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1994 J. Phys.: Condens. Matter 6 6535

(http://iopscience.iop.org/0953-8984/6/32/014)

J. Phys.: Condens. Matter 6 (1994) 6535–6542. Printed in the UK

Spin-wave theory of the phase transition induced by a magnetic field perpendicular to the CuO_2 plane in La_2CuO_4

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Received 31 January 1994, in final form 5 May 1994

Abstract. A spin-wave theory is developed for the canting antiferromagnetism by using the Holstein–Primakoff transformation. The correction arising from interactions between spin waves can be obtained easily in this theory. The theory is applied to study the field-induced transition in La_2CuO_4 when the magnetic field is perpendicular to the CuO_2 plane. The temperature dependence of the critical magnetic field predicted by our theory agrees fairly well with experiment at low temperatures. The correction terms are quite small at low temperatures, where the theory fits experiment well. This means that the simple theory, neglecting spin-wave interactions, is sufficient for practical purposes.

1. Introduction

La_2CuO_4 undergoes a transition from antiferromagnetic order to weak ferromagnetic order when a sufficiently large magnetic field H is applied perpendicular to the CuO_2 planes at a temperature below the Néel temperature T_N. This was first discovered by Thio *et al* [1] through measurements of the magnetic moment and the magnetoresistance as a function of magnetic field H and temperature T, and it was later confirmed by other research groups [2–4].

Thio *et al* [1] pointed out that their measurement demonstrates that La_2CuO_4 is a canting antiferromagnetism, and suggested the following picture for the observed magnetic field-induced transition in La_2CuO_4. Due to the Dzyaloshinskii–Moriya (DM) superexchange interaction, the spins of Cu^{2+} are canted out of the CuO_2 plane by a small angle. Each CuO_2 plane thus carries a ferromagnetic moment, but the net moment at $H = 0$ is zero because the antiferromagnetic interplanar coupling causes alternate planes to cant in opposite directions. However, above a critical field $H_c(T)$, the canting angles in all planes align in the same direction, and so a transition from antiferromagnetic to weak ferromagnetic order is observed. In their paper, Thio *et al* also developed a mean field theory for this field-induced transition. However, as Kastner *et al* [2] pointed out, the phase transition can be described by the theory of Thio *et al* close to the Néel temperature only.

In the present paper we try to treat this transition via a spin-wave theory. The spin-wave energy spectrum for the canting antiferromagnetism is derived in [3] and [5–7] by a procedure of linearizing the equations of motions for the spin operators with respect to small deviations around the classical ground state. In this paper we develop a spin-wave theory for the canting antiferromagnetism by using the Holstein–Primakoff (HP) transformation [7, 8], and study the field-induced transition in La_2CuO_4 when the magnetic field is applied perpendicular to the CuO_2 plane. Moreover, the spin-wave interactions have also been included (to first order in $1/S$).

0953-8984/94/326535+08$19.50

The paper is organized as follows. In section 2 we develop the spin-wave theory, with the model given in section 2.1 and the spin-wave theory for the canting antiferromagnetism using the HP method including the spin-wave interactions in section 2.2. Comparison between our theory and experiment, together with discussion, are in section 3.

2. Spin-wave theory

2.1. Model

The spins of Cu^{2+} in a CuO_2 plane form a square lattice. We choose the x axis to be perpendicular to the CuO_2 plane and the y and z axes to be lying in the plane, as shown in figure 1(a). We separate the lattice into sublattices A and B. As a model for our study, we consider a set of CuO_2 planes with weak antiferromagnetic interplanar coupling (figure 1(b)).

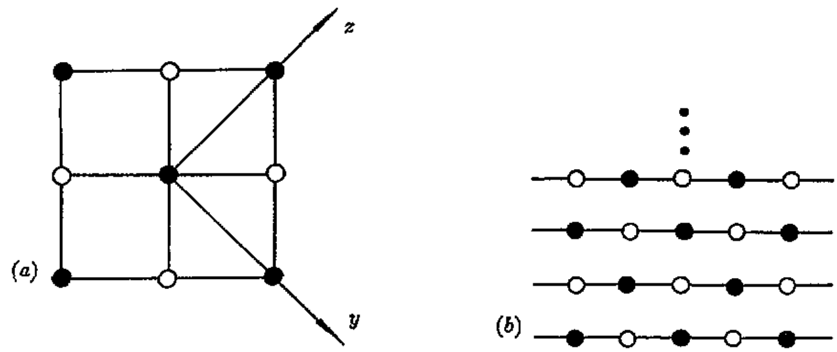

Figure 1. (a) CuO_2 plane. Only the Cu^{2+} spin sites are shown. (b) Sketch of the model used. The horizontal line refers to the CuO_2 plane. Full circle: sublattice A; open circle: sublattice B.

The Hamiltonian $\mathcal{H}$ of the system is composed of two parts: $\sum_\ell \mathcal{H}^\ell$, the sum of the planar Hamiltonians, and $\mathcal{H}_\perp$, the antiferromagnetic coupling between the planes, i.e.

$$\mathcal{H} = \sum_\ell \mathcal{H}^\ell + \mathcal{H}_\perp \tag{1}$$

$$\mathcal{H}^\ell = \mathcal{H}^\ell_{\rm ex} + \mathcal{H}^\ell_{\rm DM} + \mathcal{H}^\ell_{\rm Z} \tag{2}$$

$$\mathcal{H}_\perp = \sum_\ell \sum_i J_\perp \boldsymbol{S}^{\rm A}_{i,\ell} \cdot \boldsymbol{S}^{\rm B}_{i,\ell+1} + \sum_\ell \sum_j J_\perp \boldsymbol{S}^{\rm B}_{j,\ell} \cdot \boldsymbol{S}^{\rm A}_{j,\ell+1}. \tag{3}$$

Here and in the following ℓ labels the planes and i, j labels the site, $\mathcal{H}^\ell_{\rm ex}$ is the Heisenberg exchange Hamiltonian of the ℓth plane, $\mathcal{H}^\ell_{\rm DM}$ denotes the DM interaction which causes a small canting out of the CuO_2 plane, and $\mathcal{H}^\ell_{\rm Z}$ is the Zeeman energy of spins in the magnetic field $\boldsymbol{H} = (H, 0, 0)$. They are as follows:

$$\mathcal{H}^\ell_{\rm ex} = \sum_{i,\delta} (J^{xx} S^{\rm A}_{i,\ell;x} S^{\rm B}_{i+\delta,\ell;x} + J^{yy} S^{\rm A}_{i,\ell;y} S^{\rm B}_{i+\delta,\ell;y} + J^{zz} S^{\rm A}_{i,\ell;z} S^{\rm B}_{i+\delta,\ell;z}) \tag{4}$$

$$\mathcal{H}^\ell_{\rm DM} = \sum_{i,\delta} \boldsymbol{D}^\ell_{ii+\delta} \cdot \boldsymbol{S}^{\rm A}_{i,\ell} \times \boldsymbol{S}^{\rm B}_{i+\delta,\ell} \tag{5}$$

$$\mathcal{H}^\ell_{\rm Z} = -\sum_i g\mu_{\rm B} H S^{\rm A}_{i,\ell;x} - \sum_j g\mu_{\rm B} H S^{\rm B}_{j,\ell;x}. \tag{6}$$

The δ summation runs over the four nearest neighbours of i. J^{xx}, J^{yy} and J^{zz} are slightly different from each other with J^{zz} being the largest. This anisotropy causes the staggered moment lying in the direction of the z axis. The DM vector $\boldsymbol{D}^{\ell}_{ii+\delta}$ (with $i \in$ A and $i+\delta \in$ B) is along the y axis. It should be noted that the direction of preferred canting alternates between adjacent CuO_2 planes when $\boldsymbol{H}$ is zero. This alternation requires [10] $\boldsymbol{D}^{\ell}_{ii+\delta} = -\boldsymbol{D}^{\ell+1}_{ii+\delta}$. Combined with the symmetries given by Coffey *et al* [9], $\boldsymbol{D}^{\ell}_{ii+\delta}$ reads

$$\boldsymbol{D}^{\ell}_{ii+\delta} = (-1)^{\ell+1} D\hat{\boldsymbol{y}}. \tag{7}$$

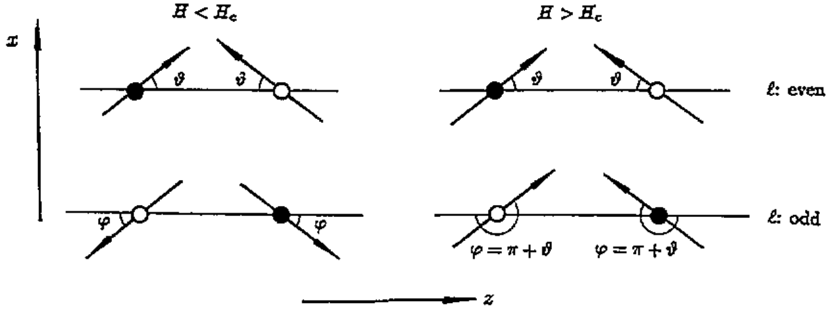

Figure 2. The spin structure under a magnetic field perpendicular to the CuO_2 plane. Full circle: sublattice A; open circle: sublattice B.

Table 1. Definition of $\theta_{\lambda,\ell}$.

λ	ℓ is odd	ℓ is even
A	$2\pi - \varphi$	ϑ
B	$\pi + \varphi$	$\pi - \vartheta$

Before applying the HP transformation to the Hamiltonians (1)–(6), we introduce a new local coordinate system $(\tilde{x}, y, \tilde{z})$ at each site to ensure that the axis $\tilde{z}$ coincides with the real canting direction of that site (see figure 2). The local coordinate system $(\tilde{x}, y, \tilde{z})$ can be obtained from the original coordinate system (x, y, z) by rotating the latter about the y axis through an angle $\theta_{\lambda,\ell}$. Here λ stands for 'A' or 'B', and the angle $\theta_{\lambda,\ell}$ is defined in table 1. $(\tilde{S}^{\lambda}_{i,\ell;x}, \tilde{S}^{\lambda}_{i,\ell;y}, \tilde{S}^{\lambda}_{i,\ell;z}) \equiv \tilde{\boldsymbol{S}}^{\lambda}_{i,\ell}$, and the three components of the spin operator at site (ℓ, i, λ) in the local coordinate system $(\tilde{x}, y, \tilde{z})$, can be obtained from $(S^{\lambda}_{i,\ell;x}, S^{\lambda}_{i,\ell;y}, S^{\lambda}_{i,\ell;z}) \equiv \boldsymbol{S}^{\lambda}_{i,\ell}$ through the transformation [11]:

$$\boldsymbol{S}^{\lambda}_{i,\ell} = \exp(\theta_{\lambda,\ell}\hat{\boldsymbol{y}}\times)\tilde{\boldsymbol{S}}^{\lambda}_{i,\ell} \tag{8}$$

where $\hat{\boldsymbol{y}}$ is the unit vector along the y axis. By using equation (8), we transform the Hamiltonian $\mathcal{H}$ into a form expressed in terms of $\tilde{S}^{\lambda}_{i,\ell;x}$, $\tilde{S}^{\lambda}_{i,\ell;y}$ and $\tilde{S}^{\lambda}_{i,\ell;z}$.

At this point we decompose the product of spin operators appearing in $\mathcal{H}_{\perp}$ in the mean field approximation. This is reasonable since the interplanar coupling $J_{\perp}$ is weak [12]. We stress that this approximation is not essential in our theory, but it does make our theory simple and direct without losing the physics of the problem. In this way, we obtain

$$\mathcal{H} = -N_{\perp}N_{\parallel}\sum_{\ell=1,2} J_{\perp}S_{\ell}S_{\ell+1}\cos(\theta_{A,\ell} - \theta_{B,\ell+1}) + N_{\perp}\sum_{\ell=1,2}\mathcal{H}^{\ell}_{\text{eff}}. \tag{9}$$

$2N_\perp$ is the number of planes and $N_\parallel$ denotes the number of sites in *one* CuO_2 plane. $\mathcal{H}_{\rm eff}^{\ell}$ is the effective Hamiltonian of the ℓth plane and is given by

$$\begin{aligned}
\mathcal{H}_{\rm eff}^{\ell} = \sum_{i,\delta} & (J^{yy}\tilde{S}^{\rm A}_{i,\ell;y}\tilde{S}^{\rm B}_{i+\delta,\ell;y} - J_\ell^{zz}\tilde{S}^{\rm A}_{i,\ell;z}\tilde{S}^{\rm B}_{i+\delta,\ell;z} - J_\ell^{xx}\tilde{S}^{\rm A}_{i,\ell;x}\tilde{S}^{\rm B}_{i+\delta,\ell;x}) \\
& - \sum_{i,\delta} J_\ell^{xz}(\tilde{S}^{\rm A}_{i,\ell;z}\tilde{S}^{\rm B}_{i+\delta,\ell;x} - \tilde{S}^{\rm A}_{i,\ell;x}\tilde{S}^{\rm B}_{i+\delta,\ell;z}) \\
& + 2J_\perp\sum_i \left[\cos(\theta_{{\rm A},\ell} - \theta_{{\rm B},\ell+1})\tilde{S}^{\rm A}_{i,\ell;z}S_{\ell+1} - \sin(\theta_{{\rm A},\ell} - \theta_{{\rm B},\ell+1})\tilde{S}^{\rm A}_{i,\ell;x}S_{\ell+1}\right] \\
& + 2J_\perp\sum_i \left[\cos(\theta_{{\rm B},\ell} - \theta_{{\rm A},\ell+1})\tilde{S}^{\rm B}_{i,\ell;z}S_{\ell+1} - \sin(\theta_{{\rm B},\ell} - \theta_{{\rm A},\ell+1})\tilde{S}^{\rm B}_{i,\ell;x}S_{\ell+1}\right] \\
& - g\mu_{\rm B}H\sum_i(\sin\theta_{{\rm A},\ell}\tilde{S}^{\rm A}_{i,\ell;z} + \cos\theta_{{\rm A},\ell}\tilde{S}^{\rm A}_{i,\ell;x}) \\
& - g\mu_{\rm B}H\sum_i(\sin\theta_{{\rm B},\ell}\tilde{S}^{\rm B}_{i,\ell;z} + \cos\theta_{{\rm B},\ell}\tilde{S}^{\rm B}_{i,\ell;x})
\end{aligned} \tag{10}$$

where

$$\begin{aligned}
J_\ell^{zz} &= J^{zz}\cos^2\theta_{{\rm A},\ell} - J^{xx}\sin^2\theta_{{\rm A},\ell} + (-1)^\ell D\sin 2\theta_{{\rm A}\lambda} \\
J_\ell^{xx} &= J^{xx}\cos^2\theta_{{\rm A},\ell} - J^{zz}\sin^2\theta_{{\rm A},\ell} + (-1)^\ell D\sin 2\theta_{{\rm A}\lambda} \\
J_\ell^{xz} &= \tfrac{1}{2}J^{xx}\sin 2\theta_{{\rm A},\ell} + \tfrac{1}{2}J^{zz}\sin 2\theta_{{\rm A},\ell} - (-1)^\ell D\cos 2\theta_{{\rm A}\lambda}.
\end{aligned} \tag{11}$$

In deriving the above equations, the relations $\langle\tilde{S}^{\lambda}_{i,\ell;z}\rangle = S_\ell$ and $\langle\tilde{S}^{\lambda}_{i,\ell;x}\rangle = \langle\tilde{S}^{\lambda}_{i,\ell;y}\rangle = 0$ are used.

2.2. *Spin-wave theory*

Now substitute the following Holstein–Primakoff transformation:

$$\begin{aligned}
\tilde{S}^{\rm A}_{i,\ell;z} &= S - a^\dagger_{\ell i}a_{\ell i} \\
\tilde{S}^{\rm A}_{i,\ell;x} &= \sqrt{\tfrac{1}{2}S}[f_{\ell i}(S)\,a_{\ell i} + a^\dagger_{\ell i}\,f_{\ell i}(S)] \\
\tilde{S}^{\rm A}_{i,\ell;y} &= -{\rm i}\sqrt{\tfrac{1}{2}S}[f_{\ell i}(S)\,a_{\ell i} - a^\dagger_{\ell i}\,f_{\ell i}(S)]
\end{aligned} \tag{12}$$

and

$$\begin{aligned}
\tilde{S}^{\rm B}_{i,\ell;z} &= S - b^\dagger_{\ell i}b_{\ell i} \\
\tilde{S}^{\rm B}_{i,\ell;x} &= \sqrt{\tfrac{1}{2}S}[g_{\ell i}(S)\,b_{\ell i} + b^\dagger_{\ell i}\,g_{\ell i}(S)] \\
\tilde{S}^{\rm B}_{i,\ell;y} &= -{\rm i}\sqrt{\tfrac{1}{2}S}[g_{\ell i}(S)\,b_{\ell i} - b^\dagger_{\ell i}\,g_{\ell i}(S)]
\end{aligned} \tag{13}$$

with $f_{\ell i}(S) = (1 - a^\dagger_{\ell i}a_{\ell i}/2S)^{1/2}$ and $g_{\ell i}(S) = (1 - b^\dagger_{\ell i}b_{\ell i}/2S)^{1/2}$, into the Hamiltonian $\mathcal{H}$, and perform the Fourier transformations of $a_{\ell i}$, $a^\dagger_{\ell i}$ and $b_{\ell i}$, $b^\dagger_{\ell i}$:

$$a_{\ell k} = \sqrt{2/N_\parallel}\sum_m {\rm e}^{{\rm i}km}a_{\ell m} \qquad a^\dagger_{\ell k} = \sqrt{2/N_\parallel}\sum_m {\rm e}^{-{\rm i}km}a^\dagger_{\ell m}$$

and similarly for $b_{\ell k}$ and $b^{\dagger}_{\ell k}$. Furthermore, we use the new operators $\alpha_{\ell k}$, $\alpha^{\dagger}_{\ell k}$ and $\beta_{\ell k}$, $\beta^{\dagger}_{\ell k}$ defined by

$$\begin{pmatrix} a_{\ell k} \\ a^{\dagger}_{\ell -k} \\ b_{\ell k} \\ b^{\dagger}_{\ell -k} \end{pmatrix} = \frac{1}{\sqrt{2}} \begin{pmatrix} \cosh\Psi^{(1)}_{\ell k} & \sinh\Psi^{(1)}_{\ell k} & \cosh\Psi^{(2)}_{\ell k} & \sinh\Psi^{(2)}_{\ell k} \\ \sinh\Psi^{(1)}_{\ell k} & \cosh\Psi^{(1)}_{\ell k} & \sinh\Psi^{(2)}_{\ell k} & \cosh\Psi^{(2)}_{\ell k} \\ \cosh\Psi^{(1)}_{\ell k} & \sinh\Psi^{(1)}_{\ell k} & -\cosh\Psi^{(2)}_{\ell k} & -\sinh\Psi^{(2)}_{\ell k} \\ \sinh\Psi^{(1)}_{\ell k} & \cosh\Psi^{(1)}_{\ell k} & -\sinh\Psi^{(2)}_{\ell k} & -\cosh\Psi^{(2)}_{\ell k} \end{pmatrix} \begin{pmatrix} \alpha_{\ell k} \\ \alpha^{\dagger}_{\ell -k} \\ \beta_{\ell k} \\ \beta^{\dagger}_{\ell -k} \end{pmatrix} \tag{14}$$

where

$$\begin{aligned} &\tanh 2\Psi^{(1)}_{\ell k} = X(k,\ell) = \frac{B_\ell \gamma_k}{A_\ell - D_\ell \gamma_k} \qquad \tanh 2\Psi^{(2)}_{\ell k} = -\frac{B_\ell \gamma_k}{A_\ell + D_\ell \gamma_k}. \\ &A_\ell = 4SJ^{zz}_\ell - 2J_\perp S_{\ell+1}\cos(\theta_{A,\ell} - \theta_{B,\ell+1}) + g\mu_B H \sin\theta_{A,\ell} \\ &B_\ell = 2SJ^{xx}_\ell + 2SJ^{yy} \qquad D_\ell = 2SJ^{xx}_\ell - 2SJ^{yy}. \end{aligned} \tag{15}$$

$\gamma_k = \frac{1}{2}(\cos k_x a + \cos k_y a)$ with a the lattice constant of the square lattice. Then the Hamiltonian can be separated into linear, quadratic and higher-order terms of the operators $\alpha_{\ell k}$, $\alpha^{\dagger}_{\ell k}$ and $\beta_{\ell k}$, $\beta^{\dagger}_{\ell k}$. The canting angles ϑ and φ are found from the vanishing of the coefficients of the terms linear in the boson operators in the expansion [13] and to order $S^{1/2}$ we have

$$4J^{xz}_\ell \bar{S}_\ell - g\mu_B H\cos\theta_{A\ell} - 2J_\perp S_{\ell+1}\sin(\theta_{A\ell} - \theta_{B\ell+1}) = 0. \tag{16}$$

Here

$$\bar{S}_\ell = S - [(3a_\ell - 2b_\ell) + (\tfrac{1}{2}d_\ell - 2e_\ell) + (k_\ell - 4l_\ell) + (3m_\ell - 2t_\ell)] \tag{17}$$

with

$$\begin{aligned} &a_\ell = \frac{1}{2N_\parallel}\sum_k \left(\frac{1}{\sqrt{1 - X(k,\ell)^2}} - 1\right) \qquad b_\ell = \frac{1}{2N_\parallel}\sum_k \gamma_k \left(\frac{1}{\sqrt{1 - X(k,\ell)^2}} - 1\right) \\ &d_\ell = \frac{1}{2N_\parallel}\sum_k \frac{X(k,\ell)}{\sqrt{1 - X(k,\ell)^2}} \qquad e_\ell = \frac{1}{2N_\parallel}\sum_k \frac{\gamma_k X(k,\ell)}{\sqrt{1 - X(k,\ell)^2}} \end{aligned} \tag{18}$$

and

$$\begin{aligned} &k_\ell = \frac{1}{2N_\parallel}\sum_k \frac{X(k,\ell)}{\sqrt{1 - X(k,\ell)^2}} n^{(\ell)}_k \qquad l_\ell = \frac{1}{2N_\parallel}\sum_k \frac{\gamma_k X(k,\ell)}{\sqrt{1 - X(k,\ell)^2}} n^{(\ell)}_k \\ &m_\ell = \frac{1}{2N_\parallel}\sum_k \frac{1}{\sqrt{1 - X(k,\ell)^2}} n^{(\ell)}_k \qquad t_\ell = \frac{1}{2N_\parallel}\sum_k \frac{\gamma_k}{\sqrt{1 - X(k,\ell)^2}} n^{(\ell)}_k. \end{aligned} \tag{19}$$

$n^{(\ell)}_k = [\exp(\beta\Omega(k,\ell)) - 1]^{-1}$ is the Bose function with $\Omega(k,\ell)$ the energy spectrum defined below. a_ℓ, b_ℓ, d_ℓ, e_ℓ and k_ℓ, l_ℓ, m_ℓ, t_ℓ, arising from the spin-wave interactions, contribute to the terms of order $S^{1/2}$ and if one only retains the lowest order, i.e. neglects the spin-wave interactions, they can be treated as zero and hence $\bar{S}_\ell = S$. Then equation (16) is same as that obtained from equations of motion which characterize the classical motion. One point should be noted: there exist four small quantities, D, $J_\perp$, $J^{zz} - J^{xx}$ and $J^{zz} - J^{yy}$, in our problem and all of them play important roles in determining the canting angles and the critical magnetic field H_c of the transition. If $D = J_\perp = J^{zz} - J^{xx} = J^{zz} - J^{yy} = 0$, then $b_\ell = d_\ell = k_\ell = t_\ell = D_\ell = 0$. Both our numerical computation and the analysis of order of magnitude show it will be sufficiently accurate if we retain terms only to the first order of b_ℓ, d_ℓ, k_ℓ, t_ℓ and D_ℓ. Hence, in the following, terms of these quantities of higher order have been dropped. By picking up all terms to order $1/S$, the Hamiltonian (10) now reads

$$\begin{aligned}\mathcal{H} = C + N_\perp \sum_{\ell=1,2}\bigg(&\sum_k[\omega(k,\ell) - (A_\ell - D_\ell\gamma_k) + \Omega(k,\ell)\,(n_{\ell k} + n'_{\ell K-k})]\\ &+ \frac{1}{2SN_\parallel}\sum_{k_1k_2}[W^{(1)}(k_1,k_2,\ell)\,n_{\ell k_1}n_{\ell k_2} + W^{(2)}(k_1,k_2,\ell)\,n'_{\ell k_1}n'_{\ell k_2}\\ &+ W^{(3)}(k_1,k_2,\ell)\,n_{\ell k_1}n'_{\ell k_2}] + \cdots\bigg)\end{aligned} \tag{20}$$

in which

$$n_{\ell k} = \alpha^\dagger_{\ell k}\alpha_{\ell k} \qquad n'_{\ell k} = \beta^\dagger_{\ell k}\beta_{\ell k}$$

$$C = 2N_\perp N_\parallel \sum_{\ell=1,2} C_\ell$$

$$\begin{aligned}C_\ell = -S^2J^{zz}_\ell - \tfrac{1}{2}g\mu_B HS\sin\theta_{A\ell} + J_\perp SS_{\ell+1}\cos(\theta_{A\ell} - \theta_{B\ell+1}) - J_\perp S_\ell S_{\ell+1}\cos(\theta_{A\ell} - \theta_{B\ell+1})\\ - \frac{1}{S}[G_\ell(a^2_\ell + e^2_\ell) - 2B_\ell a_\ell e_\ell] + \frac{1}{S}[G_\ell(m^2_\ell + 4l^2_\ell) - 4B_\ell l_\ell m_\ell].\end{aligned}$$

The energy spectrum is

$$\begin{aligned}\Omega(k,\ell) &\equiv \omega(k,\ell) + Q(k,\ell)\\ &= [(A_\ell + D_\ell\gamma_k)^2 - (B_\ell\gamma_k)^2]^{1/2} + Q(k,\ell)\end{aligned} \tag{21}$$

with

$$\begin{aligned}Q(k,\ell) = \frac{1}{S}\bigg(&\frac{2}{\sqrt{1 - X(k,\ell)^2}}[(B_\ell a_\ell - G_\ell e_\ell)\gamma_k X(k,\ell) + (B_\ell e_\ell - G_\ell a_\ell)]\\ &+ \frac{\gamma_k}{\sqrt{1 - X(k,\ell)^2}}[(2a_\ell + e_\ell)D_\ell + 2J^{xx}(d_\ell - b_\ell)]\bigg)\end{aligned}$$

which arises from the $1/S$ expansion. $G_\ell = 4SJ^{zz}_\ell$. To the order S^0 (namely with the spin-wave interactions neglected), $Q(k,\ell) = 0$, the energy spectrum (21) is written as $\Omega(k,\ell) = \omega(k,\ell)$. In the special case where $J_\perp = 0$, this spectrum is the same as that given in [3] and [5–7]. Specifically, if we further let $J^{xx} = J^{yy} = J^{zz} = J$ and $H = 0$ in our spectrum, the result $\omega(k) = 4SJ[\lambda_D(\lambda_D + \gamma_k)(1 - \gamma_k)]^{1/2}$, which is given by Coffey *et al* [9], can be obtained, with $\lambda_D = [1 + (D/J)^2]^{1/2}$. The expressions for $W^i(k_1,k_2,\ell)$ $(i = 1, 2, 3)$ are given in the appendix. $K = (\pi/a, \pi/a)$ and S_ℓ is given by

$$\begin{aligned}S_\ell &= S - \langle a^\dagger_{\ell i}a_{\ell i}\rangle = S - \langle b^\dagger_{\ell i}b_{\ell i}\rangle\\ &= S - \frac{1}{N_\parallel}\sum_k \frac{1}{\sqrt{1 - X(k,\ell)^2}}\coth\frac{\Omega(k,\ell)}{2k_BT} + \frac{1}{2}.\end{aligned} \tag{22}$$

The free energy per site now reads

$$F = \sum_{\ell=1,2} F_\ell$$

$$\begin{aligned}F_\ell = C_\ell + \frac{1}{2N_\parallel}\sum_k[\omega(k,\ell) - (A_\ell - D_\ell\gamma_k)] + \frac{k_BT}{N_\parallel}\sum_k \ln(1 - \mathrm{e}^{-\beta\Omega(k,\ell)})\\ + (1/2S)(8B_\ell m_\ell l_\ell + \tfrac{1}{2}G_\ell(m^2_\ell + 4l^2_\ell)).\end{aligned} \tag{23}$$

3. Numerical results and discussion

It is easy to verify that when $H = 0$, $\varphi = \vartheta = \frac{1}{2}\tan^{-1}(2D/(J^{zz} + J^{xx}))$ is a solution of equations (16). For a given temperature T, we solve equations (16), (22) and (23) numerically to study how the canting angles ϑ, φ and the corresponding free energy evolve with the magnetic field H from the $H = 0$ solution. Such canting states are stable only when H is sufficiently small. As H exceeds a critical value $H_c(T)$, it becomes unstable and then changes to the state with $\varphi = \pi + \vartheta$ (see figure 2). In this way, the critical magnetic field $H_c(T)$ is determined.

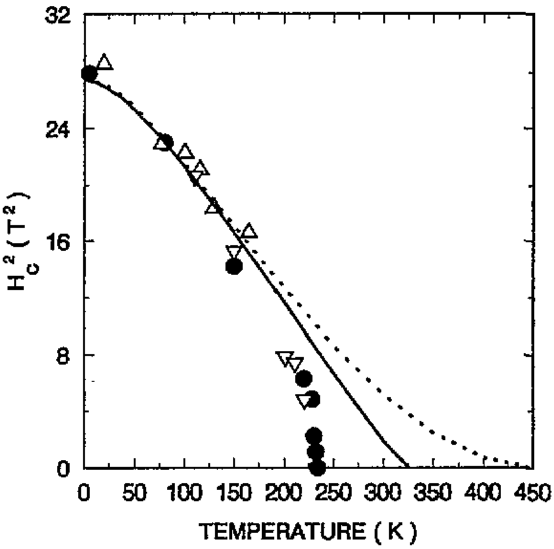

Figure 3. Square of the critical field H_c as a function of temperature. Full curve: theory with spin-wave interactions included; dotted curve: theory with spin-wave interactions neglected. Experimental points: ▽ and △ from [1]; ● from [2].

The variation of H_c^2 against temperature T is plotted in figure 3, where the full curve is that predicted by our theory with spin-wave interactions included and $S = \frac{1}{2}$. In our computation, we choose $J^{zz} \simeq 129$ meV, $J^{zz} - J^{yy} = 4.64 \times 10^{-4}$ meV, $J^{zz} - J^{xx} = 5.66 \times 10^{-3}$ meV and $D = 0.55$ meV, which coincide with the values given by Peters *et al* [3] within the experimental uncertainty, and $J_\perp = 2.122$ μeV which is between the value 2 μeV used by Thio *et al* [1] and the values 3 μeV used by Peters *et al* [3]. The lowest-order case, which is obtained by letting $a_\ell = b_\ell = d_\ell = e_\ell = k_\ell = l_\ell = m_\ell = t_\ell = 0$ in equations (16) and (20)–(23), is also plotted in the same figure as a dotted curve. The experimental data shown in figure 3 are taken from [1] and [2]. We see from the figure that our theory fits fairly well with the experiment only at low temperatures, ranging from 0 to 175 K. In our calculation we find that the transition from antiferromagnetic to weak ferromagnetic order is a first-order transition for all temperatures, however in the report of Kastner *et al* this is true only at low temperatures. This is not surprising as the spin-wave theory only holds at low temperatures. Finally, we can see from the figure that inclusion of spin-wave interactions (SWI) has little effect on the critical field $H_c(T)$ at low temperatures ranging from 0 to 160 K where both theories fit the experimental points fairly well. At high temperatures, however, SWI markedly reduce $H_c(T)$.

Acknowledgments

One of the authors (MWW) would like to thank Professor Lie-Zhao Cao and Ms Wei Sun for their invaluable support for his work.

Appendix. Expressions for $W^i(k_1, k_2, \ell)$

The expressions for $W^i(k_1, k_2, \ell)$ ($i = 1, 2, 3$) are given by

$$\begin{aligned}
W^{(1)}(k_1, k_2, \ell) = {} & [(1 - X(k_1, \ell)^2)(1 - X(k_2, \ell)^2)]^{-1/2} \\
& \times [D_\ell(\gamma_{k_1} + \gamma_{k_2})(1 + \tfrac{1}{2}X(k_1, \ell)X(k_2, \ell)) \\
& - G_\ell\left(1 + \gamma_{k_1}\gamma_{k_2} + \gamma_{k_1}\gamma_{k_2}X(k_1, \ell)X(k_2, \ell)\right) \\
& + B_\ell(\gamma_{k_1} + 2\gamma_{k_2})X(k_2, \ell)] \qquad \text{(A1)} \\
W^{(2)}(k_1, k_2, \ell) = {} & -[(1 - X'(k_1, \ell)^2)(1 - X'(k_2, \ell)^2)]^{-1/2} \\
& \times [D_\ell(\gamma_{k_1} + \gamma_{k_2})(1 + \tfrac{1}{2}X'(k_1, \ell)X'(k_2, \ell)) \\
& + G_\ell(1 + \gamma_{k_1}\gamma_{k_2} + \gamma_{k_1}\gamma_{k_2}X'(k_1, \ell)X'(k_2, \ell)) \\
& + B_\ell(\gamma_{k_1} + 2\gamma_{k_2})X'(k_2, \ell)] \qquad \text{(A2)} \\
W^{(3)}(k_1, k_2, \ell) = {} & [(1 - X(k_1, \ell)^2)(1 - X'(k_2, \ell)^2)]^{-1/2} \\
& \times [2D_\ell(\gamma_{k_1} - \gamma_{k_2})(1 + \tfrac{1}{2}X(k_1, \ell)X'(k_2, \ell)) \\
& - 2G_\ell(1 - \gamma_{k_1}\gamma_{k_2} - \gamma_{k_1}\gamma_{k_2}X(k_1, \ell)X'(k_2, \ell)) \\
& + B_\ell((\gamma_{k_1} - 2\gamma_{k_2})X'(k_2, \ell) + (2\gamma_{k_1} - \gamma_{k_2})X(k_1, \ell))] \qquad \text{(A3)}
\end{aligned}$$

in which $X'(k, \ell) = X(K - k, \ell)$.

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