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Ordering in pyrochlore compounds due to Dzyaloshinsky–Moriya interactions: the case of Cu_4O_3

Maged Elhajal¹, Benjamin Canals and Claudine Lacroix

Laboratoire Louis Néel, CNRS, 25 avenue des Martyrs, BP 166, 38042 Grenoble Cedex 9, France

E-mail: Maged.Elhajal@ipt.unil.ch

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Abstract

We investigate the effects of Dzyaloshinsky–Moriya interactions (DMI) on the magnetic properties of the pyrochlore antiferromagnet. The relevance of this interaction for such a system is shown and we describe how to take it into account in accordance with the symmetries of the pyrochlore lattice. A phase transition from a high temperature paramagnetic phase to a low temperature long range ordered state takes place as soon as DMI are present. We describe the different magnetic structures obtained depending on the geometry of the DMI and special attention is given to the compound Cu_4O_3 for which a possible explanation of the experimentally observed ordering is given.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

In this paper we first summarize some of the results of a more detailed study of Dzyaloshinsky–Moriya interactions (DMI) in the pyrochlore lattice [9] and then present a related work taking into account a lattice distortion. We show how this could explain experimental results of the low temperature magnetic structure of Cu_4O_3 [4].

2. DMI on the pyrochlore lattice

DMI are anti-symmetric exchange interactions which are written as $\mathbf{D}_{ij} \cdot (\mathbf{S}_i \times \mathbf{S}_j)$, where \mathbf{D}_{ij} is a vector related to the crystallographic structure and depends on the microscopic details of the system. It is closely related to the existence of spin–orbit coupling [1]. Deriving DMI in a microscopic approach is only feasible in some simple situations [1, 2], and otherwise is a very difficult task.

¹ Author to whom any correspondence should be addressed. Present address: Institut de Physique Théorique, BSP, Université de Lausanne, 1015 Lausanne, Switzerland.

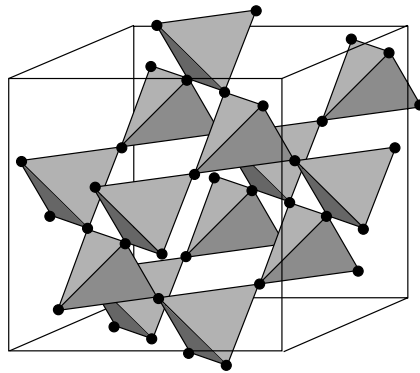


Figure 1. The low symmetry of the pyrochlore lattice. The fact that the middle point between two sites is not a centre of inversion allows for the existence of Dzyaloshinsky–Moriya interactions.

For our purposes here, it is enough to know that the direction of \mathbf{D} can be determined by inspection of the symmetry of the system (see section 2.1). The Hamiltonian we will consider is:

$$H = J \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j + \sum_{\langle ij \rangle} \mathbf{D}_{ij} (\mathbf{S}_i \times \mathbf{S}_j) \quad (1)$$

where the sums are over nearest neighbours. Usually, \mathbf{D} is much smaller than J , where J is the usual isotropic exchange.

2.1. Symmetry arguments to determine the \mathbf{D} vectors

The Hamiltonian must be invariant under the symmetry operations which leave the system invariant. This restricts the set of \mathbf{D} which can appear in the Hamiltonian. Moriya [1] enumerated a set of rules based on this fact and we applied them to the pyrochlore structure. For the present purpose, it is sufficient to say that two possible DMI can appear in the pyrochlore structure (see figures 1 and 2). This results from the fact that the plane containing two sites and the centre of the opposite bond is a mirror plane and from the symmetries of the pyrochlore lattice. Details will be given elsewhere [9].

2.2. Magnetic structures

Both of the possible DMI lead to an ordering of the pyrochlore antiferromagnet at low temperature. The two structures are described by a $\mathbf{q} = \mathbf{0}$ wavevector and the two magnetic unit cells are represented in figure 3.

The direct structure (see figure 3) is that of FeF_3 at low temperature [5]. DMI are not the only possible interactions that explain such an ordering, it appears also in the presence of single-ion anisotropies, further neighbour interactions [5, 6]. One can exclude some of these possibilities by looking at the microscopic details of the compound, for instance single ion anisotropies should not be very strong in Fe^{3+} . However, more theoretical and experimental work would be necessary to discriminate between the different scenarios.

The long ranged ordered magnetic structure of ZnCr_2O_4 at low temperature is not uniquely determined. However, one of the candidates shares some similarities with the indirect structure obtained here [8], such as planar anisotropy in the (001) planes and 90° angles between some of the spins. Although the relation between DMI and the structure of ZnCr_2O_4 is not obvious,

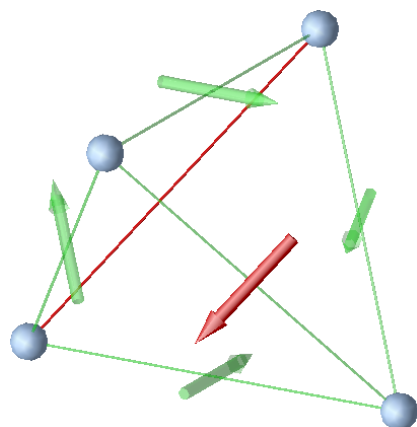


Figure 2. \mathbf{D} for the pyrochlore structure. Each \mathbf{D} is parallel to the opposite bond of the tetrahedron. There are actually two possible DMI, one of which is represented here. The other one is obtained by inverting all the \mathbf{D} . These two DMI are called ‘direct’ and ‘indirect’. See [9] for more details.

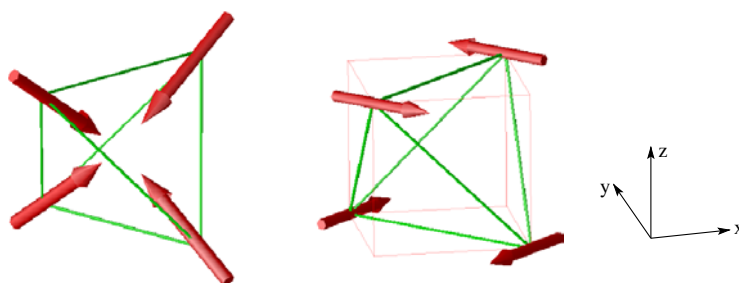


Figure 3. The two magnetic structures, obtained for ‘direct’ (left) and ‘indirect’ (right) DMI. Both of them are $\mathbf{q} = \mathbf{0}$ structures so that all tetrahedra of the pyrochlore have this structure.

it may be of interest to take into account the distortion occurring in ZnCr_2O_4 to include DMI in the same way as is done in detail in the next section for Cu_4O_3 .

3. Cu_4O_3 (paramelaconite)

Cu_4O_3 is a mineral compound in which the spin- $\frac{1}{2}$ Cu^{2+} ions form a tetragonally distorted pyrochlore lattice. The crystallographic structure is elongated along the (001) direction of the cubic cell, as detailed in [3, 4]. This involves a distortion of the tetrahedra of the pyrochlore structure as represented in figure 4. In contrast to what is predicted for the undistorted pyrochlore structure with nearest neighbour antiferromagnetic interactions, this compound orders magnetically at low temperature [4] with $\mathbf{q} = (0, 1, 1/2)$ in the cubic frame. Its magnetic structure is not fully identified and there are two candidates [4]. This ordering is not a surprise on its own, since one would expect departures from the isotropic Heisenberg Hamiltonian due to the distortion of the lattice. The most natural way to take into account this distortion is to introduce different (J) coupling constants of the isotropic exchange interaction ($\mathbf{S}_i \cdot \mathbf{S}_j$) along different types of bond. This was done in [4] and failed to reproduce the experimentally observed structures. Taking into account DMI according to the crystallographic structure

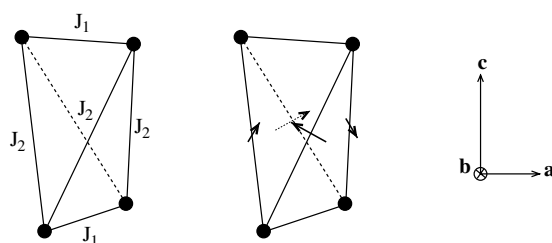


Figure 4. Tetragonal distortion in Cu_4O_3 . Left: the six bonds of each tetrahedron are no longer equivalents and it is natural to introduce two distinct coupling constants. Right: the \mathbf{D} for Cu_4O_3 . DMI on the two bonds in the (ab) planes are forbidden by symmetry. The direction of the other \mathbf{D} is slightly changed compared to the undistorted pyrochlore.

(in addition to the previously mentioned different J coupling constants), we were able to reproduce one of the two candidates for the magnetic structure.

The rest of this section is devoted to summarizing what are the most sensible isotropic exchange constants to be taken into account (as detailed in [4]), then what are the possible DMI in Cu_4O_3 , and finally we give details of the parameters which lead to the experimentally observed magnetic structure.

3.1. Rough estimate of the relevant isotropic exchange interactions

Single ion anisotropies should be small due to the isotropy of the spin- $\frac{1}{2}$ Cu^{2+} , and it is much more natural to try and explain the magnetic structure by introducing anisotropies in the interactions. The most obvious anisotropy is that the coupling constants for the six bonds of one tetrahedron are no longer equal since the six bonds are now inequivalent. One should introduce two coupling constants J_1 and J_2 as in figure 4. This does not lead to the correct magnetic structure, and a detailed analysis of the super-exchange paths suggested that there should be some non-negligible interaction between second neighbours in the same (ab) plane [4]. This super-super-exchange path is sketched in figure 5 and the corresponding coupling constant is J_3 . J_3 is also confirmed by an *ab initio* approach [7]. By varying J_1 , J_2 and J_3 , several magnetic structures can be obtained, but none of them has the same wavevector as the one observed in the experiments [4].

3.2. Moriya's rules

Since Cu_4O_3 is not an ideal pyrochlore structure and the \mathbf{D} depend strongly on the symmetry of the crystallographic structure, one expects significant departures for the \mathbf{D} compared to the undistorted pyrochlore. It is easily seen from figure 5 that the DMI between two neighbouring sites in the same (ab) plane (bonds corresponding to J_1) is forbidden by symmetry. This follows from Moriya's rules [1] because the middle point between the two sites is a centre of inversion. For the four other bonds of each tetrahedron, DMI are allowed by the symmetry since the middle point is not an inversion centre, as can be seen from figure 5. The exchange path between two Cu^{2+} in two different (ab) planes goes through one oxygen ion. According to Moriya's rules [1], the \mathbf{D} must be perpendicular to the plane defined by the two copper ions and the oxygen atom. This leaves only two possible directions for \mathbf{D} , and when one of the \mathbf{D} is fixed, then all the others in the entire lattice are fixed by symmetry. The DMI allowed by the symmetry of Cu_4O_3 are summarized in figure 4.

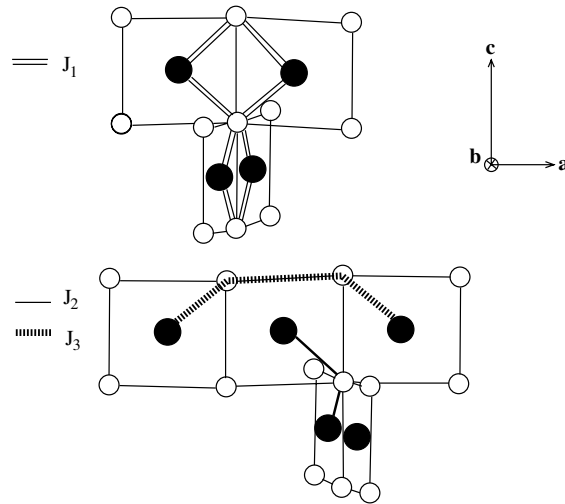


Figure 5. Exchange paths between Cu^{2+} ions (filled circles) through oxygen ions (empty circles). The first figure shows the exchange path J_1 between nearest neighbours in the same (ab) plane. The second figure shows the exchange paths J_2 between nearest neighbours in different (ab) planes and J_3 between second nearest neighbours in the same (ab) plane. Concerning DMI, they are non-zero only for nearest neighbours Cu^{2+} between different (ab) planes (see text). (Figure taken from [4]).

3.3. Magnetic structure

According to the previous section, the Hamiltonian we consider is:

$$H = J_1 \sum_{(ab) \text{ nn}} \mathbf{S}_i \mathbf{S}_j + J_2 \sum_{(c) \text{ nn}} \mathbf{S}_i \mathbf{S}_j + J_3 \sum_{(ab) \text{ nnn}} \mathbf{S}_i \mathbf{S}_j + \sum_{(c) \text{ nn}} \mathbf{D}_{ij} (\mathbf{S}_i \times \mathbf{S}_j) \quad (2)$$

where nn and nnn mean the nearest neighbours and next nearest neighbours, respectively, whereas (ab) means ‘in the same (ab) plane’ and (c) is for ‘in two neighbouring (ab) planes (along the c axis)’. We performed classical Monte Carlo simulations in order to explore the low temperature magnetic properties of the system. The simulations were done on samples up to 1024 spins. The only technical subtlety was to check that the magnetic order observed in the simulations is not due to the periodic boundary conditions on too small samples.

It is found that, except for very peculiar values of the parameters in (2), the system is always magnetically ordered at low temperature. There are several possible orderings, and we did not try to explore all the phase diagram. Instead, we looked for a region (in parameter space) where the magnetic order is the same as the one observed in Cu_4O_3 (see figure 6). The ground state is still degenerate with respect to a global rotation around the c axis, at least at $T = 0$. We did not check if this degeneracy is lifted by thermal fluctuations.

This structure is obtained for $J_1 = J_2 = 10J_3 = 10D$ with indirect DMI. We checked that it is robust with respect to small perturbations. In particular, it does not seem to be important how small J_3 and D are compared to J_1 ; this will probably only change the critical temperature, and not the ground state configuration. By contrast, D should not be too big compared to J_3 , otherwise the magnetic structure is similar to the one found in the undistorted pyrochlore with only nearest neighbour interactions. The exact direction of \mathbf{D} is not very important either. In fact, the same magnetic structure is obtained on the undistorted pyrochlore with indirect DMI and J_3 coupling. It is however more satisfactory to explain the experimental results with the \mathbf{D} vectors particularized to the distortion of Cu_4O_3 , and the distortion is necessary to justify the presence of second neighbours interactions *only* in the same (ab) plane (J_3).

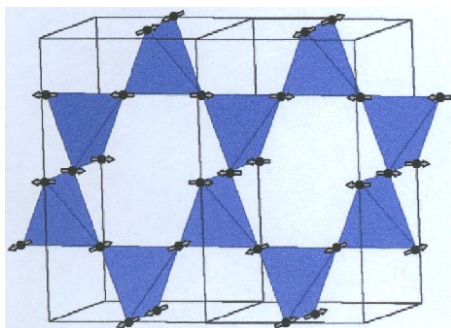


Figure 6. One of the two possible ordered magnetic structures of Cu_4O_3 as determined experimentally [4]. We obtained it theoretically with the Hamiltonian (2) with the parameters specified in the text. (Figure from [4]).

4. Conclusion

We have shown that DMI are relevant in the pyrochlore structure. Their presence leads to a long range order at low temperature and we have described the magnetic structures for both the undistorted pyrochlore lattice and the distorted pyrochlore Cu_4O_3 . The low temperature magnetic structure of this compound is not fully identified experimentally and we concentrated on one of the two possible candidates [4]. Due to the distortion, DMI are not the only departure from the ideal nearest neighbours antiferromagnet in this compound. Second neighbours interactions are important, however; DMI appear to be necessary to obtain the observed magnetic structure.

Acknowledgments

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