alpha\beta$ plane ($\alpha, \beta = x, y, z$) formed by two nearest-neighbor Ni ions, 1 and 2, and two oxygens, O₁ and O₂ (b).

$$I^{\alpha\beta} = (\frac{3}{2} - I_1^\alpha)(\frac{3}{2} - I_2^\beta) + (\frac{3}{2} - I_1^\beta)(\frac{3}{2} - I_2^\alpha), \quad (4)$$

and J_T is the coupling constant specified below. The factor $\frac{3}{2} - I_1^\alpha$ counts the number of oxygen electrons that can hop to the Ni site 1 along the bond in the α direction. This number equals 1 if the e_g electron is in the state $|I_1^\alpha = +\frac{1}{2}\rangle$, and 2 if the electron occupies the state $|I_1^\alpha = -\frac{1}{2}\rangle$. [For one hole on e_g level, as in Cu²⁺, the exchange interaction corresponding to Eq. (4) involves combinations $(\frac{1}{2} + I^\alpha)$ instead of $(\frac{3}{2} - I^\alpha)$.] The two terms in Eq. (4) correspond to the two exchange paths on the plaquette through the oxygens O₁ and O₂. The orbital coupling

$$J_T = -\frac{4t^2}{\Delta^2(2\Delta + U_p)} + \frac{2t^4}{\Delta^3} = \frac{2t^4 U_p}{\Delta^3(2\Delta + U_p)} \quad (5)$$

is the sum of the contributions of two different mechanisms of the orbital exchange. The first term in Eq. (5) is due to the exchange via the intermediate states with two holes on one of the oxygens from the plaquette. The mechanism resulting in the second term in Eq. (5) even works for infinite U_p , when the two-hole oxygen states are forbidden. To understand its origin, consider first a single Ni site inside the oxygen octahedron. The hopping of electrons from the six oxygens to nickel results in the energy decrease δE . For two neighboring Ni sites, this decrease is smaller than $2\delta E$, as the surrounding octahedra share two oxygens and two-hole oxygen states are forbidden for infinite U_p . The effect of the blocking of the two-hole states on common oxygens depends on orbital states of the e_g electrons, which gives rise to the orbital exchange with the coupling constant $+(2t^4/\Delta^3)$.

The intermediate states with two holes on one oxygen result also in a spin exchange. As in such states the holes occupy two mutually orthogonal orbitals p_α and p_β , the spin exchange is possible due only to the Hund's rule coupling J_H between the spins of the holes (in systems with the 180° bonds, the spin exchange occurs even for $J_H = 0$). The corresponding exchange Hamiltonian involves both spin and isospin operators and, to the lowest-order in powers of J_H , has the form

$$H_{TS} = -J_{TS} I^{\alpha\beta} [\frac{3}{4} + (\mathbf{S}_1 \mathbf{S}_2)], \quad (6)$$

where $J_{TS} = 4t^4 J_H / [\Delta^2(2\Delta + U_p)^2]$ and $I^{\alpha\beta}$ is given by Eq. (4). The interaction described by Eq. (6) is different from the 180° exchange in two important respects. First, since for any orbital state the expectation value $\langle I^{\alpha\beta} \rangle > 0$, the spin exchange is effectively ferromagnetic independent of an orbital state: the average spin-exchange coupling $-J_{TS} \langle I^{\alpha\beta} \rangle < 0$ is negative (in agreement with the Goodenough-Kanamori-Anderson rules). Second, since $J_H \ll U_p$, the spin-isospin coupling in the 90° systems is weaker than the orbital exchange, described by Eq. (3):

$$\frac{J_{TS}}{J_T} = \frac{J_H}{U_p} \frac{2\Delta}{(2\Delta + U_p)}.$$

Thus, in 90° -exchange systems spins and orbitals cannot strongly influence each other, i.e., they are essentially decoupled [12].

We can now obtain the Hamiltonian describing orbital interactions in the Ni layers of NaNiO_2 [see Fig. 1(a)]. In each layer Ni ions form a triangular lattice, the sites of which lie on intersections of three sets of lines parallel to the unit vectors \mathbf{e}_{xy} , \mathbf{e}_{yz} , and \mathbf{e}_{zx} , as shown in Fig. 2. It is convenient to identify $\mathbf{e}_{\alpha\beta}$ with the three unit vectors in the isospace [see Eq. (2)] by $\mathbf{n}^x = \mathbf{e}_{yz}$, $\mathbf{n}^y = \mathbf{e}_{zx}$, and $\mathbf{n}^z = \mathbf{e}_{xy}$. The bonds between the nearest-neighbor sites j and $j + \mathbf{e}_{\alpha\beta}$ of the triangular lattice are diagonals of the Ni-O plaquettes lying in the $\alpha\beta$ plane [cf. Fig. 1(b)]. Hence, the Hamiltonian describing orbital interactions on the triangular lattice has the form:

$$H_T = J_T \sum_j \sum_{\alpha \neq \beta} I_j^\alpha I_{j+\mathbf{e}_{\alpha\beta}}^\beta. \quad (7)$$

The terms linear in the operators I_j^α [see Eq. (3)] drop out, since $\sum_\alpha I_j^\alpha = 0$. The Hamiltonian Eq. (7) is invariant under the global rotation of isospin operators T^α over the angle $\frac{2\pi}{3}$ around the y axis combined with the rotation of the triangular lattice over the same angle. This transformation is equivalent to the cyclic permutation of the indices x , y , and z of the vectors $\mathbf{e}_{\alpha\beta}$ and the isospin operators I^α .

The orbital exchange on a triangular lattice is strongly frustrated and has a large number of mean-field ground states. The simplest minimal energy states are the ferro-orbital states, in which $\langle \mathbf{T}_j \rangle = T\mathbf{m}$ on all lattice sites, where \mathbf{m} is an arbitrary unit vector in the (T^x, T^z) plane and $T = \frac{1}{2}$. The energy of these states is independent of orientation of \mathbf{m} , even though the Hamiltonian (7) is not invariant under arbitrary rotations in the (T^x, T^z) plane (this continuous ground-state degeneracy is similar to one of the three-dimensional ‘‘compass’’ models [1,13]). Furthermore, there are disordered ground states, which

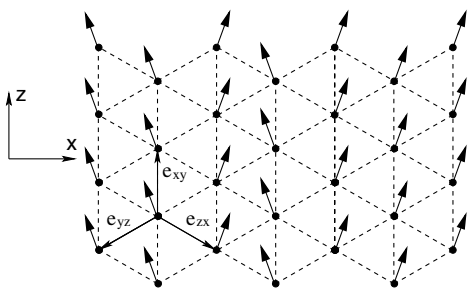


FIG. 2. The triangular lattice formed by Ni ions in the [111] plane. Shown also is a disordered mean-field ground state, in which the isospins form lines parallel to the unit vector \mathbf{e}_{xy} , such that $\langle T_j^z \rangle$ is the same on all lattice sites, while the sign of $\langle T_j^x \rangle$ varies arbitrarily from line to line.

can be obtained from the ferro-orbital states by inverting the sign of $\langle T_j^x \rangle$ on an arbitrarily selected set of lines parallel to \mathbf{e}_{xy} (see Fig. 2). Such states are ordered along the xy lines, but there are no long-range correlations between the x projections of isospins in the transverse direction. By circular permutations of the x , y , and z indices, one can obtain similar states, which are ordered only along the lines parallel to the \mathbf{e}_{yz} and \mathbf{e}_{zx} vectors.

This large ground-state degeneracy is lifted by quantum orbital fluctuations (the so-called ‘‘order-from-disorder’’ mechanism [14,15]). First, we have checked numerically that disordered states (see Fig. 2) have a higher energy of the zero-point fluctuations than the corresponding uniform states. Second, the zero-point energy of uniform states has six minima at the isospin orientations $\mathbf{m} = \pm \mathbf{n}^x, \pm \mathbf{n}^y, \pm \mathbf{n}^z$. In the original notation these are the ferro-orbital states with one of the orbitals $d_{3z^2-r^2}, d_{3x^2-r^2}, d_{3y^2-r^2}, d_{x^2-y^2}, d_{y^2-z^2}$, and $d_{z^2-x^2}$ occupied at each site. Furthermore, the anharmonicity effects [16] usually stabilize elongated octahedra, i.e., the orbitals $d_{3z^2-r^2}, d_{3x^2-r^2}$, or $d_{3y^2-r^2}$. As was mentioned above, spins in layers order ferromagnetically.

We note that the standard description of orbital excitations with noninteracting bosons [random phase approximation (RPA)] is insufficient for calculating quantum corrections to the ground-state energy of the frustrated Hamiltonian Eq. (7). In that approximation the orbital excitation spectrum for the six ground states is one-dimensional

$$\omega_{\mathbf{q}} = 3\sqrt{2}J_T \left| \sin \frac{(\mathbf{q} \cdot \mathbf{m})}{2} \right|, \quad (8)$$

which is a direct consequence of the absence of long-range correlations between chains in disordered mean-field ground states. The gapless one-dimensional spectrum leads to infrared-divergent fluctuations. To get rid of them, one has to take into account interactions between the bosons, which can be consistently done in the large isospin limit, $T \gg 1$. The interactions suppress quantum fluctuations by opening a gap and inducing a dispersion in the direction transverse to \mathbf{m} . This can be understood from the fact that the self-energy diagrams shown in Fig. 3 diverge unless a gap is introduced (in field theory a similar mechanism is known as a ‘‘dynamical mass generation’’ [17]). The physical origin of the gap is the breaking of the continuous ground-state degeneracy by quantum fluctuations [13]. In the large- T limit quantum fluctuations are relatively small and the gap $\Delta \propto J_T \sqrt{T}$ is much smaller than the bandwidth $W \sim 3\sqrt{2}J_T T$ of the orbital excitations. The dispersion in the transverse direction is of the order of Δ [18].

The main conclusions of our theory of the 90° exchange (spins are coupled much weaker than orbitals; the ground state is ordered both ferro-orbitally and ferromagnetically) are in agreement with the orbital and magnetic structure of the layered compound NaNiO_2 . This

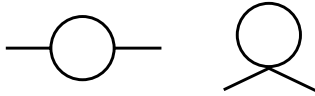


FIG. 3. The self-energy diagrams due to the cubic and the quartic interactions of orbital excitations, resulting in a gap opening and a two-dimensional dispersion.

material shows two transitions: the ferro-orbital ordering of the $d_{3z^2-r^2}$ type at $T_o = 480$ K, below which the oxygen octahedra become elongated, and the magnetic transition at the much lower temperature $T_s = 20$ K into the A-type antiferromagnetic state, in which the Ni spins in the [111] layers are ordered ferromagnetically [7].

It is, therefore, very puzzling that the structurally identical material LiNiO_2 shows neither orbital nor spin ordering. In principle, we cannot exclude that the quantum orbital fluctuations, which for $T = \frac{1}{2}$ are large, destroy the long-range ferro-orbital order (such quantum melting was discussed in the context of another frustrated system in Ref. [19]). We note that this possible orbital-liquid scenario is fundamentally different from those based on a strong interplay between orbitals and spins [4,10,11]. Actually, due to the decoupling of spins from orbitals, this orbital liquid would still have a ferromagnetic order. In fact, LiNiO_2 and NaNiO_2 have close values of the Weiss constant, corresponding to the ferromagnetic exchange in the Ni layers [7,9].

We think that the puzzles surrounding LiNiO_2 indicate the importance of disorder and electron-lattice interactions, not included in the present theory. As was argued in Refs. [7,20], the nonstoichiometry of LiNiO_2 samples results in the presence of magnetic Ni ions in the Li [111] planes, which leads to a strong interlayer coupling frustrating the three-dimensional antiferromagnetic spin ordering. This agrees with the recent observation of a spin-glass-like anomaly in magnetic susceptibility at ~ 8 K [21]. In NaNiO_2 such disorder is much weaker: as the size difference of Na and Ni ions is larger than that of Li and Ni ions, the alternating stacking of the Na and Ni [111] layers is more perfect than that of Li and Ni in LiNiO_2 . The larger radius of the Na ion also implies stronger electron-lattice interactions in NaNiO_2 that, in general, suppress quantum orbital fluctuations and stabilize a ferro-orbital ordering [22,23].

Finally, some frustration of orbital interactions (due to their anisotropy) may also occur in JT systems with simple lattices. It is, however, not as strong as in geometrically frustrated materials with the same dimensionality: e.g., while for a cubic lattice the RPA spectrum of orbital fluctuations is two-dimensional, for a pyrochlore lattice of corner sharing tetrahedra it is one-dimensional, implying larger fluctuations in the latter case. The excitations for the $d_{3z^2-r^2}$ ferro-orbital state on the pyrochlore lattice propagate only along two disconnected sets of

bonds with the $I^x I^y$ type of exchange, similarly to the case of the triangular lattice [see Eq. (8)]. However, this additional geometric frustration comes together with the spin-orbital decoupling and, therefore, cannot result in a spin-orbital liquid state. In general, orbital liquids seem to be “singular points” in the parameter space of exchange interactions (such as spin liquids [24]), which can be easily turned into ordered states by quantum effects and electron-lattice interactions. Still, the frustration in LiNiO_2 can play an important role in the strong suppression of the orbital ordering by tiny deviations from stoichiometry.

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