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1989 J. Phys.: Condens. Matter 1 4735

(http://iopscience.iop.org/0953-8984/1/28/022)

J. Phys.: Condens. Matter **1** (1989) 4735–4742. Printed in the UK

LETTER TO THE EDITOR

Long-range order in tetragonal antiferromagnets supported by quantum fluctuations

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Received 25 April 1989

Abstract. The nature of the phase transition in body-centred-tetragonal helimagnets is an argument of current theoretical interest both because high-T_c superconductors have just this structure and because the experimental data in a large number of rare earths and transition-metal compounds have suggested divergent interpretations and even the possible violation of the universality in second-order phase transitions. The theoretical approaches mainly employed are renormalisation-group analysis and Monte Carlo simulations. Both approaches are based on classical approximations. On the other hand we find that quantum fluctuations play an essential role in the low-temperature configuration in at least some regions of the parameter space. We find indeed that the model we consider can show *full frustration* in classical approximations. This frustration is lifted by the zero-point motion. The long-range order caused by quantum fluctuations differs from the order supported by thermal fluctuations and it is stable until a surprisingly high temperature: the order supported by quantum disorder prevails against the order supported by thermal disorder.

An impressive revival of the Heisenberg model with particular emphasis on the tetragonal lattice has been taking place because the magnetic properties of such models are believed to be relevant to the understanding of high-T_c superconductivity [1]. This new interest combines with a formerly active research area stimulated by the importance of the Heisenberg Hamiltonian as a model for a large number of magnetic insulators [2]. At present many questions both of basic and experimental interest await satisfactory answers. The nature of the phase transition in helimagnets remains an unsolved problem. Experimental data on Tb [3], Dy [4] and Ho [5] are differently interpreted as evidence of a first-order phase transition or, on the contrary, as indicating a second-order phase-transition, breaking the universality hypothesis. Renormalisation group (RG) analysis [6, 7] seems to support a first-order phase transition for helimagnets, where exchange competition is the source of the non-collinear spin configuration. Finally, interesting Monte Carlo (MC) simulations [8, 9] for body-centered-tetragonal (BCT) helimagnets indicate a first-order phase transition in XY-helimagnets, whereas a second-order phase transition should be expected in Heisenberg helimagnets. We notice that both approaches are based on classical approximations.

Here we consider the model of [8] and [9] but add an in-plane nearest-neighbour (NN) interaction J_1 that can be stronger than the interactions J_2 and J_3, the exchange couplings of a spin with its NN laying in the NN and in the next-nearest-neighbour (NNN) layers, respectively. J_2 and J_3 were taken into account in [8] and [9] but J_1 was neglected.

This Letter comprises the following.

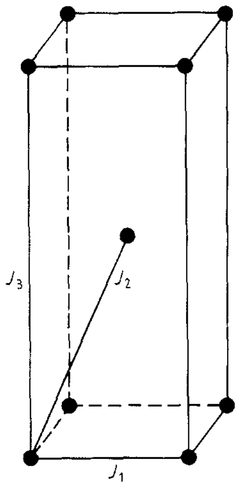

Figure 1. A body-centred-tetragonal (BCT) antiferromagnet. The exchange interactions we consider are indicated by J_1, J_2, J_3, respectively.

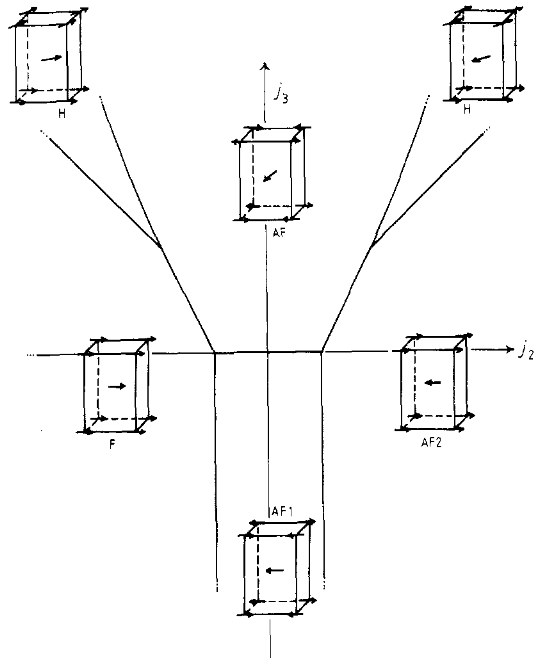

Figure 2. Zero-temperature phase diagram of the classical BCT antiferromagnet in the j_2–j_3 plane. F, AF1, AF2, H, AF configurations are described by equations (5)–(14).

(i) Calculation of the zero-temperature phase diagram of the J_1–J_2–J_3 model in the classical approximation. *Infinite degeneration lines* for $J_3 = 0$, $-1 < J_2/J_1 < 1$ and for $J_3 < 0$ with $J_2/J_1 = \pm 1$ are found.

(ii) A study of the zero-point motion that lifts the infinite degeneracy.

(iii) Consideration of the low-temperature thermodynamics of the model: we find that thermal fluctuations compete with quantum fluctuations. The order obtained by quantum disorder, however, turns out to be very hard to destroy, which is at variance with the behaviour of the rhombohedral Heisenberg antiferromagnet (RAF), where the full frustration entered into by the lattice structure is restored at intermediate temperatures by a more efficient outcome of the competition of thermal fluctuations with quantum fluctuations [10–12]. The results we obtain for the tetragonal antiferromagnet seem relevant to the understanding of the magnetic properties of La_2CuO_4 [13].

The Hamiltonian we consider is

$$H = -\sum_{\alpha} J_\alpha \sum_{i,\boldsymbol{\delta}_\alpha} \boldsymbol{S}_i \cdot \boldsymbol{S}_{i+\boldsymbol{\delta}_\alpha} \tag{1}$$

where i labels the sites of a BCT lattice and $\boldsymbol{\delta}_\alpha$ is a vector joining the site i with its NN in the plane containing the site i, if $\alpha = 1$; in the NN layers, if $\alpha = 2$; and in the NNN layers, if $\alpha = 3$, as shown in figure 1. We choose $J_1 < 0$, while J_2 and J_3 can have either signs.

In the classical approximation ($S \to \infty$) the ground-state energy of our model if the spins spiral according to a helical configuration characterised by the $\boldsymbol{Q}$ wavevector is

$$E_0 = -2J_1S^2N(\cos aQ_x + \cos aQ_y) - 8J_2S^2N\cos\tfrac{1}{2}aQ_x\cos\tfrac{1}{2}aQ_y\cos\tfrac{1}{2}cQ_z - 2J_3S^2N\cos cQ_z \qquad (2)$$

where a is the in-plane lattice constant and c is the distance between two NNN layers. The x and y axis are along two in-plane-perpendicular NN rows, and the z axis is along the c axis.

Let us define the reduced energy and exchange couplings as follows

$$e_0 = E_0/2|J_1|S^2N \qquad j_2 = J_2/J_1 \qquad j_3 = J_3/J_1. \qquad (3)$$

The minimum conditions for e_0 are

$$\sin\tfrac{1}{2}aQ_x(\cos\tfrac{1}{2}aQ_x + 2j_2\cos\tfrac{1}{2}aQ_y\cos\tfrac{1}{2}cQ_z) = 0 \qquad (4a)$$

$$\sin\tfrac{1}{2}aQ_y(\cos\tfrac{1}{2}aQ_y + 2j_2\cos\tfrac{1}{2}aQ_x\cos\tfrac{1}{2}cQ_z) = 0 \qquad (4b)$$

$$\sin\tfrac{1}{2}cQ_z(j_2\cos\tfrac{1}{2}aQ_x\cos\tfrac{1}{2}aQ_y + j_3\cos\tfrac{1}{2}cQ_z) = 0 \qquad (4c)$$

The solutions of (4) correspond to the following.

(i) A ferromagnetic (F) configuration whose wavevector and reduced energy are

$$\boldsymbol{Q} = 0 \qquad (5)$$

$$e_F = 2 + 4j_2 + j_3. \qquad (6)$$

The F phase is limited to the regions $j_2 < -1$ for $j_3 < 0$, $j_3 < -2j_2 - 2$ for $j_3 > 0$ with $-2 < j_2 < -1$ and $j_3 < -j_2$ for $j_3 > 0$ with $j_2 < -2$.

(ii) An antiferromagnetic (AF1) configuration where

$$Q_z = Q_y = \pi/a \qquad Q_z = 0 \qquad (7)$$

$$e_{AF1} = -2 + j_3. \qquad (8)$$

This phase is confined between the straight lines $j_2 = \pm 1$ for $j_3 < 0$. The line $j_3 = 0$, $-1 < j_2 < 1$ is an *infinite degeneration line* because $Q_x = Q_y = \pi/a$ but Q_z is arbitrary. The boundary lines $j_2 = \pm 1$, $j_3 < 0$ are also infinite degeneration lines with $Q_z = 0$, $Q_x = Q_y + 2\pi/a$ and Q_y arbitrary.

(iii) An antiferromagnetic (AF2) configuration where

$$Q_x = Q_y = 0 \qquad Q_z = 2\pi/c \qquad (9)$$

$$e_{AF2} = 2 - 4j_2 + j_3. \qquad (10)$$

The AF2 phase is stable for $j_2 > 1$ when $j_3 < 0$, for $j_3 < 2j_2 - 2$ when $1 < j_2 < 2$ and for $j_3 < j_2$ when $j_2 > 2$.

(iv) A helical (H) configuration where

$$Q_x = Q_y = 0 \qquad Q_z = (2/c)\cos^{-1}(-j_2/j_3) \qquad (11)$$

$$e_H = 2 - 2j_2^2/j_3 - j_3. \qquad (12)$$

The H phase is stable for $j_2 > 2$ with $j_2 < j_3 < j_2^2/2$ and for $j_2 < -2$ with $-j_2 < j_3 < j_2^2/2$.

(v) An antiferromagnetic (AF) phase where

$$Q_x = Q_y = \pi/a \qquad Q_z = \pi/c \tag{13}$$

$$e_{\mathrm{AF}} = -2 - j_3. \tag{14}$$

The AF phase is stable for $j_3 > j_2^2/2$ if $j_2 < -2$ or $j_2 > 2$, for $j_3 > -2j_2 - 2$ if $-2 < j_2 < -1$, for $j_3 > 0$ if $-1 < j_2 < 1$, and for $j_3 > 2j_2 - 2$ if $1 < j_2 < 2$.

Figure 2 shows the zero-temperature phase diagram of the BCT classical antiferromagnet in the j_2–j_3 plane.

To get quantum corrections to the classical ground state and the magnon-excitation spectrum we perform the customary steps [14]: we introduce a local quantisation axis spiralling according to a helix characterised by the wavevector $\boldsymbol{Q}$; we transform the spin operators to Bose creation and destruction operators by the Dyson–Maleev transformation, and finally we diagonalise the Bosonic equivalent Hamiltonian by keeping contributions proportional to S^2 and S.

The ground-state energy we obtain is

$$E_{\mathrm{G}} = E_0(1 + 1/S) + \tfrac{1}{2}\sum_{k} \hbar\omega_k \tag{15}$$

where E_0 is given by (2) and $\hbar\omega_k$ is the magnon energy-dispersion curve given by

$$\hbar\omega_k = 4|J_1|S(s_k d_k)^{1/2} \tag{16}$$

where

$$\begin{aligned} s_k = {} & \cos aQ_x + \cos aQ_y - \cos ak_x - \cos ak_y + 4j_2(\cos \tfrac{1}{2}aQ_x \cos \tfrac{1}{2}aQ_y \cos \tfrac{1}{2}cQ_z \\ & - \cos \tfrac{1}{2}ak_x \cos \tfrac{1}{2}ak_y \cos \tfrac{1}{2}ck_z) + j_3(\cos cQ_z - \cos ck_z) \end{aligned} \tag{17}$$

$$\begin{aligned} d_k = {} & \cos aQ_x(1 - \cos ak_x) + \cos aQ_y(1 - \cos ak_y) \\ & + 4j_2\{\cos \tfrac{1}{2}aQ_y[\cos \tfrac{1}{2}aQ_x \cos \tfrac{1}{2}cQ_z(1 - \cos \tfrac{1}{2}ak_x \cos \tfrac{1}{2}ak_y \cos \tfrac{1}{2}ck_z) \\ & - \sin \tfrac{1}{2}aQ_x \sin \tfrac{1}{2}cQ_z \sin \tfrac{1}{2}ak_z \cos \tfrac{1}{2}ak_y \sin \tfrac{1}{2}ck_z] \\ & - \sin \tfrac{1}{2}aQ_y \sin \tfrac{1}{2}ak_y[\sin \tfrac{1}{2}aQ_x \cos \tfrac{1}{2}cQ_z \sin \tfrac{1}{2}ak_x \cos \tfrac{1}{2}ck_z \\ & + \cos \tfrac{1}{2}aQ_x \sin \tfrac{1}{2}cQ_z \cos \tfrac{1}{2}ak_x \sin \tfrac{1}{2}ck_z]\} \\ & + j_3 \cos cQ_z(1 - \cos ck_z). \end{aligned} \tag{18}$$

We focus our interest on the model with $j_3 = 0$, $-1 < j_2 < 1$ where $Q_x = Q_y = \pi/a$ and Q_z is arbitrary. We stress that this is a region of physical interest, because we expect small values of j_2 and j_3. In the classical approximation ($S \to \infty$) we have found that the in-plane correlation is antiferromagnetic but any inter-plane phase relationship is allowed. An analogous behaviour is found in the classical RAF model [10–12], where an effective competition is entered into by the lattice structure that claims, for a 120°-three-sublattice, an in-plane configuration, but where the inter-plane coupling would prefer a collinear inter-plane configuration, so that a *full frustration* appears. In that model we have found [11] that the zero-point motion chooses a particular helix belonging to the infinite degeneration classical set: *order is established by quantum disorder*.

Table 1. Values of zero-point motion energy $\Delta(Q_z)$ for $Q_z = 0$ and $Q_z = \pi/c$ at selected values of j_2.

j_2	$\Delta(0)$	$\Delta(\pi/c)$
0	1.68411	1.68411
0.1	1.72193	1.72260
0.2	1.75714	1.75993
0.3	1.78968	1.79620
0.4	1.81944	1.83150
0.5	1.84622	1.86593
0.6	1.86971	1.89954
0.7	1.88943	1.93240
0.8	1.90453	1.96455
0.9	1.91335	1.99606
1	1.90994	2.02695

Here we search for an analogous phenomenon in the BCT antiferromagnet. The reduced ground-state energy is

$$e_G = E_G/2|J_1|S^2N = e_0(1 + 1/S) + (1/S)\Delta \tag{19}$$

where $e_0 = -2$ and Δ is given by

$$\Delta = \frac{1}{2\pi^3}\int_0^\pi\int_0^\pi\int_0^\pi \mathrm{d}x\,\mathrm{d}y\,\mathrm{d}z [\![\{s[d_1 - 4j_2d_2\cos(cQ_z/2)]\}^{1/2} + \{s[d_1 + 4j_2d_2\cos(cQ_z/2)]\}^{1/2}]\!] \tag{20}$$

where

$$s = 2 + \cos x + \cos y + 4j_2\cos\tfrac{1}{2}x\cos\tfrac{1}{2}y\cos\tfrac{1}{2}z \tag{21}$$

$$d_1 = 2 - \cos x - \cos y \tag{22}$$

$$d_2 = \sin\tfrac{1}{2}x\sin\tfrac{1}{2}y\cos\tfrac{1}{2}z. \tag{23}$$

In table 1 we give the value of Δ with $Q_z = 0$ and $Q_z = \pi/c$ for selected values of j_2. Notice that Δ assumes its minimum value for $Q_z = 0$ and its maximum value for $Q_z = \pi/c$. In the small j_2 limit it is possible to expand Δ in a series of powers of j_2 as follows

$$\Delta(Q_z) = a_0 + a_1j_2 - a_2j_2^2 - b_2j_2^2\cos^2(cQ_z/2) \tag{24}$$

where

$$a_0 = \frac{1}{\pi^2}\int_0^\pi\int_0^\pi \mathrm{d}x\,\mathrm{d}y(s_1d_1)^{1/2} \tag{25}$$

$$a_1 = \frac{4}{\pi^3}\int_0^\pi\int_0^\pi \mathrm{d}x\,\mathrm{d}y\,(d_1/s_1)^{1/2}\cos(x/2)\cos(y/2) \tag{26}$$

$$a_2 = \frac{1}{\pi^2}\int_0^\pi\int_0^\pi \mathrm{d}x\,\mathrm{d}y\,(d_1/s_1)^{1/2}\frac{\cos^2(x/2)\cos^2(y/2)}{s_1} \tag{27}$$

$$b_2 = \frac{1}{\pi^2}\int_0^\pi\int_0^\pi \mathrm{d}x\,\mathrm{d}y\,(s_1/d_1)^{1/2}\frac{\sin^2(x/2)\sin^2(y/2)}{d_1}. \tag{28}$$

s_1 and d_1 are given by (21) with $j_2 = 0$ and by (22), respectively. The modulation of $\Delta(Q_z)$, which appears at order j^2_{00442}, selects the helix with $Q_z = 0$. The numerical value of b_2 is 0.065062, so that $\Delta(\pi/c) - \Delta(0)$ evaluated by (24)–(28) differs from the exact one (see table 1) by a few per cent until $j_2 \simeq 0.2$. For this reason we may conclude that (24) is a realistic approximation for a large number of tetragonal antiferromagnets as, for instance, La_2CuO_4 [13]. Notice that the zero-point motion favours the AF1 configuration ($Q_x = Q_y = \pi/a$, $Q_z = 0$) that in the classical approximation should be stable for $j_3 < 0$, whereas for $j_3 > 0$ the AF configuration with $Q_x = Q_y = \pi/a$, $Q_z = \pi/c$ would become stable. This indicates an enhancement due to quantum fluctuations of the stability region of the AF1 configuration at the expense of the AF configuration. Unfortunately the evaluation of the zero-point motion energy as function of the $\boldsymbol{Q}$ wavevector away from the degeneration line is impossible because the magnon energy $\hbar\omega_k$ is well defined only for the classical helix wave vector $\boldsymbol{Q}_c$. This is a well known limitation of the perturbation approach for helix configurations [15]. Anyway, we stress the relevance of quantum fluctuations that are able to establish long-range order (LRO) in tetragonal antiferromagnets, so that it is not necessary to invoke the low-temperature orthorhombic distortion to justify LRO in La_2CuO_4 [16]. Even if the NN inter-plane exchange coupling J_2 had only one value, as for the tetragonal structure, the LRO should be given by quantum fluctuations. Notice that in La_2CuO_4 quantum corrections are expected to be important owing to the small value of the spin.

We next study the low-temperature thermodynamics of the BCT antiferromagnet with $j_3 = 0$ and small j_2. The reduced energy is given by

$$f(Q_z) = e_0\left(1 + \frac{1}{S}\right) + \frac{1}{S}\Delta(Q_z) + \frac{k_B T}{2|J_1|S^2 N}\sum_k \ln\left[1 - \exp\left(-\frac{\hbar\omega_k}{k_B T}\right)\right] \tag{29}$$

In the low temperature limit we have

$$f(Q_z) = -2(1 + 1/S) + (1/S)\Delta(Q_z) - (\zeta(3)/S)(k_B T/4|J|S)^3 g(Q_z) \tag{30}$$

where

$$g(Q_z) = \frac{1}{2\pi^2}\int_0^\pi dz \left[\left(1 + j_2 \cos\frac{z}{2}\right)^{-1}\left(1 - j_2^2 \cos\frac{cQ_z}{2}\cos^2\frac{z}{2}\right)^{-1/2}\right. \tag{31}$$

$$\left. + \left(1 - j_2^2 \cos^2\frac{cQ_z}{2}\cos^2\frac{z}{2}\right)^{-1}\left(1 - j_2^2\cos^2\frac{z}{2}\right)^{-1/2}\right]. \tag{31}$$

For small j_2, $g(Q_z)$ reduces to

$$g(Q_z) = 1/\pi[1 - (j_2/\pi) - (\tfrac{1}{4}j_2^2) - \tfrac{3}{8}j_2^2 \cos^2(\tfrac{1}{2}cQ_z)] \tag{32}$$

and

$$f(Q_z) = -2(1 + 1/S) + (1/S)[a_0(T) + a_1(T)j_2 - a_2(T)j_2^2 - b_2(T)j_2^2\cos^2(\tfrac{1}{2}cQ_z)] \tag{33}$$

where

$$a_0(T) = a_0 - (\zeta(3)/\pi)(k_B T/4|J_1|S)^3 \tag{34}$$

$$a_1(T) = a_1 + (\zeta(3)/\pi^2)(k_B T/4|J_1|S)^3 \tag{35}$$

$$a_2(T) = a_2 - (\zeta(3)/4\pi)(k_B T/4|J_1|S)^3 \tag{36}$$

$$b_2(T) = b_2 - (3\zeta(3)/8\pi)(k_B T/4|J_1|S)^3 \tag{37}$$

where a_0, a_1, a_2, b_2 are given by (25)–(28) and $\zeta(3) = 1.202$.

Notice that the minimum of the reduced free energy (33) corresponds to $Q_z = 0$ or $Q_z = \pi/c$ depending on the sign of $b_2(T)$, which is positive at low temperature and becomes negative at higher temperature. Indeed (37) may be written

$$b_2(T) = 0.065062 - 0.14348(k_B T/4|J_1|S)^3. \tag{38}$$

Equation (38) indicates that $Q_z = 0$ is stable for $T < 1.53653(2|J_1|S/k_B)$, that is to say, the stable phase corresponds to $Q_z = \pi/c$. Notice that this value of T is of the order of the NN in-plane exchange coupling so that it is quite large. In La_2CuO_4, for instance, $2|J_1| \simeq 1300$ K, $S = \frac{1}{2}$, so that the temperature at which the free energy (33) becomes Q_z-independent would be $T \simeq 1000$ K, which is much higher than the observed $T_N \simeq 300$ K. This means that the AF1 configuration ($Q_z = 0$) is well established by quantum effects and the speculation about LRO entered by orthorhombic distortion is unnecessary [16]. The surprising stability of the AF1 configuration supported by quantum fluctuations, which are generally expected to be negligible as for magnetic order, can be compared with an analogous effect we have found in the RAF model [12]. In addition, in the rhombohedral antiferromagnet full frustration destroys the LRO in the classical approximation and the LRO is restored by the zero-point motion, the tetragonal and the rhombohedral antiferromagnets, however, show a different thermal behaviour. Indeed, in the RAF model we have found that thermal fluctuations contribute to the free energy as $j^2(k_B T/|J_1|)^3$, while the zero-point energy is of order j^3 where $j = |J'/J_1|$, J_1 and J' are the in-plane and out-of-plane interactions, respectively. The temperature at which the free energy becomes Q_z-independent is consequently of order $j^{1/3}$, so that it can be quite small and a typical behaviour due to the infinite degeneracy of the classical approximation can appear in the intermediate temperature range. This possibility is ruled out for the tetragonal model because the scale of temperature is determined by the in-plane exchange interaction J_1.

We have obtained the zero-temperature phase diagram of the BCT antiferromagnet in the classical approximation. Infinite degeneracy is found for the physically interesting range of parameters $-1 < j_2 < 1, j_3 = 0$. In this range LRO is destroyed by a catastrophic magnon population number corresponding to the soft lines where $\boldsymbol{k} \in \mathcal{L}_Q$, $\mathcal{L}_Q$ being the locus of the infinite wavevectors $\boldsymbol{Q}$ characterising the infinite inequivalent isoenergetic helices that minimise the energy of the model in the classical approximation. We have found that the zero-point motion enters a modulation in the ground-state energy, establishing the AF1 configuration corresponding to $Q_z = Q_y = \pi/a$, $Q_z = 0$, We have found that this order due to quantum disorder is quite stable against thermal fluctuations. This result is of particular interest as concerns the magnetic order of La_2CuO_4 where the AF1 magnetic order can be understood as a consequence of quantum effects, whereas orthorhombic distortion could play a minor role.

Finally we stress the interest of performing MC simulation for the study of the critical behaviour of this model. Work of this kind has already been performed in the classical approximation and interesting critical properties were found, neglecting the in-plane exchange coupling J_1 [8, 9]. We stress that J_1 can be quite relevant, as is the case in La_2CuO_4. Moreover, the BCT antiferromagnet for $-1 < j_2 < 1, j_3 = 0$ is very sensitive to quantum effects that are generally neglected both in numerical and in renormalisation group analysis. Our model could show very interesting critical behaviour of a quantum nature.

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