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Frustration and zero-point motion in Heisenberg body-centred tetragonal antiferromagnets

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Abstract. Body-centred tetragonal (BCT) antiferromagnets are a central topic of both theoretical and experimental research. The Heisenberg BCT antiferromagnet shows full frustration induced by the lattice structure for all physically meaningful values of the exchange parameters. A previous analysis of the minimum-energy configuration indicated full frustration only if no coupling between next-nearest-neighbour planes is present. However, the analysis was restricted to a classical helix configuration whereas infinite non-helical configurations characterized by an arbitrary angle between the spin at the centre and the spins at the corner minimize the classical energy of the model in a substantial part of the parameter space. In any case the relevance of quantum fluctuations in solving the infinite degeneracy is confirmed. The first quantum correction to the magnon energy spectrum is shown to lift the degeneracy of the soft lines which are present in the classical approximation.

1. Introduction

Interesting anomalies in the behaviour of Heisenberg models are caused by the lattice structure itself. Rhombohedral [1] and body-centred tetragonal (BCT) [2] classical antiferromagnets show full frustration of the minimum-energy configuration. This interesting scenario was found by looking for helical configurations, but this approach does not exhaust all possible spin patterns. Indeed we show that important degrees of freedom can be neglected if one considers only helix configurations. Here we focus particularly on the BCT antiferromagnet where we find that the angle between the spin at the centre and the spins at the corners of the elementary cell is not determined for any meaningful exchange coupling. Note that such configurations are not helix configurations.

A similar result was found by Shender [3] for spin models suitable for describing some antiferromagnetic garnets.

Analysis of the zero-point motion effect is obviously useful. We confirm the conclusion obtained previously [2] (where only helical configurations were taken into account) that a particular antiferromagnetic configuration is stabilized by quantum fluctuations. Taking the arbitrary angle between the spin sublattices into account does not change our previous conclusions [2].

Our approach differs from that of Shender [3] because we account for the first quantum correction in $1/S$ to *all* orders in the inter-sublattice coupling while Shender evaluates second-order contribution only. In any case our result agrees substantially with that of Shender as concerns the quantum correction to the ground state. As for quantum corrections to the magnon spectrum we find that there is lifting of the degeneracy of the soft lines but that the soft modes at the zone centre and at the zone boundary are preserved in agreement with Goldstone's non-relativistic theorem proved for helimagnets [4].

The paper has the following format: section 2 is devoted to the evaluation of the minimum-energy configurations of the BCT antiferromagnet. In section 3 we study spin waves in the classical approximation. Section 4 concerns the zero-point motion correction of the ground-state energy and of the magnetic moment as well as the temperature dependence of the magnetization. Finally, section 5 contains a summary and concluding remarks.

2. Minimum-energy configurations of the BCT antiferromagnet

Let us consider N spins localized on the sites of a BCT lattice. We look for configurations where the spins at the centre and at the corners make up two helix sublattices, the angle θ between the spin at the centre and those at the corners being arbitrary. The exchange interactions that we consider are an in-plane nearest-neighbour coupling J_1, an inter-sublattice next-nearest-neighbour (NNN) coupling J_2 and a third-nearest-neighbour out-of-plane coupling J_3 between spins along the c axis. The Hamiltonian of our model reads

$$\mathcal{H} = -J_1 \sum_{i,\boldsymbol{\delta}_1} (\boldsymbol{S}_i^{(a)} \cdot \boldsymbol{S}_{i+\boldsymbol{\delta}_1}^{(a)} + \boldsymbol{S}_i^{(b)} \cdot \boldsymbol{S}_{i+\boldsymbol{\delta}_1}^{(b)}) - 2J_2 \sum_{i,\boldsymbol{\delta}_2} \boldsymbol{S}_i^{(a)} \cdot \boldsymbol{S}_{i+\boldsymbol{\delta}_2}^{(b)} - J_3 \sum_{i,\boldsymbol{\delta}_3} (\boldsymbol{S}_i^{(a)} \cdot \boldsymbol{S}_{i+\boldsymbol{\delta}_3}^{(a)} + \boldsymbol{S}_i^{(b)} \cdot \boldsymbol{S}_{i+\boldsymbol{\delta}_3}^{(b)}) \tag{2.1}$$

where a and b label the two tetragonal interpenetrating sublattices, i runs over the sites of each tetragonal sublattice, $\boldsymbol{\delta}_\alpha$ joins site i with the neighbours of the αth shell.

We allow for possible helical order of wavevector $\boldsymbol{Q}$ in each sublattice by the introduction of local spiralling axes as follows:

$$\begin{aligned} S_i^x(a) &= -S_i^\eta(a)\sin(\boldsymbol{Q}\cdot\boldsymbol{r}_i) + S_i^\zeta(a)\cos(\boldsymbol{Q}\cdot\boldsymbol{r}_i) \\ S_i^y(a) &= S_i^\eta(a)\cos(\boldsymbol{Q}\cdot\boldsymbol{r}_i) + S_i^\zeta(a)\sin(\boldsymbol{Q}\cdot\boldsymbol{r}_i) \\ S_i^z(a) &= -S_i^\xi(a) \end{aligned} \tag{2.2}$$

and

$$\begin{aligned} S_i^x(b) &= -S_i^\eta(b)\sin(\boldsymbol{Q}\cdot\boldsymbol{r}_i + \theta) + S_i^\zeta(b)\cos(\boldsymbol{Q}\cdot\boldsymbol{r}_i + \theta) \\ S_i^y(b) &= S_i^\eta(b)\cos(\boldsymbol{Q}\cdot\boldsymbol{r}_i + \theta) + S_i^\zeta(b)\sin(\boldsymbol{Q}\cdot\boldsymbol{r}_i + \theta) \\ S_i^z(b) &= -S_i^\xi(b). \end{aligned} \tag{2.3}$$

We are interested in finding the minimum-energy configurations and low-lying excitations. To this aim we transform the spin Hamiltonian to the bosonic equivalent one, where we retain bilinear contributions only, since this is sufficient to evaluate the zero-temperature phase diagram and magnon excitation spectrum in the classical approximation ($S \rightarrow \infty$) as well as the leading quantum corrections. The bilinear equivalent

Hamiltonian that we obtain is

$$\mathcal{H} = E_0 + \sum_k A_k(a_k^\dagger a_k + b_k^\dagger b_k) + \sum_k \tfrac{1}{2}B_k(a_k^\dagger a_{-k}^\dagger + a_k a_{-k} + b_k^\dagger b_{-k}^\dagger + b_k b_{-k})$$
$$+ \sum_k (D_k a_k^\dagger b_{-k}^\dagger + D_k^* a_k b_{-k}) + \sum_k (C_k a_k b_k^\dagger + C_k^* a_k^\dagger b_k) \qquad (2.4)$$

where a_k and b_k are Bose destruction operators of the spin wave of momentum $\boldsymbol{k}$ on the a and b sublattices, respectively, and E_0, A_k, B_k, C_k, D_k are functions of the variational parameters $\boldsymbol{Q}$ and θ given by

$$E_0 = -J_1NS^2 \sum_{\delta_1} \cos(\boldsymbol{Q}\cdot\boldsymbol{\delta}_1) - J_2NS^2\cos\theta \sum_{\delta_2}\cos(\boldsymbol{Q}\cdot\boldsymbol{\delta}_2) - J_3NS^2\sum_{\delta_3}\cos(\boldsymbol{Q}\cdot\boldsymbol{\delta}_3) \qquad (2.5)$$

$$A_k = 2J_1S\sum_{\delta_1}\{\cos(\boldsymbol{Q}\cdot\boldsymbol{\delta}_1) - \tfrac{1}{2}\cos(\boldsymbol{k}\cdot\boldsymbol{\delta}_1)[1+\cos(\boldsymbol{Q}\cdot\boldsymbol{\delta}_1)]\} + 2J_2S\cos\theta\sum_{\delta_2}\cos(\boldsymbol{Q}\cdot\boldsymbol{\delta}_2)$$
$$+ 2J_3S\sum_{\delta_3}\{\cos(\boldsymbol{Q}\cdot\boldsymbol{\delta}_3) - \tfrac{1}{2}\cos(\boldsymbol{k}\cdot\boldsymbol{\delta}_3)[1+\cos(\boldsymbol{Q}\cdot\boldsymbol{\delta}_3)]\} \qquad (2.6)$$

$$B_k = -J_1S\sum_{\delta_1}\cos(\boldsymbol{k}\cdot\boldsymbol{\delta}_1)[1+\cos(\boldsymbol{Q}\cdot\boldsymbol{\delta}_1)] - J_3S\sum_{\delta_3}\cos(\boldsymbol{Q}\cdot\boldsymbol{\delta}_3)[1-\cos(\boldsymbol{Q}\cdot\boldsymbol{\delta}_3)] \qquad (2.7)$$

$$C_k = -J_2S\sum_{\delta_2}\{\cos(\boldsymbol{Q}\cdot\boldsymbol{\delta}_2)[1+\cos\theta\cos(\boldsymbol{Q}\cdot\boldsymbol{\delta}_2)] + \mathrm{i}\sin(\boldsymbol{k}\cdot\boldsymbol{\delta}_2)\sin\theta\sin(\boldsymbol{Q}\cdot\boldsymbol{\delta}_2)\} \qquad (2.8)$$

$$D_k = -J_2S\sum_{\delta_2}\{\cos(\boldsymbol{k}\cdot\boldsymbol{\delta}_2)[1-\cos\theta\cos(\boldsymbol{Q}\cdot\boldsymbol{\delta}_2)] + \mathrm{i}\sin(\boldsymbol{k}\cdot\boldsymbol{\delta}_2)\sin\theta\sin(\boldsymbol{Q}\cdot\boldsymbol{\delta}_2)\}. \qquad (2.9)$$

In a previous communication [2] we assumed that $\theta = 0$ which oversimplifies the problem, because we find that this degree of freedom is crucial at least for classical systems. Let us introduce the reduced classical energy

$$e_0 \equiv E_0/2|J_1|NS^2 = \cos Q_x + \cos Q_y$$
$$+ 4j_2\cos\theta\cos(\tfrac{1}{2}Q_x)\cos(\tfrac{1}{2}Q_y)\cos(\tfrac{1}{2}Q_z) + j_3\cos Q_z \qquad (2.10)$$

where the in-plane lattice constant and the distance between two NNN layers are assumed to be unity ($a = c = 1$) and $j_\alpha = J_\alpha/J_1$. The minimum-energy conditions in the classical approximation are

$$\sin(\tfrac{1}{2}Q_x)[\cos(\tfrac{1}{2}Q_x) + j_2\cos\theta\cos(\tfrac{1}{2}Q_y)\cos(\tfrac{1}{2}Q_z)] = 0 \qquad (2.11a)$$
$$\sin(\tfrac{1}{2}Q_y)[\cos(\tfrac{1}{2}Q_y) + j_2\cos\theta\cos(\tfrac{1}{2}Q_x)\cos(\tfrac{1}{2}Q_z)] = 0 \qquad (2.11b)$$
$$\sin(\tfrac{1}{2}Q_z)[j_2\cos\theta\cos(\tfrac{1}{2}Q_x)\cos(\tfrac{1}{2}Q_y) + j_3\cos(\tfrac{1}{2}Q_z)] = 0 \qquad (2.11c)$$
$$j_2\sin\theta\cos(\tfrac{1}{2}Q_x)\cos(\tfrac{1}{2}Q_y)\cos(\tfrac{1}{2}Q_z) = 0. \qquad (2.11d)$$

If one assumes that $\theta = 0$, *ab initio* equations (2.11) reduce to equations (4) of [2]. Note that equations (4*a*) and (4*b*) of [2] contain a misprint: a factor of 2 has to be omitted.

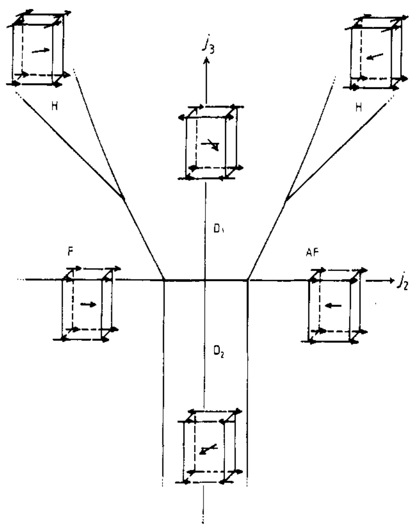

Figure 1. Zero-temperature phase diagram of the classical BCT antiferromagnet in the j_2–j_3 plane. The angle θ between the spin at the centre and the spin at the corner is arbitrary in the D_1 and D_2 phases.

The zero-temperature phase diagram is shown in figure 1. The phase diagram of the BCT antiferromagnet consists of the same six regions as in [2] where the AF and AF_1 phases are replaced by the new *degenerate* phases D_1 and D_2, respectively. The degenerate phase D_1 is characterized by $Q_x = Q_y = Q_z = \pi$ and θ *arbitrary*. Its reduced energy is $e_{D_1} = -2 - j_3$. Note that the AF configuration in [2] is one of the infinite configurations of the D_1 configuration corresponding to the choice $\theta = 0$.

The degenerate phase D_2 is characterized by $Q_x = Q_y = \pi$, $Q_z = 0$ and θ *arbitrary*. The reduced energy of this phase is $e_{D_2} = -2 + j_3$. The AF_1 phase in [2] is one of the infinite configurations of D_2 corresponding to $\theta = 0$.

Note that for $j_3 = 0$ there is the inset of a new degeneracy because in that case also the z component of the helix wavevector becomes arbitrary. In the D_1 and D_2 phases the a and b sublattices are decoupled while for $j_3 = 0$ *each* plane is decoupled from any other. The H, F and AF phases are the same as the H, F and AF_2 phases in [2].

Full frustration caused by the lattice structure and (or) suitable competition of the exchange interactions was found in a number of classical Heisenberg models on periodic lattices. Localized spins on tetragonal, hexagonal [5], face-centred cubic and rhombohedral [1] lattices can support disordered configurations in addition to the well known collinear or helical ones. In the minimum-energy configuration, spins can explore, with zero energy cost, infinite inequivalent configurations that lead to the absence of long-range order (LRO) in 3D Heisenberg models at any finite temperature. These classical Heisenberg models behave like *liquids* even if spins lie on the sites of a perfectly periodic lattice. There is no *condensation* into an ordered spin pattern at any finite temperature and the only kind of order that one can see is short ranged.

The BCT antiferromagnet shows a similar behaviour for $j_3 = 0$ and $-1 < j_2 < 1$. Note that the arbitrary angle between the two antiferromagnetic sublattices for $j_3 \neq 0$ does not destroy LRO. On the contrary, full frustration appears for $J_3 = 0$ because in this case the magnon dispersion curve shows *soft lines* that lead to a catastrophically huge number of spin deviations.

Note that the arbitrary angle θ between the two sublattices of the classical BCT antiferromagnet would show itself in the Bragg peak intensity. Indeed the intensity of the $(2h + 1, 2k, 2l)$ and $(2h, 2k + 1, 2l + 1)$ peaks is proportional to $1 + \cos\theta$ while the intensity of the $(2h, 2k + 1, 2l)$ and $(2h + 1, 2k, 2l + 1)$ peaks is proportional to

$1 - \cos\theta$. For $\theta = 0$ or $\theta = \pi$, one half of the peaks disappear. This scenario occurs for a single-domain sample, whereas the simultaneous presence of all values of θ in multiple-domain samples leads to Bragg peaks of equal intensity. These peculiarities are related to classical spins so that, in real compounds, elastic scattering profiles as described above might appear in an intermediate range of temperatures where the quantum nature of the spins is expected to play a minor role.

3. Classical spin waves in BCT antiferromagnets

In this section we diagonalize the bilinear bosonic equivalent Hamiltonian (2.4) for exchange couplings supporting the D_2 configuration, which is suitable for describing the high-T_c superconductor La_2CuO_4 [6]. For this choice equations (2.5)–(2.9) become

$$A_k = 8|J_1|S[1 - \tfrac{1}{2}j_3(1 - \cos k_z)] \quad (3.1)$$

$$B_k = 4|J_1|S(\cos k_x + \cos k_y) \quad (3.2)$$

$$C_k = 8|J_1|Sj_2 \cos(\tfrac{1}{2}k_z)[\cos(\tfrac{1}{2}k_z)\cos(\tfrac{1}{2}k_x)\cos(\tfrac{1}{2}k_y) + \cos\theta \sin(\tfrac{1}{2}k_x)\sin(\tfrac{1}{2}k_y)] \quad (3.3)$$

$$D_k = 8|J_1|Sj_2 \cos(\tfrac{1}{2}k_z)[\cos(\tfrac{1}{2}k_z)\cos(\tfrac{1}{2}k_x)\cos(\tfrac{1}{2}k_y) - \cos\theta \sin(\tfrac{1}{2}k_x)\sin(\tfrac{1}{2}k_y)]. \quad (3.4)$$

To diagonalize the Hamiltonian (2.4) the following Bogoliubov transformation is required:

$$\alpha_k = u_k a_k + l_k a^{\dagger}_{-k} - m_k b_k - v_k b^{\dagger}_{-k} \quad (3.5)$$

$$\beta_k = r_k b_k + t_k b^{\dagger}_{-k} - w_k a_k - s_k a^{\dagger}_{-k} \quad (3.6)$$

instead of the usual Bogoliubov transformation that involves only two boson operators. Equations (3.5) and (3.6) reduce the Hamiltonian (2.4) to the diagonal form

$$\mathscr{H}_0 = E_0 + \Delta E + \sum_k E_k^+ \alpha_k^{\dagger}\alpha_k + \sum_k E_k^- \beta_k^{\dagger}\beta_k \quad (3.7)$$

where E_k^+ and E_k^- are given by

$$(E_k^{\pm})^2 = (A_k \pm C_k)^2 - (B_k \pm D_k)^2 \quad (3.8)$$

and the leading quantum correction ΔE to the classical ground-state energy is

$$\Delta E = -\sum_k A_k + \tfrac{1}{2}\sum_k (E_k^+ + E_k^-). \quad (3.9)$$

In order to obtain the diagonal Hamiltonian (3.7) the coefficients of the Bogoliubov transformation have to be chosen as

$$u_k = \sqrt{(E_k^+ + A_k + C_k)/4E_k^+} \qquad m_k = -u_k \quad (3.10)$$

$$l_k = \sqrt{(-E_k^+ + A_k + C_k)/4E_k^+} \qquad v_k = -l_k \quad (3.11)$$

$$r_k = \sqrt{(E_k^- + A_k - C_k)/4E_k^-} \qquad w_k = r_k \quad (3.12)$$

$$t_k = \sqrt{(-E_k^- + A_k - C_k)/4E_k^-} \qquad s_k = t_k. \quad (3.13)$$

It is interesting to evaluate $E_k^{\mp}$ along the lines $(0, 0, k_z)$ and (π, π, k_z) where

$$E_k^{\mp}(0,0,k_z) = 8|J_1|S\sqrt{|j_3|(1-\cos k_z)[1 \pm j_2 \cos(\tfrac{1}{2}k_z)]} \tag{3.14}$$

$$E_k^{\mp}(\pi,\pi,k_z) = 8|J_1|S\sqrt{|j_3|(1-\cos k_z)[1 \pm j_2 \cos\theta \cos(\tfrac{1}{2}k_z)]}. \tag{3.15}$$

It is obvious that for $j_3 = 0$ the magnon energy spectrum develops *soft lines* that disappear for $j_3 \neq 0$ even if soft modes remain at the zone centre $(0, 0, 0)$ and at the zone boundary $(\pi, \pi, 0)$ in agreement with Goldstone's non-relativistic theorem extended to helimagnets [4].

4. Quantum and thermal fluctuations in BCT antiferromagnets

In this section we look at the effect of quantum fluctuations on the arbitrary angle θ between the two antiferromagnetic sublattices we have found in the D_2 phase. We focus on the D_2 phase because the magnetic structure observed in pure La_2CuO_4 [6] corresponds to that D_2 configuration with $\theta = 0$.

We find that the zero-point motion supports the order that we called AF_1 in [2] since it selects the angle $\theta = 0$. The ground-state energy obtained summing the leading quantum contribution to the classical energy is

$$E_G = E_0 + \Delta E = 2|J_1|NS^2[(-2 + j_3)(1 + 1/S) + \Delta/S] \tag{4.1}$$

where

$$\Delta = \frac{1}{4\pi^3}\int_{-\pi}^{\pi}\int_{-\pi}^{\pi}\int_{-\pi}^{\pi} dx\,dy\,dz\,\tfrac{1}{2}[\varepsilon^+(x,y,z) + \varepsilon^-(x,y,z)] \tag{4.2}$$

with

$$\varepsilon^{\pm}(x,y,z) = \sqrt{(a \pm c_1 \pm c_2\cos\theta)^2 - (b \pm c_1 \mp c_2 \cos\theta)^2} \tag{4.3}$$

$$a = 1 + \tfrac{1}{2}(1 - \cos z) \tag{4.4}$$

$$b = \tfrac{1}{2}(\cos x + \cos y) \tag{4.5}$$

$$c_1 = j_2 \cos(\tfrac{1}{2}x)\cos(\tfrac{1}{2}y)\cos(\tfrac{1}{2}z) \tag{4.6}$$

$$c_2 = j_2 \sin(\tfrac{1}{2}x)\sin(\tfrac{1}{2}y)\cos(\tfrac{1}{2}z). \tag{4.7}$$

We find that for any $j_3 \leqslant 0$ the AF_1 configuration is established because $\theta = 0$ or $\theta = \pi$ minimizes the zero-point motion energy. This can be seen analytically, by expanding Δ for small j_2 and j_3. In particular, for $j_3 = 0$, one obtains

$$\Delta = a_1 - a_2(1 + \cos^2\theta)j_2^2 \tag{4.8}$$

where

$$a_1 = \frac{8}{\pi^2}\int_0^{\pi/2}\int_0^{\pi/2} dx\,dy\,\sqrt{(\sin^2 x + \sin^2 y)(\cos^2 x + \cos^2 y)} = 1.684\,105\,2 \tag{4.9}$$

$$a_2 = \frac{2}{\pi^2}\int_0^{\pi/2}\int_0^{\pi/2} dx\,dy\,\frac{(\sin^2 x + \sin^2 y)^{1/2}}{(\cos^2 x + \cos^2 y)^{3/2}}\sin^2 x \sin^2 y = 0.065\,062\,6. \tag{4.10}$$

Equation (4.8) should be compared with equation (14) of [3] where only second-order contributions in j_2 have been accounted for.

We have also evaluated the zero-temperature spin reduction obtained from the mean value of the spin component along the local quantization axis:

$$\langle S_i^\zeta \rangle = S - \Delta S - \Delta S(T) \tag{4.11}$$

where

$$\Delta S = \frac{1}{N}\sum_k \left(\frac{A_k + C_k - E_k^+}{2E_k^+} + \frac{A_k - C_k - E_k^-}{2E_k^-} \right) \tag{4.12}$$

$$\Delta S(T) = \frac{1}{N}\sum_k \left(\frac{A_k + C_k}{E_k^+} \langle \alpha_k^\dagger \alpha_k \rangle + \frac{A_k - C_k}{E_k^-} \langle \beta_k^\dagger \beta_k \rangle \right) \tag{4.13}$$

with

$$\langle \alpha_k^\dagger \alpha_k \rangle = 1/[\exp(E_k^+/k_B T) - 1] \qquad \langle \beta_k^\dagger \beta_k \rangle = 1/[\exp(E_k^-/k_B T) - 1]. \tag{4.14}$$

Using equations (4.3)–(4.6) the zero-temperature spin reduction (4.12) reduces to

$$\Delta S = \frac{1}{(2\pi)^3} \int_{-\pi}^{\pi} \int_{-\pi}^{\pi} \int_{-\pi}^{\pi} \mathrm{d}x\, \mathrm{d}y\, \mathrm{d}z \left(\frac{a + c_1 + c_2 \cos\theta - \varepsilon^+}{4\varepsilon^+} + \frac{a - c_1 - c_2 \cos\theta - \varepsilon^-}{4\varepsilon^-} \right). \tag{4.15}$$

The finite value ΔS indicates that the zero-temperature coherent fluctuations do not destroy LRO. For $j_3 = 0$ and small j_2 we have

$$\Delta S = s_1 + (s_2 + s_3 \cos^2\theta) j_2^2 \tag{4.16}$$

where

$$s_1 = \frac{2}{\pi^2} \int_0^{\pi/2} \int_0^{\pi/2} \mathrm{d}x\, \mathrm{d}y \left(\frac{1}{\sqrt{(\sin^2 x + \sin^2 y)(\cos^2 x + \cos^2 y)}} - 1 \right) = 0.1965 \tag{4.17}$$

$$s_2 = \frac{1}{2\pi^2} \int_0^{\pi/2} \int_0^{\pi/2} \mathrm{d}x\, \mathrm{d}y \frac{(3 - 2\cos^2 x - 2\cos^2 y)\cos^2 x \cos^2 y}{(\sin^2 x + \sin^2 y)^{1/2} (\cos^2 x + \cos^2 y)^{5/2}} = 0.0182 \tag{4.18}$$

$$s_3 = \frac{2}{2\pi^2} \int_0^{\pi/2} \int_0^{\pi/2} \mathrm{d}x\, \mathrm{d}y \frac{(2\cos^2 x + 2\cos^2 y - 1)\sin^2 x \sin^2 y}{(\sin^2 x + \sin^2 y)^{5/2} (\cos^2 x + \cos^2 y)^{1/2}} = -0.0398. \tag{4.19}$$

Let us consider the effect of thermal fluctuations on the LRO within the simple spin-wave approximation. For small j_2 and j_3 we may evaluate analytically the temperature dependence of the magnetization (4.12) because for a wide range of temperatures one has

$$|j_3| \ll t \equiv k_B T / 8|J_1| S \ll 1 \tag{4.20}$$

so that the sum over k_x and k_y in (4.12) may be performed in the long-wavelength limit. When this is done, one obtains

$$\Delta S(T) = -\frac{2t}{\pi^2} \int_0^{\pi} \mathrm{d}z \ln\left[1 - \exp\left(-\frac{1}{t} \sqrt{2|j_3|} \sin(\tfrac{1}{2}z) \right) \right] \tag{4.21}$$

which for $|j_3| \ll 1$ becomes

$$\Delta S(T) = -(2t/\pi) \ln(\sqrt{|j_3|}/2t). \tag{4.22}$$

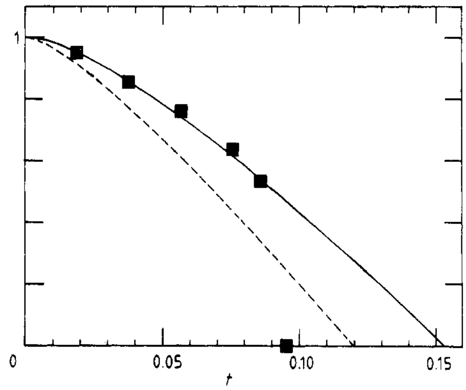

Figure 2. Reduced spontaneous magnetization m versus reduced temperature t for selected values of the intra-sublattice exchange coupling: ———, $J_3 = 10^{-4}|J_1|$; – – –, $J_3 = 10^{-5}|J_1|$; ■, experimental data of Budnick *et al.* [8].

It is easily seen that for $S = \frac{1}{2}$, $|j_3| = 10^{-5}$, the magnetization vanishes for $t_N \simeq 0.12$ which for $2|J_1| \simeq 1300$ K (a value suitable for La_2CuO_4) leads to a Néel temperature T_N of about 310 K. Note that the numerical value obtained from the simple spin-wave approach should be reliable because of the absence of renormalization of elementary excitations observed experimentally [7]. We would like to remark that the magnetization curve decreases linearly as shown by equation (4.22) in agreement with μ-spin rotation measurements by Budnick *et al* [8]. Figure 2 shows the thermal behaviour of the reduced spontaneous magnetization $m = 1 - \Delta S(T)/(S - \Delta S)$ versus the reduced temperature $t = k_B T/8|J_1|S$ for $j_3 = -10^{-4}$ (full curve) and for $j_3 = -10^{-5}$ (broken curve).

Note that simple spin-wave theory gives a clear indication of absence of LRO at any finite temperature if $j_3 = 0$. On the contrary, for any $j_3 \neq 0$ the logarithmic divergence in (4.22) is suppressed so that simple spin-wave theory suggests that any exchange coupling along the c axis leads to LRO, whereas LRO is absent when only coupling between adjacent planes J_2 is accounted for even if the zero-point motion energy picks a particular helix ($Q_x = Q_y = \pi$, $Q_z = 0$) out of the infinite helices which are isoenergetic in the classical approximation. However, the absence of LRO at any finite temperature for $j_3 = 0$ seems to be an artefact of the simple spin-wave approximation since the disappearance of the *soft lines* in the spectrum is expected as a consequence of higher-order quantum corrections. If so, no catastrophic population numbers related to the presence of soft lines are expected and LRO should appear again. Note that the disappearance of the soft lines in the elementary excitation spectrum as well as the removing of the ground-state infinite degeneracy is a necessary condition to have *order by quantum disorder*.

In the framework of a perturbation expansion in $1/S$, the spin-wave spectrum E_k^I containing the first-order quantum correction in the AF_1 configuration is given by [4]

$$(E_k^I)^2 = (E_k)^2\left[1+\frac{1}{S}\left(1-\frac{1}{N}\sum_q\frac{A_q}{E_q}\right)\right]-\frac{1}{S}\left(A_k\frac{1}{N}\sum_q\frac{1}{E_q}-\frac{1}{N}\sum_q D_{k-q}\frac{S_kS_q+D_kD_q}{2E_q}\right) \tag{4.23}$$

where

$$A_k = \tfrac{1}{2}(D_k + S_k) \tag{4.24}$$

$$D_k = 8|J_1|S[1 - \tfrac{1}{2}(\cos k_x + \cos k_y) + 2j_2 \sin(\tfrac{1}{2}k_x)\sin(\tfrac{1}{2}k_y)\cos(\tfrac{1}{2}k_z) + \tfrac{1}{2}|j_3|(1 - \cos k_z)] \quad (4.25)$$

$$S_k = 8|J_1|S[1 + \tfrac{1}{2}(\cos k_x + \cos k_y) + 2j_2 \sin(\tfrac{1}{2}k_x)\sin(\tfrac{1}{2}k_y)\cos(\tfrac{1}{2}k_z) + \tfrac{1}{2}|j_3|(1 - \cos k_z)]. \quad (4.26)$$

Note that equation (4.23) holds for collinear configurations whereas for helical configurations further perturbative contributions should be accounted for [4]. We evaluate E_k^I along the soft lines of E_k for $j_3 = 0$ and small j_2. We obtain

$$E_k^I(0, 0, k_z) = E_k^I(\pi, \pi, k_z) = 8|J_1|Sc_0(|j_2|/\sqrt{S})\sqrt{1 - \cos(\tfrac{1}{2}k_z)} \quad (4.27)$$

where

$$c_0 = \frac{1}{\pi^2}\int_0^\pi\int_0^\pi \mathrm{d}x\,\mathrm{d}y\,\frac{(\cos^2 x + \cos^2 y)^{1/2}}{(\sin^2 x + \sin^2 y)^{3/2}}\sin^2 x \sin^2 y = 0.130\,125\,2. \quad (4.28)$$

Equation (4.27) clearly shows that first-order quantum corrections remove soft lines so that no divergent occupation numbers are produced at a finite temperature. This seems to substantiate the hypothesis of the ordering effect by quantum fluctuations in systems that would be fully frustrated in the classical approximation. Such a suggestion [2] was advanced to explain the magnetic order in pure La_2CuO_4, because the ordering due to orthorhombic distortion seems insufficient to give a reliable value of the critical temperature [9]. Note that the raising of the degeneracy of the soft lines due to quantum fluctuations can be as relevant as the raising due to the spin–spin interaction J_3 along the c axis as one can see by comparison of equations (3.14) and (3.15) with equation (4.27). On the other hand we have shown that a weak interaction $|j_3| = 10^{-5}$ is sufficient to give a transition temperature $T_N \simeq 300$ K for parameter values suitable for La_2CuO_4 and the same could be obtained for $j_3 = 0$ taking into account the zero-point motion in the frame of the spin-wave theory including the leading quantum corrections into the magnon dispersion curve. Indeed for $|j_2| = 10^{-2}$, equations (3.14) and (4.27) become, respectively,

$$E_k^{\mp}(0, 0, k_z) \simeq 12\,\mathrm{K}|\sin(\tfrac{1}{2}k_z)| \quad (4.29)$$

$$E_k^I(0, 0, k_z) \simeq 7\,\mathrm{K}|\sin(\tfrac{1}{4}k_z)| \quad (4.30)$$

where K indicates kelvin.

5. Conclusions and comments

We have studied the Heisenberg BCT antiferromagnet in the classical approximation evaluating the zero-temperature phase diagram in the parameter space and the spin-wave normal modes. The interest in this model is due both to its rich phenomenology and to the possible insight into the magnetic properties of physical compounds such as La_2CuO_4 which becomes a high-T_c superconductor on suitable doping [6].

The present paper substantiates and improves the previous analysis [2] that looked at helical configurations only as possible ground-state configurations. Here we take into account the possibility of two interpenetrating sublattices whose magnetizations make an arbitrary angle θ. We find that this is the case for meaningful exchange parameters; θ is arbitrary for $-1 < j_2 < 1$. For zero coupling between spins of the same sublattice along the c axis ($j_3 = 0$), the BCT antiferromagnet shows an additional infinite degeneracy because the z component of the helix wavevector becomes arbitrary.

Evaluation of the elementary excitations allows us to estimate the order parameter in the classical approximation that suggests the existence of LRO for any $j_3 \neq 0$, whereas no LRO is expected if $j_3 = 0$ and $T \neq 0$. LRO should be present with $j_3 = 0$ only for $T = 0$, as indicated by a finite value of the spin reduction.

Quantum contributions are taken into account to leading order of $1/S$ for the D_2 phase that reduces to the configuration of pure La_2CuO_4 when the arbitrary angle θ between the two interpenetrating sublattices vanishes. We find that the zero-point motion energy forces θ to vanish for any $j_3 \leqslant 0$. This could be an ordering effect in addition to the orthorhombic distortion [9] for La_2CuO_4.

The evaluation of the leading quantum correction to the magnon energy seems to confirm such a hypothesis, because we find that degeneracies of the soft lines one has in the classical approximation for $j_3 = 0$ related to the arbitrary phase relation of spins along the c axis are lifted. It is worthwhile noting that this quantum lifting of the degeneracy of the soft lines is of the same order of magnitude as the lifting of the typical values of j_3.

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