Low-temperature properties of quantum Heisenberg helimagnets

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Low-temperature properties of the body-centered-tetragonal Heisenberg helimagnet are investigated by use of temperature-dependent spin waves. The calculation of the spin waves takes into account magnon-magnon interactions via a random-phase approximation. A quantum correction to the classical ground state, and the temperature dependence of the turn angle Q and of the basal-plane magnetization are shown.

I. INTRODUCTION

Magnetic properties and phase transitions in helimagnets have been extensively studied for nearly three decades. Since the pioneer theoretical works by Yoshimori[1] and Villain,[2] there has been a large number of studies dealing with the helimagnets.[3,4] In a previous paper,[5] the nature of the helical transition in a body-centered-tetragonal (bct) lattice with classical XY and Heisenberg spins has been investigated. It was found that for XY spins the transition associated with the loss of helical ordering is of first order and it is followed by a second-order transition associated with the breaking of basal-plane ordering. For Heisenberg spins, only a second-order transition was found.

In this paper, we are interested in the low-temperature behaviors of bct helimagnets with *quantum* Heisenberg spins. It is known that low-temperature properties of magnetic materials are governed by spin-wave (SW) excitations. In a first approximation, the free SW spectrum at temperature $T=0$ is often used to calculate physical quantities such as magnetization and free energy at finite T. Recently, Rastelli, Reatto, and Tassi[6] studied the effect of magnon-magnon interaction at $T=0$ via a perturbation calculation. For finite T, twenty years ago Nagai[7] proposed a method to take into account these interactions via a random-phase-approximation (RPA) decoupling scheme for antiferromagnets. This method has been used by Harada and Motizuki[8] to calculate the SW at finite T in a general helical structure. We shall apply this method to a bct Heisenberg helimagnet.

In Sec. II the method is briefly reviewed. Results for the bct helimagnet are shown and discussed in Sec. III. Concluding remarks are given in Sec. IV.

II. METHOD

The Hamiltonian is written as

$$H=-\sum_{i,j} J(R_{ij})\mathbf{S}_i\cdot\mathbf{S}_j\,, \tag{1}$$

where S_i is a quantum Heisenberg spin of the magnitude S at the lattice site R_i and $J(R_{ij})$ is the exchange integral between pairs of spins at distance $R_{ij}=R_i-R_j$.

In this paper, we take $J(R_{ij})=J_1$ (>0 or <0) between nearest neighbors (NN's) in the $\langle 111\rangle$ directions of a bct lattice and $J(R_{ij})=J_2$ (<0) between NN along the c axis (Fig. 1). The classical ground-state energy per spin is given by

$$U_0=-4J_1\cos(Q_0)-J_2\cos(2Q_0)\,, \tag{2}$$

where Q_0 is the angle between spins belonging to two adjacent basal planes (spins in each plane are parallel) given by

$$\cos(Q_0)=-J_1/J_2\,. \tag{3}$$

In the following, we choose a particular spin configuration in which spin S_i is quantized along the ζ_i axis in the basal plane. The ξ_i axis is taken in the basal plane and is perpendicular to the ζ_i axis and the η_i axis is taken in such a way as to form a direct trihedron (ζ_i,ξ_i,η_i) (see Fig. 1). For numerical convenience, we introduce a very small stabilizing field which acts along each local quantization axis. The Zeeman energy is given by

$$-h\sum_i S_{i\zeta}\,. \tag{4}$$

We use this stabilizing field instead of an easy-plane anisotropy to avoid the Goldstone mode at wave vector $k=0$ for numerical iteration input. The classical ground state is not affected by h. The effects of h and an easy-plane anisotropy on the SW will be considered.

The calculations are done with the following steps:

(i) H is written in the local coordinates and the Holstein-Primakoff transformation is used with operators a and $a^\dagger$ to expand H up to four-operator terms: $H=H_0+H_1+H_2+H_3+H_4+\cdots$, where H_0 contains the nonoperator terms and H_i ($i=1$-4) are i-operator terms. H_1 vanishes by inversion symmetry.[8]

(ii) Fourier transforms and the RPA decoupling scheme are used.[7] H_3 vanishes in this scheme while H_4 becomes two operator terms which modify H_2.

(iii) The resulting Hamiltonian is diagonalized by the Bogoliubov transformation with new operators α and $\alpha^\dagger$.

For details of the calculations, the reader is referred to Ref. 8. The result is

$$H=H_0'+H_2'\,,$$

where H_0' (the nonoperator terms) and H_2' are given by

$$H_0' = -J(Q)NS(S+1) - (S/2)\sum_k \{A_1(k,Q,T)+A_2(k,Q,T)+\langle a_k^\dagger a_k\rangle[A_1(k,Q,T)+A_2(k,Q,T)]$$
$$+\langle a_k^\dagger a_{-k}^\dagger\rangle[B_1(k,Q,T)+B_2(k,Q,T)]\}, \quad (5)$$
$$H_2' = (S/2)\sum_k \hbar\omega_k(Q,T)(a_k^\dagger a_k + a_k a_k^\dagger) = (S/2)\sum_k \hbar\omega_k(Q,T) + S\sum_k \hbar\omega_k(Q,T)a_k^\dagger a_k, \quad (6)$$

where the last equality follows from the commutation relation between operators and N is the number of spins. In Eq. (5), a constant has been omitted and Q is the value of the turn angle at T. Other notations are

$$\hbar\omega_k(Q,T) = [A^2(k,Q,T) - B^2(k,Q,T)]^{1/2}, \quad (7)$$
$$A(k,Q,T) = A_0(k,Q) + A_1(k,Q,T) + A_2(k,Q,T), \quad (8a)$$
$$B(k,Q,T) = B_0(k,Q) + B_1(k,Q,T) + B_2(k,Q,T), \quad (8b)$$
$$A_0(k,Q) = 2J(Q) - J(k) - \tfrac{1}{2}[J(k+Q)+J(k-Q)] + h/S, \quad (9a)$$
$$B_0(k,Q) = J(k) - \tfrac{1}{2}[J(k+Q)+J(k-Q)], \quad (9b)$$
$$A_1(k,Q,T) = -(1/NS)\sum_q [A_0(k,Q)+A_0(q,Q)-2J(Q)+J(k-q+Q)+J(k-q-Q)]\langle a_q^\dagger a_q\rangle, \quad (9c)$$
$$A_2(k,Q,T) = -(1/NS)\sum_q [0.5B_0(k,Q)+B_0(q,Q)]\langle a_q^\dagger a_{-q}^\dagger\rangle, \quad (9d)$$
$$B_1(k,Q,T) = -(1/NS)\sum_q [0.5B_0(q,Q)+B_0(k,Q)]\langle a_q^\dagger a_q\rangle, \quad (9e)$$
$$B_2(k,Q,T) = -(1/NS)\sum_q \{0.5[A_0(k,Q)+A_0(q,Q)] - 2J(Q)+J(k-q+Q)+J(k-q-Q)\}\langle a_q^\dagger a_{-q}^\dagger\rangle, \quad (9f)$$
$$\langle a_q^\dagger a_q\rangle = (\langle n_q\rangle + \tfrac{1}{2})A(q,Q,T)/\hbar\omega_q(Q,T) - \tfrac{1}{2}, \quad (9g)$$
$$\langle a_q^\dagger a_{-q}^\dagger\rangle = -(\langle n_q\rangle + \tfrac{1}{2})B(q,Q,T)/\hbar\omega_q(Q,T), \quad (9h)$$
$$\langle n_q\rangle = \{\exp[S\hbar\omega_q(Q,T)/k_BT] - 1\}^{-1} \quad (9i)$$

[the minus sign of (9h) was missing in Ref. 8]. The Fourier transform $J(k)$ for the bct model studied here is

$$J(k) = 8J_1\cos(k_x)\cos(k_y)\cos(k_z) + 2J_2\cos(2k_z) = 4J_2[-4\cos Q_0\cos(k_x)\cos(k_y)\cos(k_z) + 2J_2\cos(2k_z)], \quad (10)$$

where Eq. (3) has been used. For convenience, the lattice constant has been taken equal to 2 and the following notations have been used in the above equations: $k = (k_x, k_y, k_z)$, $Q_0 = (0,0,Q_{0z})$, and $Q = (0,0,Q_z)$ (only the z component of Q is taken in our approximation which is reasonable because of the in-plane stabilizing field). At this stage, it is noted that without magnon-magnon interaction the SW spectrum is given by

$$\hbar\omega_k^0(Q) = [A_0^2(k,Q) - B_0^2(k,Q)]^{1/2}. \quad (11)$$

At a given T, one has to solve in a self-consistent way Eqs. (7)–(9), even at $T=0$ where $\langle n_q\rangle$ in Eq. (9) is zero, to obtain the SW spectrum for each value of Q (Q is allowed to vary around Q_0). Once this is done, the free energy is calculated from the following formula:

$$F = H_0' + (S/2)\sum_k \hbar\omega_k(Q,T)$$
$$+ k_BT\sum_k \ln\{1 - \exp[-S\hbar\omega_k(Q,T)/k_BT]\}, \quad (12)$$

where k_B is the Boltzmann constant. The value of Q corresponding to the minimum of F at a given T, which will be denoted as Q_c below, indicates the most stable state. The local magnetization is then calculated from

$$m = S - (1/N)\sum_q \langle a_q^\dagger a_q\rangle. \quad (13)$$

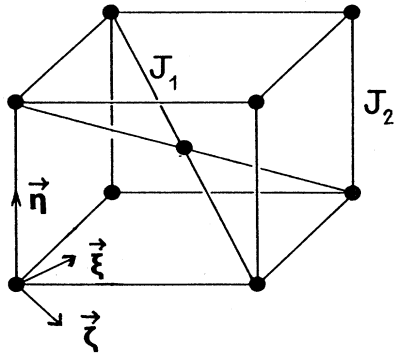

FIG. 1. bct lattice with interactions J_1 and J_2 along diagonal and vertical (c-axis) directions, respectively. Local frame (ζ,ξ,η) is indicated.

III. RESULTS AND DISCUSSION

We show here two examples where $J_1 > 0$ and $J_1 < 0$ ($J_2 < 0$) with $S = 2$. For the former, we choose $Q_0 = 60°$

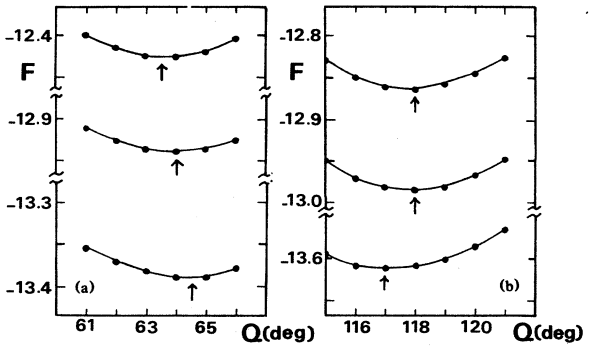

FIG. 2. (a) Free energy F vs Q (in degrees) for $Q_0=60°$ at $T=0$, 4, and 5 (from above) with $h=0.04$. (b) F vs Q for $Q_0=120°$ at $T=0$, 3, and 5 (from above) with $h=0.02$. Arrows indicate minimum F.

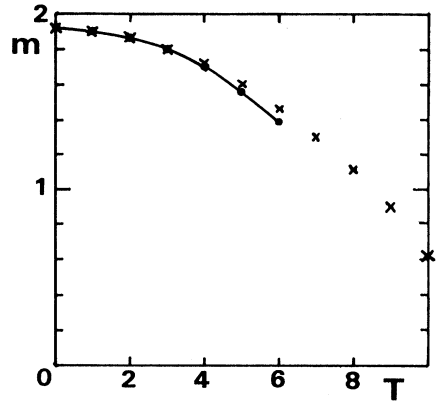

FIG. 3. Local magnetization m (at minimum F) vs T for $Q_0=60°$ and $h=0.04$ obtained from self-consistent SW (solid circles). No more self-consistent SW found for $T>6$. Results from a first approximation (see text) are shown by crosses for comparison.

$(-J_1/J_2=0.5)$ and for the latter, $Q_0=120°$ $(-J_1/J_2=-0.5)$. Quantities such as h, F, $\hbar\omega_k$, and T will be measured in units of $|J_2|$ and $k_B=1$. Note that self-consistent SW's are obtained up to a certain temperature above which the iterations do not converge. The reader is referred to Ref. 7 for detailed discussion about this point. Figure 2 shows F vs Q for various T obtained with self-consistent SW. The number of points in the first Brillouin zone was taken to be 40^3 and five iterations are sufficient at high T with an accuracy of about 10^{-4}. Some remarks about Fig. 2 are in order. (i) For $Q_0=60°$, Q_c is 63° 30′ at $T=0$ and slowly increases to 64° 30′ at $T=5$. No self-consistent solution is found for $T>6$. (ii) For $Q_0=120°$, Q_c is 118° at $T=0$ and remains so with increasing T and slowly decreases only just before no more self-consistent solution is found for $T>6$.

The local magnetization m for $Q_0=60°$ is displayed in Fig. 3 as a function of T, where results obtained by replacing A and B, in Eqs. (9g) and (9h), with A_0 and B_0 are also shown for comparison. This approximation gives results at high T contrary to the self-consistent calculation. The zero-point spin contractions are 3.5 and 6% for $Q_0=60°$ and 120°, respectively.

In order to see the effects of different terms in Eq. (12), we show in Table I the case of $Q_0=60°$ at $T=1$ and 5. All the terms are insensitive to Q, except the SW energy E_{SW} [second term of Eq. (12)] which decreases with increasing Q, making the minimum of F to move to higher Q. At higher T the entropy term becomes important and its decrease with increasing Q makes Q_c larger. The same is found for $Q_0=120°$ except E_{SW} increases with increasing Q, making Q_c lower.

Let us show now the temperature dependence of the SW spectrum for $Q_0=60°$ and $h=0.02$ in Fig. 4, where the free SW spectrum is also presented for comparison. $\hbar\omega_k^0(Q)$ is zero at $k=0$ and $k=Q$ for $h=0$. However, $\hbar\omega_k(Q,T)$ is positive definite at and around Q_0, as can be seen by expanding (7) in the vicinity of Q_0 at low temperatures. This allows the SW to be excited at and around

TABLE I. Contributions of various terms in F for $Q_0=60°$ and $h=0.04$, E_1 is the first term of H_0' [see Eq. (5)], E_2 are the first two terms in the curly brackets, and E_3 are the remaining terms of H_0', E_{SW} and E_e are the second and third terms of F [Eq. (12)], respectively. Upper and lower parts are results for $T=1$ and 5, respectively. See text for comments.

Q	E_1	E_2	E_3	E_{SW}	E_e	F
61	−17.9940	−0.1145	0.0032	5.7051	−0.0058	−12.4064
62	−17.9776	−0.1197	0.0046	5.6763	−0.0058	−12.4221
63	−17.9492	−0.1250	0.0060	5.6433	−0.0058	−12.4306
64	−17.9088	−0.1304	0.0074	5.6058	−0.0058	−12.4318
65	−17.8563	−0.1360	0.0088	5.5640	−0.0058	−12.4254
66	−17.7912	−0.1418	0.0101	5.5175	−0.0058	−12.4112
61	· · ·	0.3229	−0.0404	5.3463	−0.9893	−13.3550
62	· · ·	0.3180	−0.0402	5.3188	−0.9904	−13.3714
63	· · ·	0.3135	−0.0402	5.2866	−0.9933	−13.3825
64	· · ·	0.3094	−0.0403	5.2497	−0.9981	−13.3882
65	· · ·	0.3056	−0.0406	5.2080	−1.0050	−13.3882
66	· · ·	0.3022	−0.0409	5.1612	−1.0140	−13.3827

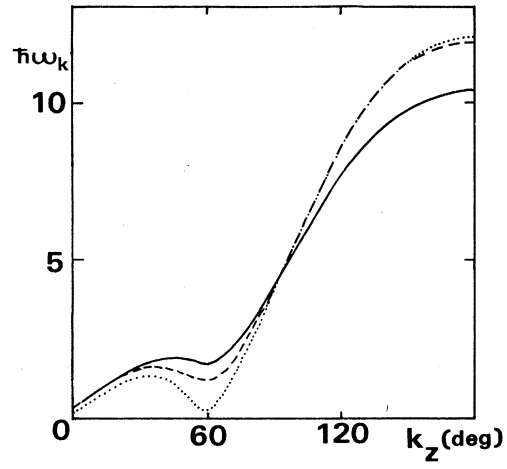

FIG. 4. SW spectrum for $k_x=k_y=0$ at $Q=60°$ in the case $Q_0=60°$ with $h=0.02$. Note that k_z is shown here in degrees (to recover the original k_z in radians, a conversion factor is needed). Dashed and solid lines are self-consistent SW at $T=0$ and $T=6$, respectively. Dotted line shows free SW using Eq. (11).

Q_0, driving the minimum of F elsewhere, as seen above. Recently, Rastelli *et al.*[6] have found that at $T=0$ the turn angle of the classical ground state does not change with quantum corrections, which include corrections due to first-order perturbation of four-operator terms and second-order perturbation of three-operator terms. It was shown numerically that the former and the latter cancel out leaving the turn angle of the classical ground state unchanged in a simple-cubic helical model. However, it is not clear to us whether this results form the fact that the bilinear (two-operator) terms are first diagonalized and higher-order terms are considered as perturbations. In order to estimate the effects of H_3 we calculated the second-order perturbation of H_3 as follows (the expression of H_3 has been given in Ref. 8): transforming the operators a and $a^\dagger$ into α and $\alpha^\dagger$ and using the eigenstates of α and $\alpha^\dagger$, one obtains the second-order contribution of H_3 which is too long to write down here. Numerical calculations done at $T=0$ show that this is of the order of 10^{-10} too small to affect $\hbar\omega_k(Q,T)$. Even at $T=1$ where more SW are excited, it is of the order to 10^{-3}, while the contribution of H_4 is of the order of 10^{-1}.

The effect of h at $T=0$ is not significant. For $h=0.002$, 0.01, 0.02, and 0.04 the zero-point spin contractions are all 3.5%. However, Q_c does not vary significantly with increasing T for very small h. Besides, one notices that at $T=0$, the correction Q_c-Q_0 is positive or negative. We conjecture that it depends on the sign of J_1.

Finally, let us mention that the results shown above are without anisotropy and the inclusion in the self-consistent calculation of an easy-plane anisotropy of the type DS_{in}^2, where $D>0$ (see additional terms given in Ref. 8) yields no change in the value of Q_c, at least for values we have taken $D=0.01$ and 0.02 (in units of $|J_2|$).

IV. CONCLUDING REMARKS

We have shown that quantum correction to the classical ground state is important within our approximation in disagreement with Rastelli *et al.*[6] This problem, therefore, remains open for future investigations. Within our approximation, the turn angle is found to be rather insensitive to T, in agreement with experiments[9] on VF_2 which has the same lattice structure studied here. Experiments on rare-earth metals Tb, Dy, and Ho (Ref. 10) show, on the other hand, that Q decreases with decreasing T. This is probably due to the fact that these elements have a ferromagnetic ordering at temperatures below the helical phase; Q decreases due to the enhancement of ferromagnetic interaction at low T. Therefore, the variation of Q depends on the interactions in the system.

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