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Patterns of symmetry breaking in systems of coupled tetrahedra

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Abstract
The phases of a system of weakly coupled tetrahedra ($S = 1/2$), in the presence of both Heisenberg and antisymmetric Dzyaloshinsky–Moriya interactions, are discussed. While non-magnetic dimer order is dominant for the specific interactions considered, we find that Dzyaloshinsky–Moriya induced magnetic long-range order can also emerge. The presence of antisymmetric interactions also leads to non-trivial effects (e.g. ordering) in an external magnetic field.

1. Introduction

The Heisenberg model on the pyrochlore lattice (a 3D network of corner-sharing tetrahedra) is strongly frustrated and represents a long-standing theoretical challenge as to the nature of the ground state and excitations. The situation is most unclear for low values of the spin when no magnetic order seems to be present and the possibility of singlet (dimer) order is under debate [1, 2].

The purpose of this paper is to look at a different class of models, where the tetrahedra are coupled not via their corners, but almost in a 2D square lattice-like arrangement. Our work was partially motivated by recent experimental studies of the $S = 1/2$ material $Cu_2Te_2O_5Br_2$ [3], a representative of such a geometry. This compound exhibits a number of interesting properties, such as a phase transition at $T_c \approx 11$ K into a phase whose nature is still unclear (possibly weakly ordered magnetically or disordered). Quite unusually, a sharp peak appears for $T < T_c$ in Raman spectroscopy (measuring $S = 0$ excitations) at energy (singlet 'gap') $\Delta \approx 24$ K. This value, well below the two-triplet continuum [3], in combination with the sharpness of the peak, suggests that this mode is a well-defined low-energy singlet excitation of the system. Moreover, the singlet mode shifts to lower energy as a function of temperature and the gap seems to disappear at T_c, signalling that the phase transition itself possibly takes place in the

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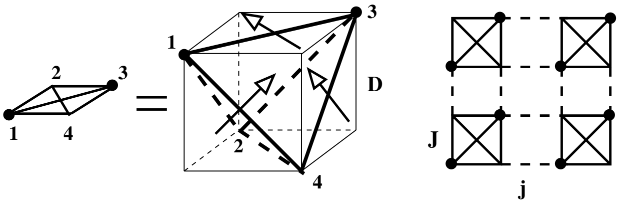


(a) (b)

Figure 1. (a) A tetrahedron with DM interactions represented by the arrows. (b) A 2D lattice of coupled tetrahedra.

singlet sector. Finally, both $\Delta(T \ll T_c)$ and T_c show a weak increase as a function of external magnetic field.

The goal of the present work is however more general than explaining the experimental data described above (much still remains unclear in that respect and no attempt of specific fits will be made). We will aim to analyse the possible ground states of a system of weakly interacting tetrahedra, starting from the case of Heisenberg exchange (section 2) and then also introducing antisymmetric, Dzyaloshinsky–Moriya (DM) interactions (section 3). We will show that the presence of both types of interactions can naturally lead to relevant low-energy singlet dynamics coexisting, in a certain range of parameters, with weak magnetism.

2. Dimer order in a system of weakly interacting tetrahedra

We start with the $S = 1/2$ Heisenberg model represented in figure 1(b) and will assume that the Heisenberg exchanges within the tetrahedra (J) are strong, whereas the inter-tetrahedral exchanges (j) are weak, i.e. $J \gg j$. Both are assumed to be antiferromagnetic $J, j > 0$. We represent the tetrahedra as plaquettes, as shown in figure 1(a) (where the arrows should be ignored for now, as they are the DM vectors discussed in the next section). This model has already been discussed in [4], and we will summarize the results we need here.

For a single tetrahedron ($j = 0$) the ground state is doubly degenerate and consists of two singlets ($S = 0$):

$$\begin{aligned} |s_1\rangle &= \frac{1}{\sqrt{3}}\left[\left(\begin{array}{c}\circ\!-\!\circ\\ \circ\!-\!\circ\end{array}\right) + \left(\begin{array}{cc}\circ & \circ\\ | & |\\ \circ & \circ\end{array}\right)\right] \\ |s_2\rangle &= \left[\left(\begin{array}{cc}\circ & \circ\\ | & |\\ \circ & \circ\end{array}\right) - \left(\begin{array}{c}\circ\!-\!\circ\\ \circ\!-\!\circ\end{array}\right)\right] \end{aligned} \tag{1}$$

where $\circ\!-\!\circ$ in (1) represents a singlet formed by two spins. The excitations consist of three triplets ($S = 1$) with high energy J and even higher spin 2 states. As long as $j \ll J$ we can concentrate on the singlet dynamics only, whereas certainly in the regime $j \sim J$ a transition to a magnetic state can occur [5]. To see how the degeneracy is lifted by finite $j \ll J$ it is convenient to use the pseudospin language where pseudospin $T_z = 1/2$ corresponds to $|s_1\rangle$ and $T_z = -1/2$ corresponds to $|s_2\rangle$. Then one can write an effective Hamiltonian in the singlet subspace with the result (to second order)

$$\begin{aligned} H_{\text{eff}} = &-\frac{J_{\text{eff}}}{2}\sum_{\langle \mathbf{i},\mathbf{j}\rangle}\left[T_{x,\mathbf{i}}T_{x,\mathbf{j}} + \frac{1}{3}T_{z,\mathbf{i}}T_{z,\mathbf{j}} + \frac{\mathrm{e}^{\mathrm{i}\mathbf{Q}\cdot(\mathbf{i}-\mathbf{j})}}{\sqrt{3}}(T_{z,\mathbf{i}}T_{x,\mathbf{j}} + T_{x,\mathbf{i}}T_{z,\mathbf{j}})\right] \\ &- h_{\text{eff}}\sum_i T_{z,\mathbf{i}}, \qquad \text{where } J_{\text{eff}} = \frac{j^2}{2J}, \quad h_{\text{eff}} = \frac{j^2}{6J}. \end{aligned} \tag{2}$$

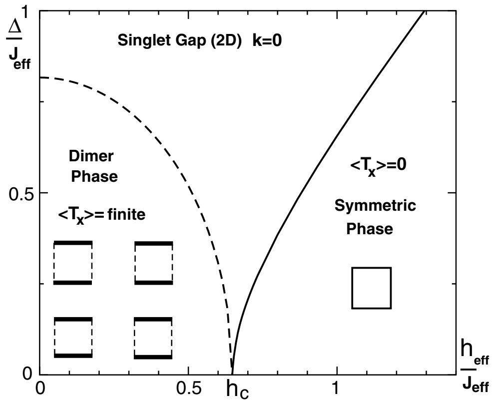


Figure 2. The phase diagram of the model (2).

Here the indexes $\mathbf{i}, \mathbf{j}$ represent different tetrahedra with summation over nearest neighbours, and $\mathbf{Q} = (\pi, 0)$.

In figure 2 the gap Δ is plotted as a function of the effective field h_{eff}, and one finds two phases (with an Ising transition between them): (I) the symmetry-broken phase with $\langle T_{x,\mathbf{i}}\rangle \neq 0$ for $h_{\text{eff}} < h_{\text{c}}$, and (II) the symmetric phase for $h_{\text{eff}} > h_{\text{c}}$. It is clear from equation (2) that $h_{\text{eff}}/J_{\text{eff}} = 1/3$ meaning that the model (2) is in the phase with $\langle T_{x,\mathbf{i}}\rangle \neq 0$. It can be easily seen that this phase corresponds to dimerization, i.e. stronger spin–spin correlations on some bonds relative to others as shown in figure 2.

Thus a system of weakly coupled tetrahedra dimerizes spontaneously at $T = 0$. Since an Ising symmetry is broken, there is a finite (small) singlet gap $\Delta \sim J_{\text{eff}}$ with all spin 1 excitation much higher in energy. This phase persists up to $T_{\text{c}} \sim J_{\text{eff}}$. Magneto-elastic couplings could also contribute to the dimerization tendency as discussed for the pyrochlore case [6, 7], but in our scenario dimerization occurs due to purely Heisenberg exchanges.

3. Quantum phases in the presence of Dzyaloshinsky–Moriya (DM) interactions

For a system with DM interactions of low enough symmetry, originating from the spin–orbit interaction, are always present [8, 9]. Whether the DM interaction is present on a particular bond depends on the symmetry of the environment, and instead of performing the analysis for a specific material we will make the following assumptions in our model:

(i) DM interactions are present on the tetrahedra only (where the Heisenberg exchange is also dominant); and
(ii) DM interactions are present on all tetrahedral bonds.

Then on a single tetrahedron we have $H_{\text{tetrahedron}} = J\sum_{k,l}\mathbf{S}_k\cdot\mathbf{S}_l + \sum_{k,l}\mathbf{D}_{k,l}\cdot(\mathbf{S}_k\times\mathbf{S}_l)$, $k, l = 1, 2, 3, 4$, where the DM vectors $\mathbf{D}_{k,l}$ reside on the six bonds of the tetrahedron, with directions as shown in figure 1(a) (only three vectors are drawn for clarity), so the tetrahedral group is respected. All the vectors thus have equal magnitude which we call D from now on.

The presence of the DM interactions does not lift the degeneracy of the ground state on one tetrahedron, but admixes triplets to the two singlets (1). We represent the three triplets by p_μ, q_μ, t_μ, $\mu = x, y, z$, in the notation of [10]. Since D is expected to be small, at most several per cent of the Heisenberg exchange, we can work for simplicity in the perturbative

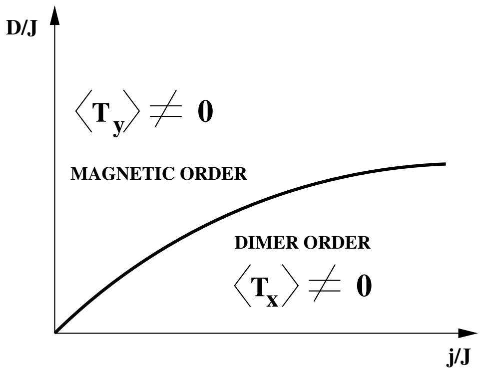


Figure 3. A schematic phase diagram of the model (4).

limit $D \ll J$. Instead of (1) the ground state becomes

$$
\begin{aligned}
|s_1\rangle_{\rm DM} &= |s_1\rangle + \frac{{\rm i}3D}{2\sqrt{6}J}[|p_x\rangle - |p_y\rangle + |q_x\rangle + |q_y\rangle] \\
|s_2\rangle_{\rm DM} &= |s_1\rangle + \frac{{\rm i}D}{2\sqrt{2}J}[|p_x\rangle + |p_y\rangle + |q_x\rangle - |q_y\rangle] + {\rm i}\frac{D}{J}|t_z\rangle.
\end{aligned} \tag{3}
$$

Working within this subspace we find that the effective Hamiltonian (2) is modified already at first order in the inter-tetrahedral exchange j and takes the form, to lowest order in D/J,

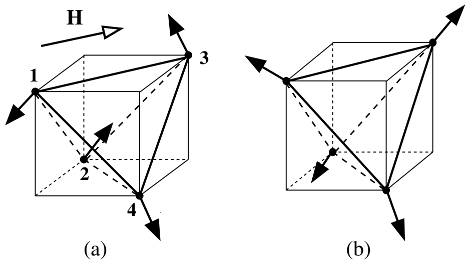
$$
H_{\rm eff}^{\rm DM} = H_{\rm eff} - j\frac{4D^2}{3J^2}\sum_{\langle \mathbf{i},\mathbf{j}\rangle} T_{y,\mathbf{i}}T_{y,\mathbf{j}}. \tag{4}
$$

The additional term generated by D creates the possibility of ordering in the y pseudospin component $T_{y,\mathbf{i}}$. Since the effective Hamiltonian has the form of a ferromagnetic spin model with Ising anisotropy, the y ordering can occur only if the interaction strength in that direction is larger than the one in the x direction. In the perturbative limit discussed above for example this gives the critical strength: $D_{\rm c}/J \sim \sqrt{j/J}$. The more general situation has to be analysed numerically, which we will not do here. Quite generally we expect two phases, represented schematically in figure 3: one with $\langle T_{y,\mathbf{i}}\rangle \neq 0$ (and $\langle T_{x,\mathbf{i}}\rangle = 0$) for $D > D_{\rm c}$, and another with $\langle T_{x,\mathbf{i}}\rangle \neq 0$ ($\langle T_{y,\mathbf{i}}\rangle = 0$) for $D < D_{\rm c}$. These phases are (pseudospin) ferromagnetic, and from now on we skip the lattice index and write for their respective order parameters: $\langle T_{x,\mathbf{i}}\rangle = \langle T_x\rangle$, $\langle T_{y,\mathbf{i}}\rangle = \langle T_y\rangle$. We proceed now to analyse the structure of the two phases in terms of real spins.

3.1. Magnetic field effects in the dimer phase

First let us consider the phase with $\langle T_x\rangle \neq 0$. This is the dimerized phase discussed in section 2, with small additional modulation of the correlations due to the presence of the DM interactions. This phase still has no magnetic order (at zero external field), in the sense that $\langle \mathbf{S}_k\rangle = 0$, $k = 1, 2, 3, 4$, on every site of the tetrahedron.

However, in an external (uniform) magnetic field $-\mathbf{H}\cdot\mathbf{S}^{\rm tot}$, due to the admixture of $S = 1$ excitations to the ground state (3), long-range (field induced) order appears and the physical quantities such as gaps start depending on the field. For example in an external magnetic field



(a) (b)

Figure 4. (a) The spin arrangement in an external field **H** in the phase $\langle T_x \rangle \neq 0$ (the tilt of spins in the field direction is not shown). (b) The spin arrangement in the phase $\langle T_y \rangle \neq 0$, at zero external field.

in the plane, $\mathbf{H} = \frac{H}{\sqrt{2}}(1, 1, 0)$ (i.e. along the diagonal connecting sites 1 and 3; see figure 4(a)):

$$\frac{T_c(H)}{T_c(0)} \approx 1 + 0.35\left(\frac{J}{j}\right)^2 \frac{D^2 H^2}{J^4}, \qquad \frac{\Delta(H)}{\Delta(0)} \approx 1 + 0.86\left(\frac{J}{j}\right)^2 \frac{D^2 H^2}{J^4}. \tag{5}$$

This (weak) magnetic field dependence is due to an energy splitting between the two ground states (3) in a magnetic field. We also note that even though (5) suggests that the phase is more stable in a magnetic field, this result is not universal and strongly depends on the field orientation (in particular the tendency is reversed for a field in the (0, 0, 1) direction).

The spin structure in a field has two components. The first one is a magnetic moment in the field direction (spins on all sites align along the field), proportional to $\frac{D^2 H}{J^3}$. This is a single-tetrahedron ($j = 0$) contribution. The second one is a long-range component, originating form inter-tetrahedral interactions, generating the pattern shown in figure 4(a). In this pattern $\langle \mathbf{S}_3 \rangle = -\langle \mathbf{S}_4 \rangle$, $\langle \mathbf{S}_1 \rangle = -\langle \mathbf{S}_2 \rangle$, and the spins on sites 2 and 4 point along the diagonals of the cube (towards and out of the cube's centre, respectively). The value of the magnetic moment in this pattern (length of arrows) is: $|\mathbf{S}_k| \sim \frac{DH}{J^2}\langle T_x \rangle$. Since the pseudospin interactions are ferromagnetic (2), the magnetic field induced pattern of figure 4(a) is the same on all tetrahedra.

3.2. Magnetic order induced by Dzyaloshinsky–Moriya interactions

Now we discuss the regime where the DM interaction is strong enough to produce the non-zero average $\langle T_y \rangle \neq 0$. Only the case of zero external magnetic field is considered since it already yields non-trivial magnetic order.

We find that the $T_{y,\mathbf{i}}$ ordering corresponds to four-sublattice magnetic long-range order where the moments on each tetrahedron point along the cube's diagonals outward from the centre; figure 4(b). The value of the moment is proportional to the DM interaction: $|\mathbf{S}_k| \sim \frac{D}{J}\langle T_y \rangle$. As a function of temperature this moment is finite up to a critical temperature with a scale set by (4): $T_c^{\mathrm{DM}} \sim j\frac{D^2}{J^2}$. It should be kept in mind that for the tetrahedral lattice studied in this work (figure 1(b)) the DM induced magnetic order competes with the non-magnetic dimer order, and the former can 'win' over the latter only when $D > D_c$. We will not attempt to discuss here how feasible this condition is for specific materials. It is certainly clear that the presence of additional Heisenberg interactions that frustrate the dimer order will make it easier for the DM induced magnetic order to emerge.

Finally we note that DM induced ordering has been discussed recently from a semiclassical perspective for the case of the Kagome [11] and the pyrochlore [12] lattices. However, the mechanism discussed here is rather different. We have shown, for the lattice of figure 1(b) and in the extreme quantum limit $S = 1/2$, that magnetic order can be generated due to lifting of the degeneracy in the subspace of tetrahedra states (3). In the presence of DM interactions these states are not pure singlets but break spin-rotational invariance ($\langle (\mathbf{S}^{\mathrm{tot}})^2 \rangle \neq 0$, $D \neq 0$), making it possible to have long-range magnetic order in the whole lattice system. The resulting magnetic moment is small, governed by the ratio D/J. Alternatively, in the phase with no magnetic moment (dimer phase), DM interactions can manifest themselves by non-trivial magnetic field induced order and dependence of observables on the magnetic field. We believe the above effects to be generically important for systems composed of tetrahedra, since typically the (low) symmetry of such systems allows the presence of DM interactions. The size of these effects, and consequently their possible observability, depends on the specific values of the interactions and their distribution around the lattice.

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