

# Temperature dependence of the frustrated Heisenberg model in the fcc type-I configuration

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The Heisenberg model for the fcc type-I configuration is studied with the use of the interacting spin-wave theory at the order  $1/S$ ,  $S$  being the spins coupled antiferromagnetically. The continuous degeneracy of the ground state of the nearest-neighbor Heisenberg Hamiltonian ( $H_{nn}$ ) and the conditions for different types of interactions susceptible to stabilize a multi- $\mathbf{k}$  structure remain identical in the whole antiferromagnetic range in temperature. This set includes local anisotropy and four-spin and biquadratic exchange interactions as well as effects of the crystal electric field and quadrupolar interaction. The temperature dependence of the low-frequency magnetic excitations corresponding to  $H_{nn}$  is calculated analytically. For the double- $\mathbf{k}$  and triple- $\mathbf{k}$  configurations the quartic terms renormalize in great part the overall Heisenberg antiferro-coupling, giving schematically the behavior  $(\sqrt{1-\bar{n}/S})\sqrt{a+b(1-\bar{n}/S)}$  where  $\bar{n}$  has the temperature dependence of the Bose-Einstein distribution. Concerning the terms to be added to  $H_{nn}$  to lift the degeneracy of the ground state, they can be separated into two classes following their leading temperature dependences which are  $1-t\bar{n}/S$ , where  $t=1$  or 3. Available experiments are discussed.

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## I. INTRODUCTION

Type-I fcc antiferromagnets are typical examples of three-dimensional frustrated spin systems. The most general spin configuration for this kind of magnetic order can be written as a superposition of the three wave vectors  $K_1=(2\pi/a)\times(1,0,0)$ ,  $K_2=(2\pi/a)(0,1,0)$ , and  $K_3=(2\pi/a)(0,0,1)$ , where  $a$  is the lattice constant. This structure can then be in a single- $\mathbf{k}$ , double- $\mathbf{k}$ , or triple- $\mathbf{k}$  state, the triple- $\mathbf{k}$  structure being an extension of the  $120^\circ$  spin structure on the triangular lattice to the three-dimensional case.

Since the first experimental evidence of fcc type-I frustrated compounds, a great deal of both theoretical and experimental interests have arisen. In particular, there are compounds where the phonon effects are well known, thus allowing, in principle, a precise identification of the excitation spectrum on a large domain in temperature.<sup>1-3</sup> In this respect, NpBi and USb compounds present strong similarities (see, for instance, Ref. 4). For both, magnetic excitation spectra have been measured on a temperature range lower than the Néel temperatures of 192.5 and 213 K, respectively. They give strong indications that a noncollinear triple- $\mathbf{k}$  spin structure is realized on the whole antiferromagnetic domain. In both alloys the magnetic excitation spectra consist of two branches with longitudinal and transverse polarizations, respectively. On the other hand, the semiconductor MnTe<sub>2</sub>, which crystallizes with the pyrite-type structure, undergoes to a second-order antiferromagnetic (AF) transition at  $T_N=86.5$  K. The nature of its magnetic structure has been subject to controversy until recently. Indeed the experiment of Burllet *et al.*<sup>5</sup> assesses definitely the conclusion that the corresponding spin configuration is of the triple- $\mathbf{k}$  type-I structure. In contrast to previous claims,<sup>6</sup> this structure is stable on the whole AF domain.

Until now, most theoretical works have concentrated on the properties of the ground state at  $T=0$  and essentially concern the terms to be added to the isotropic Heisenberg

model to lift the degeneracy of this ground state. Effects due to biquadratic and four-spin exchange interactions in fcc spin lattices were first discussed, to our knowledge, by Yoshimori and Inagaki<sup>7</sup> and Yosida and Inagaki<sup>8</sup> in a classical treatment of the Heisenberg Hamiltonian (see also Ref. 9). In Ref. 10 a local anisotropy energy was considered in a phenomenological model, in which higher-order spin interactions were taken into consideration. The magnetic excitation spectra for type-I antiferromagnets have been modeled by Hälgl and Furrer<sup>1</sup> within the random phase approximation by taking into account the crystal-field, anisotropic bilinear exchange and isotropic quadrupolar interactions. Jensen and Bak<sup>11</sup> calculated the spin-wave spectra of USb with triple- $\mathbf{k}$  magnetic structure by using a Hamiltonian including, in addition to the AF exchange coupling between nearest neighbors  $H_{nn}$ , the pseudodipolar interaction and a crystal-field term with cubic symmetry. However, almost nothing is known about finite-temperature properties including the stability of various spin orderings. The purpose of this paper is to investigate the stability of the Néel order and the temperature dependence of the spin-wave spectra. The three possible AF structures are in general considered. A triple- $\mathbf{k}$  structure is, sometimes, only studied, but it is just an exercise to extend the formalism to others multi- $\mathbf{k}$  states. In Ref. 12 we have proposed a theoretical framework to study with reasonable efforts frustrated magnets in the fcc type-I configuration. Here we apply this framework to evaluate the  $1/S$  quantum corrections to the spin-wave spectra through the Holstein-Primakoff representation of spins. We prove that this theory gives relevant information within the whole range of the AF phase. For instance, the softening temperature  $T_s$ , where the spin-wave frequencies drop to zero is the Néel temperature, i.e., the temperature for which the magnetization disappears, at least for the triple- $\mathbf{k}$  configuration. We give strong reasons to think that, if some structure is stabilized at  $T=0$ , it remains the same on the whole AF domain of temperature. Moreover, we obtain analytical results on the temperature behavior of the spin-wave spectra.

This paper is organized as follows. In Sec. II formal theory is settled down. Invariances of the magnon spectra under some substitutions are used to shrink the unit magnetic cell and to establish some results, drastically simplifying the perturbative treatment of the Hamiltonian. These simplifications allow us to make new predictions about the temperature dependence of the spin-wave spectra of  $H_{nn}$  in Sec. III and of a set of supplementary interactions in Sec. IV. This set includes four-spin and biquadratic exchange interactions, local anisotropy and quadrupolar interaction. The crystal electric field is also studied. The experimental relevance of these results is discussed in the Conclusion.

## II. GENERAL FORMULATION

### A. Heisenberg model

The Hamiltonian we start from reads

$$H_{nn} = J \sum_{\langle i,j \rangle} (\mathbf{S}_{1i} \cdot \mathbf{S}_{2j} + \mathbf{S}_{1i} \cdot \mathbf{S}_{3j} + \mathbf{S}_{1i} \cdot \mathbf{S}_{4j} + \mathbf{S}_{2i} \cdot \mathbf{S}_{3j} + \mathbf{S}_{2i} \cdot \mathbf{S}_{4j} + \mathbf{S}_{3i} \cdot \mathbf{S}_{4j}), \quad (1)$$

with  $J > 0$ .  $\mathbf{S}_{Ii}$  represents the magnetic moment, located at the  $i$  th site, and corresponding to the sublattice  $I$ . The summation  $\langle i,j \rangle$  is taken over all nearest-neighbor pairs. The crystal structure of fcc is such that each site has 12 nearest-neighbor sites which belong four by four to the other three sublattices. Thus the coordination number  $z$  is 4. The magnetic moments  $\mathbf{S}_I$  are located at  $\delta_i$  in the unit magnetic cell as follows:  $\delta_1 = (0,0,0)$ ,  $\delta_2 = (0,1/2,1/2)$ ,  $\delta_3 = (1/2,0,1/2)$ , and  $\delta_4 = (1/2,1/2,0)$ . In order to consider simultaneously the single- $\mathbf{k}$ , double- $\mathbf{k}$ , and triple- $\mathbf{k}$  states we choose in full generality the direction of the magnetizations on sublattices located on the unit cell at  $\delta_i$  as  $(\alpha, \beta, \gamma)$ ,  $(-\alpha, \beta, -\gamma)$ ,  $(-\alpha, -\beta, \gamma)$ , and  $(\alpha, -\beta, -\gamma)$ , respectively. Here  $\alpha = \sin\vartheta \cos\varphi$ ,  $\beta = \sin\vartheta \sin\varphi$ , and  $\gamma = \cos\vartheta$  are polar directional cosines satisfying  $\alpha^2 + \beta^2 + \gamma^2 = 1$ . The triple- $\mathbf{k}$  state is given by  $\alpha = \beta = \gamma = 1/\sqrt{3}$  and corresponds to the AF state observed in the USb and MnTe<sub>2</sub> compounds.<sup>1,5</sup> Fixing, for instance,  $\theta \equiv \pi/2$ , which means a coplanar structure, the double- $\mathbf{k}$  model is recovered for  $\varphi = \pi/4$  (shown in Fig. 1 of Ref. 12). Finally, the single- $\mathbf{k}$  model is obtained from the condition  $\theta = 0$ . The ground state given by  $H_{nn}$  has a two-dimensional infinite degeneracy, since its energy is independent of the polar angles  $(\vartheta, \varphi)$ , betraying the high degree of frustration of this model. This high degeneracy can be lifted by additional terms in the Hamiltonian.

Various types of interactions have been studied in Ref. 12. In particular local anisotropy, four-spin exchange interactions, and biquadratic interactions have been considered. Their role in stabilizing a noncollinear magnetic state has been clarified. In spite of the great number of studies, nothing is well established since no clear signal emerges from the data. This is the reason why we made a systematic study of these interactions, extending our analysis<sup>12</sup> to quadrupolar interactions and crystal-electric-field effects.

In full generality the Hamiltonian (1) is expressed in terms of four types of bosonic operators  $\lambda = a, b, c$ , and  $d$  for

sublattices 1, 2, 3, and 4, respectively, these operators satisfying the commutation relations of elementary boson operators. Following the standard analysis within the linear spin-wave theory based on the Holstein-Primakoff representation,<sup>13</sup> we take a local coordinate system  $\xi\eta\zeta$  at each site of the lattice. The  $\zeta$  axis is taken to be the classical direction of spins in the ground state. The resulting transformation at each sublattice is expressed in terms of the polar coordinates  $(\theta, \varphi)$  which define the antiferromagnetic configuration of the four sublattices. Finally, the Hamiltonian (1) is given in terms of the Fourier transforms of the bosonic operators by ( $N$  being the total number of sites)

$$\frac{N\mathcal{H}_2}{JS} = -2zS + \sum_k \{ P_k \Omega_k + [ Q_k \Lambda_k + R_k^x \Sigma_k^x + R_k^y \Sigma_k^y + R_k^z \Sigma_k^z + V_k^x \Delta_k^x + V_k^y \Delta_k^y + V_k^z \Delta_k^z + \text{H.c.} ] \}, \quad (2)$$

where  $\Omega_k = n_k^a + n_k^b + n_k^c + n_k^d$ ,  $\Lambda_k = a_k a_{-k} + b_k b_{-k} + c_k c_{-k} + d_k d_{-k}$ ,  $\Sigma_k^x = a_k b_{-k} + c_k d_{-k}$ ,  $\Sigma_k^y = a_k c_{-k} + b_k d_{-k}$ ,  $\Sigma_k^z = a_k d_{-k} + b_k c_{-k}$ ,  $\Delta_k^x = a_k^\dagger b_k + c_k^\dagger d_k$ ,  $\Delta_k^y = a_k^\dagger c_k + b_k^\dagger d_k$ , and  $\Delta_k^z = a_k^\dagger d_k + b_k^\dagger c_k$ . For the sake of simplicity we assume  $\varphi = \pi/4$ . Then the coefficients are  $P_k = 4$ ,  $Q_k = 0$ ,  $R_k^x = 2C_{k_y} C_{k_z} (\sin^2\theta + 2i \cos\theta)$ ,  $R_k^y = 4C_{k_x} C_{k_z} \sin^2\theta$ ,  $R_k^z = 2C_{k_x} C_{k_y} (\sin^2\theta - 2i \cos\theta)$ ,  $V_k^x = 2C_{k_x} C_{k_z} \sin^2\theta$ ,  $V_k^y = -4C_{k_x} C_{k_z} \cos^2\theta$ , and  $V_k^z = 2C_{k_x} C_{k_y} \sin^2\theta$ . The structure factors are defined by  $C_{k_i} = \cos[a(k_i/2)]$  ( $i = x, y, z$ ) and  $n_k^a = a_k^\dagger a_k$ . From now on, we set the lattice constant  $a = 1$ . Up to now, the approximation used to obtain analytical results assumes that the different bosonic operators are independent of the sublattice to which they are bounded. This approach has been considered for the first time in Ref. 14 and has been proved to give the exact spin-wave dispersions in Ref. 12. The exact diagonalization of Hamiltonian (2) may be carried out in two different ways. The first approach, used in Ref. 12 consists of obtaining analytically through the MAPLE mathematical manipulation language<sup>15</sup> the four spin-wave dispersions. The complete solution requires knowledge of the Bogoliubov transformation to which we turn next.

### B. Bogoliubov transformation

We introduce a set of spin-wave operators  $\alpha_k, \alpha_k^\dagger, \beta_k, \beta_k^\dagger, \gamma_k, \gamma_k^\dagger, \delta_k, \delta_k^\dagger$  through the Bogoliubov transformations ( $k > 0$ ) which are a straightforward generalization of those given by Oguchi, Nishimori, and Taguchi in Ref. 14:

$$a_k = \frac{1}{2\sqrt{2}} [A_k \exp(-i\phi_k^A) + B_k \exp(-i\phi_k^B) + C_k \exp(-i\phi_k^C) + D_k \exp(-i\phi_k^D)],$$

$$b_k = \frac{1}{2\sqrt{2}} [A_k \exp(-i\phi_k^A) - B_k \exp(-i\phi_k^B) + C_k \exp(-i\phi_k^C) - D_k \exp(-i\phi_k^D)],$$

$$\begin{aligned}
c_k &= \frac{1}{2\sqrt{2}} [A_k \exp(-i\phi_k^A) + B_k \exp(-i\phi_k^B) \\
&\quad - C_k \exp(-i\phi_k^C) - D_k \exp(-i\phi_k^D)], \\
d_k &= \frac{1}{2\sqrt{2}} [A_k \exp(-i\phi_k^A) - B_k \exp(-i\phi_k^B) \\
&\quad - C_k \exp(-i\phi_k^C) + D_k \exp(-i\phi_k^D)], \quad (3)
\end{aligned}$$

where  $X_k = C_k^X(x_k + x_{-k}) + S_k^X(x_k^\dagger - x_{-k}^\dagger)$ , for  $X=A, B, C, D$  and  $x = \alpha, \beta, \gamma, \delta$ , respectively. For  $k < 0$  we have  $X_{-k} = C_k^X(x_k - x_{-k}) + S_k^X(x_k^\dagger + x_{-k}^\dagger)$ . The operators  $\alpha_k, \beta_k, \gamma_k$ , and  $\delta_k$  commute between themselves and satisfy the canonical commutation relations

$$[\alpha_k, \alpha_q^\dagger] = [\beta_k, \beta_q^\dagger] = [\gamma_k, \gamma_q^\dagger] = [\delta_k, \delta_q^\dagger] = \delta_{kq}.$$

The coefficients  $C_k^X, S_k^X$ , and the phases  $\phi_k^X$  are chosen so that the off-diagonal terms in Eq. (2) vanish, that is,

$$\begin{aligned}
(C_k^X)^2 &= \frac{1}{2} \left( \frac{P_k^X}{\sqrt{(P_k^X)^2 - |Q_k^X|^2}} + 1 \right), \\
(S_k^X)^2 &= \frac{1}{2} \left( \frac{P_k^X}{\sqrt{(P_k^X)^2 - |Q_k^X|^2}} - 1 \right), \quad (4) \\
\cos^2 \phi_k^X &= \frac{1}{2} \left( 1 \pm \frac{\text{Re } Q_k^X}{|Q_k^X|} \right), \quad \sin^2 \phi_k^X = \frac{1}{2} \left( 1 \mp \frac{\text{Re } Q_k^X}{|Q_k^X|} \right), \quad (5)
\end{aligned}$$

where

$$\begin{aligned}
P_k^A &= \frac{\sin^2 \theta}{2} (C_{k_y} C_{k_z} + C_{k_x} C_{k_y}) - \cos^2 \theta C_{k_x} C_{k_z} + 1, \\
Q_k^A &= -\frac{\sin^2 \theta}{2} (2C_{k_x} C_{k_z} + C_{k_y} C_{k_z} + C_{k_x} C_{k_y}) \\
&\quad + i \cos \theta (C_{k_x} C_{k_y} - C_{k_y} C_{k_z}), \\
P_k^B &= -\frac{\sin^2 \theta}{2} (C_{k_y} C_{k_z} + C_{k_x} C_{k_y}) - \cos^2 \theta C_{k_x} C_{k_z} + 1, \\
Q_k^B &= -\frac{\sin^2 \theta}{2} (2C_{k_x} C_{k_z} - C_{k_y} C_{k_z} - C_{k_x} C_{k_y}) \\
&\quad - i \cos \theta (C_{k_x} C_{k_y} - C_{k_y} C_{k_z}), \\
P_k^C &= \frac{\sin^2 \theta}{2} (C_{k_y} C_{k_z} - C_{k_x} C_{k_y}) + \cos^2 \theta C_{k_x} C_{k_z} + 1, \\
Q_k^C &= -\frac{\sin^2 \theta}{2} (-2C_{k_x} C_{k_z} + C_{k_y} C_{k_z} - C_{k_x} C_{k_y}) \\
&\quad - i \cos \theta (C_{k_x} C_{k_y} + C_{k_y} C_{k_z})
\end{aligned}$$

$$\begin{aligned}
P_k^D &= -\frac{\sin^2 \theta}{2} (C_{k_y} C_{k_z} - C_{k_x} C_{k_y}) + \cos^2 \theta C_{k_x} C_{k_z} + 1, \\
Q_k^D &= -\frac{\sin^2 \theta}{2} (-2C_{k_x} C_{k_z} - C_{k_y} C_{k_z} + C_{k_x} C_{k_y}) \\
&\quad + i \cos \theta (C_{k_x} C_{k_y} + C_{k_y} C_{k_z}). \quad (6)
\end{aligned}$$

Following this procedure, we obtain the diagonal form of the Hamiltonian:

$$\begin{aligned}
H_D &= \sum_{X=A, B, C, D} \sum_k \left( -\frac{1}{2} P_k^X + \frac{1}{2} \sqrt{(P_k^X)^2 - |Q_k^X|^2} \right. \\
&\quad \left. + \sqrt{(P_k^X)^2 - |Q_k^X|^2} x_k^\dagger x_k \right).
\end{aligned}$$

The magnon energy corresponding to branch A is given by

$$\begin{aligned}
\omega_A(k) &\equiv \Omega(C_{k_x}, -C_{k_y}, C_{k_z}) = \{ \gamma^2 [(1 - C_{k_x} C_{k_z})^2 - C_{k_y}^2 (C_{k_x} \\
&\quad - C_{k_z})^2] + \alpha^2 [(1 + C_{k_x} C_{k_y})^2 - C_{k_z}^2 (C_{k_x} + C_{k_y})^2] \\
&\quad + \beta^2 [(1 + C_{k_y} C_{k_z})^2 - C_{k_x}^2 (C_{k_y} + C_{k_z})^2] \}^{1/2}. \quad (7)
\end{aligned}$$

Note that the energy  $\Omega(C_{k_x}, C_{k_y}, C_{k_z})$  is measured in units of the spin-wave velocity, i.e., the prefactor  $v = 4JS$  is understood. The set of spin-wave dispersions as well as the set of parameters of the Bogoliubov transformation is invariant with respect to the simultaneous replacements  $C_{k_i} \Rightarrow -C_{k_i}$  ( $i = x, y, z$ ). First, we observe that the other spectrum branches are straightforwardly deduced from the expression above by changing the signs of the cosines  $C_{k_i}$ :  $\omega_B(k) \equiv \Omega(C_{k_x}, C_{k_y}, C_{k_z})$ ,  $\omega_C(k) \equiv \Omega(C_{k_x}, C_{k_y}, -C_{k_z})$ , and  $\omega_D(k) \equiv \Omega(-C_{k_x}, C_{k_y}, C_{k_z})$ . Besides the parameters of the Bogoliubov transformation (6) are related by the same substitutions. Indeed, the pairs  $(P_k^X, Q_k^X)$  for  $X = B, C, D$  are obtained from  $X = A$  by changing  $C_{k_i}$  into  $-C_{k_i}$  with  $i = y, x, z$ , respectively. Thus, the whole parametrization given by the Bogoliubov transformation obeys this invariance. Other invariances are hidden in these expressions. Restricting ourselves to the set  $\{P_k^X, \omega_X(k); X = A, B, C, D\}$  and to the triple- $\mathbf{k}$  state, we observe that this set is invariant under the permutations  $(C_{k_x}, C_{k_y}, C_{k_z}) \Rightarrow (C_{k_y}, C_{k_z}, C_{k_x})$  and  $(C_{k_x}, C_{k_y}, C_{k_z}) \Rightarrow (C_{k_z}, C_{k_x}, C_{k_y})$ . Indeed, if we denote by (P1) and (P2) respectively, these transformations of the structure factors, we see that according to expressions (6) and (7) and  $\alpha^2 = \beta^2 = \gamma^2$ , (P1) implies

$$\begin{aligned}
(P_k^A, \omega_A(k)) &\Rightarrow (P_k^C, \omega_C(k)), \quad (P_k^B, \omega_B(k)) \Rightarrow (P_k^B, \omega_B(k)), \\
(P_k^C, \omega_C(k)) &\Rightarrow (P_k^D, \omega_D(k)), \quad (P_k^D, \omega_D(k)) \Rightarrow (P_k^A, \omega_A(k)), \quad (8)
\end{aligned}$$

whereas for (P2) we find

$$\begin{aligned} (P_k^A, \omega_A(k)) &\Rightarrow (P_k^D, \omega_D(k)), & (P_k^B, \omega_B(k)) &\Rightarrow (P_k^B, \omega_B(k)), \\ (P_k^C, \omega_C(k)) &\Rightarrow (P_k^A, \omega_A(k)), & (P_k^D, \omega_D(k)) &\Rightarrow (P_k^C, \omega_C(k)). \end{aligned} \quad (9)$$

When a double- $\mathbf{k}$  structure is considered, expressions of the spin-wave spectra and of the parameters of the Bogoliubov transformation, under the exchange (E),  $C_{k_x} \Leftrightarrow C_{k_z}$  ( $C_{k_y}$  being invariant), become

$$(P_k^C, \omega_C(k)) \Leftrightarrow (P_k^D, \omega_D(k)), \quad (10)$$

the two remaining sets  $A$  and  $B$  being unchanged.

### C. Consequences of the spectrum invariance

These invariances have very important consequences. First, let us consider for instance the staggered magnetization  $M(T)$  at finite temperature:

$$\begin{aligned} M(T) &= \frac{1}{N} \sum_i (\langle S_{1i}^z \rangle + \langle S_{2i}^z \rangle + \langle S_{3i}^z \rangle + \langle S_{4i}^z \rangle) \\ &= \frac{1}{N} \left( 4S - \sum_{\lambda=a,b,c,d} \sum_i \langle \lambda_i^\dagger \lambda_i \rangle \right). \end{aligned}$$

Using the Bogoliubov transformation we obtain

$$\begin{aligned} &\sum_{\lambda=a,b,c,d} \sum_i \langle \lambda_i^\dagger \lambda_i \rangle \\ &= \sum_{Xx=A\alpha, B\beta, C\gamma, D\delta} \sum_k \{ [(C_k^X)^2 + (S_k^X)^2] \langle x_k^\dagger x_k \rangle + (S_k^X)^2 \} \\ &+ \sum_{Xx=A\alpha, B\beta, C\gamma, D\delta} \sum_{k>0} C_k^X S_k^X [ \langle (x_k^\dagger)^2 \rangle + \langle (x_k)^2 \rangle \\ &- \langle (x_{-k}^\dagger)^2 \rangle - \langle (x_{-k})^2 \rangle ]. \end{aligned}$$

Obviously, since the expectation values do not depend on the sign of the variable introduced by the Fourier transformation, the staggered magnetization is given by

$$\begin{aligned} M(T) &= \frac{1}{N} \sum_k \left( 4S - \sum_{X=A, B, C, D} [(C_k^X)^2 + (S_k^X)^2] n_k^B(X) \right. \\ &\left. + (S_k^X)^2 \right), \end{aligned} \quad (11)$$

where  $n_k^B(X) = [\exp \beta \omega_X(k) - 1]^{-1}$  is the Bose-Einstein distribution. Let us now consider the staggered magnetization corresponding to a given sublattice. Converting the sum of the Fourier transformation to an integral over the Brillouin zone (BZ), we get from the expression above

$$M_X(T) = \frac{1}{V_0} \int_{BZ} dk_x dk_y dk_z f_X(C_{k_x}, C_{k_y}, C_{k_z}), \quad (12)$$

where  $V_0$  is the volume of the BZ and

$$f_X(C_{k_x}, C_{k_y}, C_{k_z}) = S - [(C_k^X)^2 + (S_k^X)^2] n_k^B(X) - (S_k^X)^2.$$

Of interest to us in the sequel are relations involving the four integrands above along with the expression  $f(C_{k_x}, C_{k_y}, C_{k_z}) = \sum_X f_X(C_{k_x}, C_{k_y}, C_{k_z})$ . They are the explicit forms of the invariance of both the spin-wave spectra and the Bogoliubov transformation (4),(5):

$$\begin{aligned} f_B(C_{k_x}, C_{k_y}, C_{k_z}) &= f_A(C_{k_x}, -C_{k_y}, C_{k_z}), \\ f_C(C_{k_x}, C_{k_y}, C_{k_z}) &= f_A(-C_{k_x}, C_{k_y}, C_{k_z}), \\ f_D(C_{k_x}, C_{k_y}, C_{k_z}) &= f_A(C_{k_x}, C_{k_y}, -C_{k_z}), \end{aligned} \quad (13)$$

$$\begin{aligned} f(C_{k_x}, C_{k_y}, C_{k_z}) &= f(-C_{k_x}, C_{k_y}, C_{k_z}) = f(C_{k_x}, -C_{k_y}, C_{k_z}) \\ &= f(C_{k_x}, C_{k_y}, -C_{k_z}). \end{aligned} \quad (14)$$

Let us take advantage of these properties. First of all, they allow us to get a very satisfactory self-consistent description of the spin-wave physics. Indeed, the main applications of a spin-wave approach are to calculate the reduction of the spontaneous magnetization by quantum fluctuations at  $T=0$ , to determine the temperature dependence of the spontaneous magnetization, and to estimate the critical temperature as the temperature at which this magnetization vanishes. In general, when as in our case they are several sublattices, different choices lead to different solutions, thus rendering the overall framework inconclusive. Now we proceed to prove that these solutions are identical, an important characteristic of this framework. In fact, we have as many possibilities as the number of sublattices to manage the calculation. For example, Eq. (11) gives the solution  $M(T)$  corresponding to the case of considering together the four sublattices. However, it is also possible to work with only one of the four dispersion relations, say,  $\omega_A(k)$ , with the expression of  $M_A(T)$  given by Eq. (12). To see how this is possible we must carefully study the properties of the integration on the BZ.

fcc lattices are in general described by a set of ten integrals. The invariance of the integrand under the transformation  $k_i \Rightarrow -k_i$  allows us to reduce this set to the three following contributions:

$$\begin{aligned} M_A^I(T) &= \frac{1}{V_0} \int_0^\pi dk_z \int_0^{\pi-k_z} dk_x \int_0^{2\pi} dk_y f_A(C_{k_x}, C_{k_y}, C_{k_z}), \\ M_A^{II}(T) &= \frac{1}{V_0} \int_0^\pi dk_z \int_{\frac{\pi}{2}-k_z}^{2\pi} dk_x \int_0^{3\pi-k_x-k_z} dk_y \\ &\quad \times f_A(C_{k_x}, C_{k_y}, C_{k_z}), \\ M_A^{III}(T) &= \frac{1}{V_0} \int_\pi^{2\pi} dk_z \int_0^{3\pi-k_z} dk_x \int_0^{3\pi-k_x-k_z} dk_y \\ &\quad \times f_A(C_{k_x}, C_{k_y}, C_{k_z}), \end{aligned} \quad (15)$$

and the final expression is

$$M_A(T) = 8[M_A^I(T) + M_A^{II}(T) + M_A^{III}(T)].$$

By using the obvious invariance of the integrand

$$f_A(C_{k_x}, C_{k_y}, C_{k_z}) \equiv f_A(-C_{k_x}, -C_{k_y}, -C_{k_z}) \quad (16)$$

and by changing the integration variables  $k_i \Rightarrow 2\pi - k_i$ , it is possible to rewrite these integrals as

$$\begin{aligned} M_A^I(T) &= \frac{1}{V_0} \int_0^\pi dk_z \int_{\pi+k_z}^{2\pi} dk_x \int_0^{2\pi} dk_y f_A(C_{k_x}, C_{k_y}, C_{k_z}), \\ M_A^{II}(T) &= \frac{1}{V_0} \int_0^\pi dk_z \int_0^{\pi+k_z} dk_x \int_0^{\pi+k_x-k_z} dk_y \\ &\quad \times f_A(C_{k_x}, C_{k_y}, C_{k_z}), \\ M_A^{III}(T) &= \frac{1}{V_0} \int_0^\pi dk_z \int_0^{\pi+k_z} dk_x \int_{\pi+k_x-k_z}^{2\pi} dk_y \\ &\quad \times f_A(C_{k_x}, C_{k_y}, C_{k_z}). \end{aligned}$$

Consequently the full integration reads

$$M_A(T) = \frac{8}{V_0} \int_0^\pi dk_z \int_0^{2\pi} dk_x \int_0^{2\pi} dk_y f_A(C_{k_x}, C_{k_y}, C_{k_z}). \quad (17)$$

Now for the choice  $\omega_B(k)$ , the integration on the Brillouin zone is

$$\begin{aligned} M_B(T) &= \frac{1}{V_0} \int_{BZ} dk_x dk_y dk_z f_A(C_{k_x}, -C_{k_y}, C_{k_z}) \\ &= 8[M_B^I(T) + M_B^{II}(T) + M_B^{III}(T)]. \end{aligned}$$

Again, using Eq. (16) we rewrite the first two integrals on the right-hand side (RHS) of the relation above by changing  $C_{k_y} \Rightarrow -C_{k_y}$ . The third integral is modified through  $C_{k_x} \Rightarrow -C_{k_x}$  and  $C_{k_z} \Rightarrow -C_{k_z}$  and the overall result is identical to Eq. (17). Using the same tools it is easy to show that

$$\frac{M(T)}{4} = M_A(T) = M_B(T) = M_C(T) = M_D(T) \quad (18)$$

$$= \frac{8}{V_0} \int_0^\pi dk_z \int_0^{2\pi} dk_x \int_0^{2\pi} dk_y f_A(C_{k_x}, C_{k_y}, C_{k_z}). \quad (19)$$

This proves the self-consistency as mentioned before. Moreover, let us observe that it is possible to extend the integration domain of  $k_z$  to  $[0, 2\pi]$ , thus recovering the overall symmetry between the three variables  $(k_x, k_y, k_z)$ . This means that the cumbersome numerical integration on the fcc BZ is replaced by a simple cubic integration. In order to prove this result it is sufficient to consider the peculiar case of  $M_A(T)$ . After the substitutions  $k_x \Rightarrow 2\pi - k_x$ ,  $k_y \Rightarrow 2\pi - k_y$ , and  $k_z \Rightarrow 2\pi - k_z$  in the first two integrals  $M_A^I(T)$  and  $M_A^{II}(T)$  in Eqs. (15) and the use of Eq. (16) one can easily find that

$$M_A(T) = \frac{8}{V_0} \int_0^\pi dk_z \int_0^{2\pi} dk_x \int_0^{2\pi} dk_y f_A(C_{k_x}, C_{k_y}, C_{k_z}).$$

Besides, as a by-product of the discussion above, we have obtained a powerful tool to build the interacting spin-wave theory of such spin systems, namely, the universality of the different averages over the ground state of the number operators corresponding to the four sublattices:

$$\begin{aligned} \langle n^a \rangle &= \langle n^b \rangle = \langle n^c \rangle = \langle n^d \rangle = \bar{n} \\ &= \frac{1}{V_0} \int_{BZ} \{[(C_k^X)^2 + (S_k^X)^2] n_k^B(X) + (S_k^X)^2\}. \quad (20) \end{aligned}$$

To conclude we establish a general result which is a straightforward consequence of the discussion above. Indeed, it can be stated as a kind of theorem, namely, all integrals of the form  $\mathcal{I}(X) = \int_{BZ} \mathcal{F}_X(C_{k_x}, C_{k_y}, C_{k_z})$  where the expressions  $\mathcal{F}_X(C_{k_x}, C_{k_y}, C_{k_z})$  obey to the invariance properties (16) can be rewritten as an integration over a simple cubic lattice. In addition  $\mathcal{I}(A) = \mathcal{I}(B) = \mathcal{I}(C) = \mathcal{I}(D)$ .

#### D. Perturbative treatment

The various terms of interaction between magnons are treated in the standard way. We require that the number and momentum of magnons always be conserved. Taking the example of two sublattices  $a$  and  $b$ , we have only to consider in the decoupling process pair averages like  $\langle a_k^\dagger a_k \rangle$ ,  $\langle b_k^\dagger b_k \rangle$ ,  $\langle a_k^\dagger b_k \rangle$  and their conjugates. Indeed, annihilating a magnon in the  $a$  sublattice corresponds to the process of creating a magnon on the same sublattice or on another sublattice  $b$ ,  $c$ , or  $d$ . Obviously the modified spin-wave theory introduced by Takahashi<sup>16</sup> for Heisenberg ferromagnets and later applied to antiferromagnets<sup>17</sup> is in agreement with these conservation laws. Then we calculate all kinds of interactions by combining every possible pairs of operators into their averages.

The main hindrance to straightforward calculations is the occurrence in the decoupling process of cumbersome pair averages different from the average of the particle number operator. In some peculiar cases<sup>18</sup> (for recent works see Refs. 19 and 20), the nonlinear spin-wave theory can be computed by using some complicated and, sometimes, obscure self-consistent equations between several pair averages. This obstacle disappears in the framework of the fcc frustrated Heisenberg antiferromagnet. Let us illustrate this statement on the following trilinear expression  $\sum_{\langle i,j \rangle} n_i^a b_j^\dagger$ , where in momentum space it reads

$$\sum_{\langle i,j \rangle} n_i^a b_j^\dagger = \frac{1}{\sqrt{N}} \sum_{k,k'} \cos \frac{(k_y - k'_y)}{2} \cos \frac{(k_z - k'_z)}{2} a_k^\dagger a_k b_{k'-k}^\dagger.$$

In the decoupling process where we employ Wick's theorem, only the pair averages  $\langle a_k^\dagger a_k \rangle$  and  $\langle a_k b_k^\dagger \rangle$  do not vanish and

$$\sum_{\langle i,j \rangle} \langle n_i^a b_j^\dagger \rangle = \frac{1}{\sqrt{N}} \left( z b_0^\dagger \sum_k \langle n_k^a \rangle + 4 a_0^\dagger \sum_k C_{k_y} C_{k_z} \langle a_k b_k^\dagger \rangle \right). \quad (21)$$

Following the same procedure the average  $\sum_{\langle i,j \rangle} n_i^a b_j$  can be written as

$$\sum_{(i,j)} \langle n_i^a b_j \rangle = \frac{1}{\sqrt{N}} \left( z b_0 \sum_k \langle n_k^a \rangle + 4 a_0 \sum_k C_{k_y} C_{k_z} \langle a_k^\dagger b_k \rangle \right). \quad (22)$$

Let us now discuss the last contributions on the RHS of the above equations. As a consequence of the Bogoliubov transformation (4),(5) it follows that

$$\begin{aligned} & \sum_k (\Delta_k^x + \text{H.c.}) \\ &= \frac{1}{2} \sum_k (A_k^\dagger A_k - B_k^\dagger B_k + C_k^\dagger C_k - D_k^\dagger D_k + \text{H.c.}), \\ & \sum_k (\Delta_k^z + \text{H.c.}) \\ &= \frac{1}{2} \sum_k (A_k^\dagger A_k - B_k^\dagger B_k - C_k^\dagger C_k + D_k^\dagger D_k + \text{H.c.}), \\ & \sum_k (\Delta_k^y + \text{H.c.}) \\ &= \frac{1}{2} \sum_k (A_k^\dagger A_k + B_k^\dagger B_k - C_k^\dagger C_k - D_k^\dagger D_k + \text{H.c.}). \end{aligned}$$

These relations allow us to calculate the averages on the RHS of Eqs. (21) and (22). Since

$$\langle X_k^\dagger X_k \rangle = [(\mathcal{C}_k^X)^2 + (\mathcal{S}_k^X)^2] n_k^B(X) + (\mathcal{S}_k^X)^2,$$

it follows that

$$\begin{aligned} F_x(C_{k_x}, C_{k_y}, C_{k_z}) &= 4 C_{k_y} C_{k_z} \langle \Delta_k^x + \text{H.c.} \rangle \\ &= 2 C_{k_y} C_{k_z} [g(C_{k_x}, -C_{k_y}, C_{k_z}) \\ &\quad - g(C_{k_x}, C_{k_y}, C_{k_z}) + g(C_{k_x}, C_{k_y}, -C_{k_z}) \\ &\quad - g(-C_{k_x}, C_{k_y}, C_{k_z})], \quad (23) \end{aligned}$$

where  $g(C_{k_x}, C_{k_y}, C_{k_z}) = \langle B_k^\dagger B_k \rangle$ . Note also that

$$\begin{aligned} F_z(C_{k_x}, C_{k_y}, C_{k_z}) &= 4 C_{k_x} C_{k_y} \langle \Delta_k^z + \text{H.c.} \rangle \\ &= 2 C_{k_x} C_{k_y} [g(C_{k_x}, -C_{k_y}, C_{k_z}) \\ &\quad - g(C_{k_x}, C_{k_y}, C_{k_z}) - g(C_{k_x}, C_{k_y}, -C_{k_z}) \\ &\quad + g(-C_{k_x}, C_{k_y}, C_{k_z})] \quad (24) \end{aligned}$$

and

$$\begin{aligned} F_y(C_{k_x}, C_{k_y}, C_{k_z}) &= 4 C_{k_x} C_{k_z} \langle \Delta_k^y + \text{H.c.} \rangle \\ &= 2 C_{k_x} C_{k_z} [g(C_{k_x}, -C_{k_y}, C_{k_z}) \\ &\quad + g(C_{k_x}, C_{k_y}, C_{k_z}) - g(C_{k_x}, C_{k_y}, -C_{k_z}) \\ &\quad - g(-C_{k_x}, C_{k_y}, C_{k_z})]. \quad (25) \end{aligned}$$

Therefore, the following properties follow directly from Eqs. (23)–(25),

$$\begin{aligned} F_\sigma(C_{k_x}, C_{k_y}, C_{k_z}) &= F_\sigma(-C_{k_x}, C_{k_y}, C_{k_z}) = F_\sigma(C_{k_x}, -C_{k_y}, C_{k_z}) \\ &= F_\sigma(C_{k_x}, C_{k_y}, -C_{k_z}), \end{aligned}$$

and are valid for  $\sigma = x, y, z$ . It then turns out that these expressions satisfy the conditions allowing us to apply the theorem proved in the preceding section, namely,

$$\begin{aligned} \mathcal{I}_\sigma &= \frac{1}{V_0} \int_{BZ} F_\sigma(C_{k_x}, C_{k_y}, C_{k_z}) \\ &\equiv \frac{16}{V_0} \int_0^{2\pi} dk_z \int_0^{2\pi} dk_x \int_0^{2\pi} dk_y F_\sigma(C_{k_x}, C_{k_y}, C_{k_z}). \end{aligned}$$

More specific properties depend on the particular multi- $\mathbf{k}$  structure which is stabilized. For the highly noncollinear triple- $\mathbf{k}$  state, the expressions above satisfy the permutation invariances  $P1$  and  $P2$  [Eqs. (8) and (9)],

$$F_x(C_{k_x}, C_{k_y}, C_{k_z}) = F_z(C_{k_y}, C_{k_z}, C_{k_x}) = -F_y(C_{k_z}, C_{k_x}, C_{k_y}), \quad (26)$$

allowing us to establish [since from Eq. (26) the three variables of integration are equivalent]

$$\mathcal{I}_x = \mathcal{I}_z = -\mathcal{I}_y \equiv \mathcal{I}. \quad (27)$$

For the double- $\mathbf{k}$  state we can observe from the exchange (E) [see Eq. (10)] that

$$\mathcal{I}_x = \mathcal{I}_z, \quad (28)$$

$\mathcal{I}_y$  being unchanged. Finally for the single- $\mathbf{k}$  state we have

$$\mathcal{I}_x = \mathcal{I}_z = 0, \quad (29)$$

since in that case  $\sum_k (\Delta_k^x + \text{H.c.}) = \sum_k (\Delta_k^z + \text{H.c.}) = 0$ .

The next step is to find phenomenological expressions for these integrals. It seems quite reasonable to assume the universality of the pair averages  $\langle \lambda^\dagger \mu \rangle$  and their conjugates, with  $\lambda \neq \mu$  and  $\lambda, \mu = a, b, c$  or  $d$ . Thus they can be written as

$$\begin{aligned} \mathcal{I}_\sigma &= \frac{4}{V_0} \int_{BZ} C_{\sigma'} C_{\sigma''} \langle \lambda^\dagger \mu \rangle = \frac{4}{V_0} \int_{BZ} C_{\sigma'} C_{\sigma''} \{ [(\mathcal{C}_k^B)^2 \\ &\quad + (\mathcal{S}_k^B)^2] n_k^B(B) + (\mathcal{S}_k^B)^2 \}, \quad (30) \end{aligned}$$

where to  $\sigma = (x, y, z)$  corresponds, respectively,  $[\sigma', \sigma''] = ([y, z], [x, z], [x, y])$ . This expression is, of course, consistent with the relations (27) and (28). Indeed,  $g(C_{k_x}, C_{k_y}, C_{k_z}) \equiv [(\mathcal{C}_k^B)^2 + (\mathcal{S}_k^B)^2] n_k^B(B) + (\mathcal{S}_k^B)^2$  with the parametrization corresponding to the triple- $\mathbf{k}$  state has the property

$$g(C_{k_x}, C_{k_y}, C_{k_z}) = g(C_{k_y}, C_{k_z}, C_{k_x}) = g(C_{k_z}, C_{k_y}, C_{k_x}),$$

whereas for the double- $\mathbf{k}$  case one only has

$$g(C_{k_x}, C_{k_y}, C_{k_z}) = g(C_{k_z}, C_{k_y}, C_{k_x}).$$

### III. HEISENBERG ISOTROPIC HAMILTONIAN

Within the spin-wave theory at fourth order to which we shall restrict ourselves, the effective Hamiltonian acquires the form  $\mathcal{H} = \mathcal{H}_1 + \mathcal{H}_2 + \mathcal{H}_3 + \mathcal{H}_4$ , where  $\mathcal{H}_n$  is the term of  $n$ th order in the Bose operators. Here we evaluate the  $1/\sqrt{S}$  and  $1/S$  quantum corrections to  $\mathcal{H}_2$  by taking into account  $\mathcal{H}_3$  and  $\mathcal{H}_4$ . We replace, as explained before, two of the three (four) operators in  $\mathcal{H}_3$  ( $\mathcal{H}_4$ ) with the expectation values in the ground state of  $\mathcal{H}_2$ , ending up with a term which is linear (bilinear) in Bose operators. In the following we prove that the double infinite degeneracy of the ground state persists on the whole antiferromagnetic range of temperature when the third-order terms in the bosonic operators are calculated.

#### A. Odd contribution

Generally, the requirement that the linear part  $\mathcal{H}_1$  of the Hamiltonian vanish leads to classical conditions for the orientation of the equilibrium magnetic moment of each sublattice. When this Hamiltonian describes a frustrated spin system this part disappears whatever may be the angles  $(\vartheta, \varphi)$ , thus betraying the strong degeneracy of the corresponding ground state. The odd term of higher-order  $\mathcal{H}_3$  gives some insight into the relative stability of the magnetic states given by  $\mathcal{H}_1$  or to the permanency of the degeneracy at the quantum level. This term is separated into an imaginary part proportional to

$$\text{Im } \mathcal{H}_3 = \sin \theta (\mathbf{S}_1^\eta \mathbf{S}_2^\rho - \mathbf{S}_1^\rho \mathbf{S}_2^\eta - \mathbf{S}_1^\eta \mathbf{S}_4^\rho + \mathbf{S}_1^\rho \mathbf{S}_4^\eta + \mathbf{S}_2^\eta \mathbf{S}_3^\rho - \mathbf{S}_2^\rho \mathbf{S}_3^\eta + \mathbf{S}_3^\eta \mathbf{S}_4^\rho - \mathbf{S}_3^\rho \mathbf{S}_4^\eta)$$

and a real part

$$\text{Re } \mathcal{H}_3 = -\sin 2\theta (\mathbf{S}_1^\xi \mathbf{S}_3^\rho + \mathbf{S}_1^\rho \mathbf{S}_3^\xi - \mathbf{S}_2^\xi \mathbf{S}_4^\rho - \mathbf{S}_2^\rho \mathbf{S}_4^\xi) + \frac{\sin 2\theta}{2} (\mathbf{S}_1^\xi \mathbf{S}_2^\rho - \mathbf{S}_1^\rho \mathbf{S}_2^\xi + \mathbf{S}_3^\xi \mathbf{S}_4^\rho - \mathbf{S}_3^\rho \mathbf{S}_4^\xi + \mathbf{S}_1^\xi \mathbf{S}_4^\rho - \mathbf{S}_1^\rho \mathbf{S}_4^\xi - \mathbf{S}_2^\xi \mathbf{S}_3^\rho + \mathbf{S}_2^\rho \mathbf{S}_3^\xi).$$

The contribution of third order in bosonic operators, which is of the order  $1/S$  with respect to the linear contribution, takes the form

$$\text{Im } \mathcal{H}_3 = \sqrt{\frac{S}{2}} \sin \theta \sum_{\langle i,j \rangle} \{ (n_i^a - n_i^c) (b_j^\dagger - b_j + d_j - d_j^\dagger) + (n_i^b - n_i^d) (c_j^\dagger - c_j + a_j - a_j^\dagger) \},$$

It is straightforward to evaluate the average of this quantum correction by using Eqs. (20)–(22). This results in

$$\langle \text{Im } \mathcal{H}_3 \rangle = 4 \sqrt{\frac{S}{2}} \sin \theta (a_0^\dagger - a_0 - b_0^\dagger + b_0 + c_0^\dagger - c_0 - d_0^\dagger + d_0) (\mathcal{I}_x - \mathcal{I}_z).$$

Now, we may invoke Eqs. (27)–(29) to prove that  $\langle \text{Im } \mathcal{H}_3 \rangle \equiv 0$ , whatever may be the state considered.

The corresponding real part is given by

$$\begin{aligned} \text{Re } \mathcal{H}_3 = & \sqrt{\frac{S}{8}} \sin 2\theta \sum_{\langle i,j \rangle} \{ (a_i + a_i^\dagger) (n_j^b + n_j^d - 2n_j^c) + (c_i + c_i^\dagger) \\ & \times (n_j^b + n_j^d - 2n_j^a) - (b_i + b_i^\dagger) (n_j^a + n_j^c - 2n_j^d) \\ & - (d_i + d_i^\dagger) (n_j^a + n_j^c - 2n_j^b) \}. \end{aligned}$$

Again using Eqs. (20)–(22),

$$\begin{aligned} (\text{Re } \mathcal{H}_3) = & -2 \sqrt{\frac{S}{2}} \sin 2\theta (a_0^\dagger + a_0 - b_0^\dagger - b_0 + c_0^\dagger + c_0 \\ & - d_0^\dagger - d_0) (\mathcal{I}_x + \mathcal{I}_z + 2\mathcal{I}_y). \end{aligned}$$

This average value is zero for the single- $\mathbf{k}$  and double- $\mathbf{k}$  states since then the prefactor  $\sin 2\theta$  vanishes and for the triple- $\mathbf{k}$  state by using Eq. (27). Thus we have proved that, at this order, the degeneracy of the ground state remains true over the entire AF domain in temperature.

#### B. Quartic contribution

The following is an attempt to go beyond the linear spin-wave theory by considering the interaction  $\mathcal{H}_4$  given by the fourth-order perturbation with respect to the number of bosonic operators. The scalar products of spins containing even products of bosonic operators can be written in the local coordinate system as

$$\mathbf{S}_1 \cdot \mathbf{S}_2 = -(\mathbf{S}_1^\xi \cdot \mathbf{S}_2^\eta + \mathbf{S}_1^\eta \cdot \mathbf{S}_2^\xi) \cos \theta + \mathbf{S}_1^\xi \cdot \mathbf{S}_2^\xi \sin^2 \theta - \mathbf{S}_1^\zeta \cdot \mathbf{S}_2^\zeta \cos^2 \theta, \quad (31)$$

$$\mathbf{S}_1 \cdot \mathbf{S}_3 = (\mathbf{S}_1^\xi \cdot \mathbf{S}_3^\zeta - \mathbf{S}_1^\zeta \cdot \mathbf{S}_3^\xi) \cos 2\theta - \mathbf{S}_1^\eta \cdot \mathbf{S}_3^\eta, \quad (32)$$

$$\mathbf{S}_1 \cdot \mathbf{S}_4 = (\mathbf{S}_1^\xi \cdot \mathbf{S}_4^\eta + \mathbf{S}_1^\eta \cdot \mathbf{S}_4^\xi) \cos \theta + \mathbf{S}_1^\xi \cdot \mathbf{S}_4^\xi \sin^2 \theta - \mathbf{S}_1^\zeta \cdot \mathbf{S}_4^\zeta \cos^2 \theta. \quad (33)$$

Other contributions are easily obtained by changing spin labels. In fact the scalar products of any pairs of spins laying on opposite edges of the basic tetrahedron are identical. Thus  $\mathbf{S}_3 \cdot \mathbf{S}_4$  is deduced from  $\mathbf{S}_1 \cdot \mathbf{S}_2$  by replacing  $1 \rightarrow 3$  and  $2 \rightarrow 4$ , whereas  $\mathbf{S}_2 \cdot \mathbf{S}_4$  ( $\mathbf{S}_2 \cdot \mathbf{S}_3$ ) is deduced from  $\mathbf{S}_1 \cdot \mathbf{S}_3$  ( $\mathbf{S}_1 \cdot \mathbf{S}_4$ ) by the substitutions  $1 \rightarrow 2$  and  $3 \rightarrow 4$  ( $1 \rightarrow 2$  and  $4 \rightarrow 3$ ).

At the fourth order in Bose operators, the term  $\mathbf{S}_1^\xi \mathbf{S}_2^\xi$  takes the form

$$\begin{aligned} \mathbf{S}_1^\xi \mathbf{S}_2^\xi = & \sum_{\langle i,j \rangle} \left[ \left( \frac{S}{2} \right) \left( (a_i b_j + a_i^\dagger b_j + \text{H.c.}) - \frac{1}{8} [a_i^\dagger a_i^2 b_j + a_i^\dagger a_i^2 b_j^\dagger \right. \right. \\ & + (a_i^\dagger)^2 a_i b_j + (a_i^\dagger)^2 a_i b_j^\dagger + a_i b_j^\dagger b_j^2 + a_i (b_j^\dagger)^2 b_j \\ & \left. \left. + a_i^\dagger b_j^\dagger b_j^2 + a_i^\dagger (b_j^\dagger)^2 b_j \right) \right]. \end{aligned} \quad (34)$$

It is straightforward to generalize the averages (21) and (22). For instance, we have

$$\begin{aligned} \left\langle \sum_{\langle i,j \rangle} a_i^\dagger a_i^2 b_j \right\rangle = & \frac{16}{N} \left[ 2 \sum_{k_1} \langle n_{k_1}^a \rangle \sum_{k_2} C_{k_{2y}} C_{k_{2z}} a_{k_2} b_{-k_2} \right. \\ & \left. + \sum_{k_1} C_{k_{1y}} C_{k_{1z}} \langle a_{k_1}^\dagger b_{k_1} \rangle \sum_{k_2} a_{k_2} a_{-k_2} \right]. \end{aligned}$$

Transforming the sum on  $k_2$  in integration over the BZ we obtain

$$\left\langle \sum_{\langle i,j \rangle} a_i^\dagger a_i^2 b_j \right\rangle = \sum_k (8\bar{n} C_{k_y} C_{k_z} a_k b_{-k} + 4\mathcal{I}_x a_k a_{-k}).$$

The other terms in Eq. (34) come out easily and

$$\begin{aligned} \langle \mathbf{S}_1^\xi \mathbf{S}_2^\xi + \mathbf{S}_3^\xi \mathbf{S}_4^\xi \rangle &= \sum_k \left\{ 2S \left( 1 - \frac{\bar{n}}{S} \right) C_{k_y} C_{k_z} (\Sigma_k^x + \Delta_k^x + \text{H.c.}) \right. \\ &\quad \left. - \frac{\mathcal{I}_x}{2} (4\Omega_k + \Lambda_k + \Lambda_k^\dagger) \right\}. \end{aligned} \quad (35)$$

The first term on the RHS above is the same as the one for the noninteracting case in which  $S$  is replaced by  $S(1 - \bar{n}/S)$ . It is straightforward, although lengthy, to generalize the above result to the other scalar products in Eqs. (31)–(33). The final results are

$$\begin{aligned} \langle \mathbf{S}_1^\eta \mathbf{S}_3^\eta + \mathbf{S}_2^\eta \mathbf{S}_4^\eta \rangle &= - \sum_k \left\{ 2S \left( 1 - \frac{\bar{n}}{S} \right) C_{k_x} C_{k_z} (\Sigma_k^y - \Delta_k^y + \text{H.c.}) \right. \\ &\quad \left. - \frac{\mathcal{I}_y}{2} (4\Omega_k - \Lambda_k - \Lambda_k^\dagger) \right\}, \\ \langle \mathbf{S}_1^\xi \mathbf{S}_2^\eta + \mathbf{S}_1^\eta \mathbf{S}_2^\xi + \mathbf{S}_3^\xi \mathbf{S}_4^\eta + \mathbf{S}_3^\eta \mathbf{S}_4^\xi \rangle \\ &= \sum_k \left\{ 4S \left( 1 - \frac{\bar{n}}{S} \right) C_{k_y} C_{k_z} [i(\Sigma_k^x)^\dagger + \text{H.c.}] \right. \\ &\quad \left. - \mathcal{I}_x (i\Lambda_k^\dagger + \text{H.c.}) \right\}. \end{aligned} \quad (36)$$

The calculation of a mean-value-like  $\langle \mathbf{S}_1^\xi \mathbf{S}_2^\xi \rangle$  requires special care. Indeed, at this order such a contribution is given by

$$\langle \mathbf{S}_1^\xi \mathbf{S}_2^\xi \rangle = zS^2 - zS \sum_i (n_i^a + n_i^b) + \left\langle \sum_{\langle i,j \rangle} n_i^a n_j^b \right\rangle.$$

After the Fourier transformation we obtain

$$\begin{aligned} \left\langle \sum_{\langle i,j \rangle} n_i^a n_j^b \right\rangle &= 4\bar{n} \sum_k (n_k^a + n_k^b) \\ &\quad + \frac{4}{N} \sum_{k,q} (C_{k_y - q_y} C_{k_z - q_z} \langle a_k b_k^\dagger \rangle a_q^\dagger b_q + \text{H.c.}). \end{aligned} \quad (37)$$

Because of the natural assumption  $\langle a_k b_k^\dagger \rangle \equiv \langle a_{-k} b_{-k}^\dagger \rangle$  we found

$$\begin{aligned} &\sum_{k,q} (C_{k_y - q_y} C_{k_z - q_z} \langle a_k b_k^\dagger \rangle a_q^\dagger b_q + \text{H.c.}) \\ &= \left( \sum_k C_{k_y} C_{k_z} \langle a_k b_k^\dagger \rangle \sum_q C_{q_y} C_{q_z} a_q^\dagger b_q \right. \\ &\quad \left. + \sum_k S_{k_y} S_{k_z} \langle a_k b_k^\dagger \rangle \sum_q S_{q_y} S_{q_z} a_q^\dagger b_q + \text{H.c.} \right), \end{aligned}$$

where  $S_{k_i} = \sin[k_i/2]$  ( $i=x,y,z$ ). Formally, the average  $\langle \sum_{\langle i,j \rangle} n_i^a n_j^b \rangle$  may be written as

$$\begin{aligned} \left\langle \sum_{\langle i,j \rangle} n_i^a n_j^b \right\rangle &= 4\bar{n} \sum_k (n_k^a + n_k^b) + \left( \mathcal{I}_x \sum_k C_{k_y} C_{k_z} a_k^\dagger b_k \right. \\ &\quad \left. + \mathcal{I}_x' \sum_k S_{k_y} S_{k_z} a_k^\dagger b_k + \text{H.c.} \right), \end{aligned}$$

where the expression of  $\mathcal{I}_x'$  is deduced from  $\mathcal{I}_x$  by replacing the factor  $C_{k_y} C_{k_z}$  by  $S_{k_y} S_{k_z}$ . Now, defining  $F'_x(C_{k_x}, C_{k_y}, C_{k_z}) = S_{k_y} S_{k_z} \langle \Delta_k^x + \text{H.c.} \rangle$  it can be shown that

$$\begin{aligned} F'_x(C_{k_x}, C_{k_y}, C_{k_z}) &= F'_x(-C_{k_x}, C_{k_y}, C_{k_z}) \\ &= -F'_x(C_{k_x}, -C_{k_y}, C_{k_z}) \\ &= -F'_x(C_{k_x}, C_{k_y}, -C_{k_z}) \end{aligned}$$

and more generally  $F'_\sigma(C_{k_\sigma}) = F'_\sigma(-C_{k_\sigma})$  and  $F'_\sigma(C_{k_\omega}) = -F'_\sigma(-C_{k_\omega})$  if  $\omega \neq \sigma$ . Since these expressions obey relation (16), we can apply the theorem stated in the former section, namely,  $\int_{\text{BZ}} F'_\sigma(C_{k_x}, C_{k_y}, C_{k_z}) \equiv 4 \int_0^{2\pi} dk_x \int_0^{2\pi} dk_y \int_0^{2\pi} dk_z F'_\sigma(C_{k_x}, C_{k_y}, C_{k_z})$ . From  $F'_\sigma(C_{k_\omega}) = -F'_\sigma(-C_{k_\omega})$  it follows that  $F'_\sigma(C_{k_\omega}) = 0$ . Then necessarily  $\mathcal{I}'_\sigma = 0$ .

Accordingly we have

$$\begin{aligned} \langle \mathbf{S}_1^\xi \mathbf{S}_2^\xi + \mathbf{S}_3^\xi \mathbf{S}_4^\xi \rangle &= 2zS^2 - zS \left( 1 - \frac{\bar{n}}{S} \right) \sum_k \Omega_k \\ &\quad + \mathcal{I}_x \left( \sum_k C_{k_y} C_{k_z} \Delta_k^x + \text{H.c.} \right). \end{aligned} \quad (38)$$

Now with the help of Eqs. (31)–(33) and according to Eqs. (35), (36), and (38) it is straightforward to calculate  $\mathcal{H}_2 + \mathcal{H}_4$ . The resulting Hamiltonian can be written as in Eq. (2). The new coefficients are

$$P_k^{(2+4)} = 4 \left( 1 - \frac{\bar{n}}{S} \right) - \frac{2}{S} (\mathcal{I}_x + \mathcal{I}_z + 2\mathcal{I}_y) \sin^2 \theta,$$

$$Q_k^{(2+4)} = -\frac{\sin^2 \theta}{2S} (\mathcal{I}_x + \mathcal{I}_z) + \frac{\mathcal{I}_y}{S} \cos^2 \theta,$$

$$R_k^{x(2+4)} = 2 \left( 1 - \frac{\bar{n}}{S} \right) C_{k_y} C_{k_z} (\sin^2 \theta + 2i \cos \theta),$$

$$R_k^{y(2+4)} = 4 \left( 1 - \frac{\bar{n}}{S} \right) C_{k_x} C_{k_z} \sin^2 \theta,$$

$$R_k^{z(2+4)} = 2 \left( 1 - \frac{\bar{n}}{S} \right) C_{k_x} C_{k_y} (\sin^2 \theta - 2i \cos \theta),$$

$$V_k^{x(2+4)} = 2 \left( 1 - \frac{\bar{n}}{S} \right) C_{k_y} C_{k_z} \sin^2 \theta - \frac{\cos^2 \theta}{S} \mathcal{I}_x C_{k_y} C_{k_z},$$



$$V_k^{y(2+4)} = -4 \left(1 - \frac{\bar{n}}{S}\right) C_{k_x} C_{k_z} \cos^2 \theta + \frac{\cos 2\theta}{S} \mathcal{I}_y C_{k_x} C_{k_z},$$

$$V_k^{z(2+4)} = 2 \left(1 - \frac{\bar{n}}{S}\right) C_{k_x} C_{k_y} \sin^2 \theta - \frac{\cos^2 \theta}{S} \mathcal{I}_z C_{k_x} C_{k_y}. \quad (39)$$

Now we are ready to give the main result of this paper. Since the Hamiltonian defined by the above parameters can be diagonalized by the Bogoliubov transformation (3), we can write the corresponding set of spin-wave dispersions. For sake of clarity, we give the formulas for the three allowed spin states. We restrict ourselves to terms which are not quadratic in the integrals  $\mathcal{I}_\lambda$ ,  $\lambda = x, y, z$ . Indeed, as we are discussing now, those terms denoted by  $\langle \Omega^{(4)} \rangle_{II}$  are negligible in the most interesting cases. They can be obtained, if necessary, from the formulas above. Beginning with the single- $\mathbf{k}$  state, recalling that  $\mathcal{I}_x = \mathcal{I}_z = 0$ , and denoting  $\mathcal{I}_y$  by  $\mathcal{I}_1$ ,

$$\omega_B^2(k) \equiv \left(1 - \frac{\bar{n}}{S}\right)^2 [(1 - C_{k_x} C_{k_z})^2 - C_{k_y}^2 (C_{k_x} - C_{k_z})^2] \quad (40)$$

$$+ \frac{\mathcal{I}_1}{2S} \left(1 - \frac{\bar{n}}{S}\right) C_{k_x} C_{k_z} (1 - C_{k_x} C_{k_z}) + \langle \Omega^{(4)} \rangle_{II}. \quad (41)$$

For this state the spin-wave dispersion relations contain two branches, as it should, since there are as many branches as magnetic sublattices. The other branch is deduced from the expression above by changing the sign of  $C_{k_x}$  or  $C_{k_z}$ .

For the double- $\mathbf{k}$  state, by imposing  $\theta = \pi/2$ , Eq. (39), results in

$$\omega_B^2(k) \equiv \left(1 - \frac{\bar{n}}{S}\right)^2 (1 - C_{k_x} C_{k_z})$$

$$\times (1 + C_{k_x} C_{k_z} - C_{k_x} C_{k_y} - C_{k_y} C_{k_z}) - \frac{2}{S} \left(1 - \frac{\bar{n}}{S}\right)$$

$$\times (\mathcal{I}_2 + \mathcal{J}_2) [1 - \frac{1}{2}(C_{k_x} C_{k_y} + C_{k_y} C_{k_z})] - \frac{1}{4S} \left(1 - \frac{\bar{n}}{S}\right)$$

$$\times \mathcal{I}_2 (C_{k_x} C_{k_y} + C_{k_y} C_{k_z} - 2C_{k_x} C_{k_z}) + \langle \Omega^{(4)} \rangle_{II}. \quad (42)$$

This expression depends on two quantities which are defined by  $\mathcal{I}_2 = \mathcal{I}_x = \mathcal{I}_z$  and  $\mathcal{J}_2 = \mathcal{I}_y$ .

Note that  $\theta = \cos^{-1} \sqrt{\frac{1}{3}}$  gives the triple- $\mathbf{k}$  state. The corresponding spectrum branch is

$$\omega_B^2(k) \equiv \left(1 - \frac{\bar{n}}{S}\right)^2 [1 - \frac{2}{3}(C_{k_x} C_{k_y} + C_{k_y} C_{k_z} + C_{k_x} C_{k_z})$$

$$+ \frac{2}{3} C_{k_x} C_{k_y} C_{k_z} (C_{k_x} + C_{k_y} + C_{k_z}) - \frac{1}{3} (C_{k_x}^2 C_{k_y}^2$$

$$+ C_{k_y}^2 C_{k_z}^2 + C_{k_x}^2 C_{k_z}^2)] + \frac{\mathcal{I}_3}{2S} \left(1 - \frac{\bar{n}}{S}\right) [C_{k_x} C_{k_z}$$

$$- \frac{1}{3} (C_{k_x} C_{k_y} + C_{k_y} C_{k_z} + C_{k_x} C_{k_z})^2] + \langle \Omega^{(4)} \rangle_{II}, \quad (43)$$

where  $\mathcal{I}_x = \mathcal{I}_z = -\mathcal{I}_y \equiv \mathcal{I}_3$ . In addition, the other spin-wave frequencies associated with Eqs. (42) and (43) are obtained from these expressions by changing the signs of  $C_{k_x}$ ,  $C_{k_y}$ , and  $C_{k_z}$  respectively.

In both cases the resulting magnon energy assumes the form

$$\omega(k) = \sqrt{\left(1 - \frac{\bar{n}}{S}\right) \left[ \left(1 - \frac{\bar{n}}{S}\right) \Omega^{(2)} + \langle \Omega^{(4)} \rangle_I \right] + \langle \Omega^{(4)} \rangle_{II}}. \quad (44)$$

Let us first discuss the term  $\langle \Omega^{(4)} \rangle_{II}$ . We have to evaluate the importance of the different integrals entering in this formulation. We choose the limit of zero temperature since then they become independent of the Heisenberg coupling  $J$ . In this limit these integrals are given by formulas (30) where now  $n_k^B(B) \Rightarrow 0$ . A numerical evaluation of such quantities, however, is complicated by the singular nature of the integrand. In fact these integrals are analog to the expressions of the zero-point spin reduction within the antiferromagnetic spin-wave theory. They ask for an analytical treatment, a well-known fact in quasi-one-dimensional antiferromagnets.<sup>21</sup> Let us consider the concrete example of  $\mathcal{I}_1$  for the single- $\mathbf{k}$  state:

$$\mathcal{I}_1 \simeq \int_0^{2\pi} dk_z \int_0^{2\pi} dk_x \int_0^{2\pi} dk_y C_{k_x} C_{k_z}$$

$$\times \left[ \frac{1 - C_{k_x} C_{k_z}}{[(1 - C_{k_x} C_{k_z})^2 - C_{k_y}^2 (C_{k_x} - C_{k_z})^2]} - 1 \right].$$

The  $k_y$  integration yields

$$\mathcal{I}_1 \simeq 4 \int_0^{2\pi} dk_z \int_0^{2\pi} dk_x C_{k_x} C_{k_z} \left[ K \left( \frac{C_{k_x} - C_{k_z}}{1 - C_{k_x} C_{k_z}} \right) - 1 \right].$$

Here  $K(k)$  is the complete elliptic integral of the first kind with modulus  $k$ . The analytical property of this function near  $k' = 0$ , where  $k' = \sqrt{(1 - C_{k_x}^2)(1 - C_{k_z}^2)/(1 - C_{k_x} C_{k_z})}$  is known from<sup>22</sup>  $K \simeq \ln(4/k')$ . The divergent logarithmic behavior along the directions  $k_x = 0$  and  $k_z = 0$  cannot be regarded as a wrong indication since the original Heisenberg Hamiltonian corresponds to an unphysical situation. Indeed, when systems experimentally realized are considered, the continuous degeneracy of the ground state has to be lifted by some exchange term stabilizing a multi- $\mathbf{k}$  structure.<sup>12</sup> Then a gap appears in the spin-wave spectrum and there are no more divergencies. Moreover, these quantities decrease as the gap increases, in such a way that the value at the weak gap is a good estimate. This is the reason why we have evaluated the above integrals by introducing at hand a gap of the order of  $4JS/200$ . Assuming  $V_0 = (2\pi)^3$ , we obtain (in  $a^3$  units)

$$\mathcal{I}_1 \simeq -0.84, \quad \mathcal{I}_2 \simeq -0.25, \quad \mathcal{J}_2 \simeq -0.13, \quad \mathcal{I}_3 \simeq -0.12. \quad (45)$$

These values can be interpreted as follows. The contribution of the triple- $\mathbf{k}$  state to the spin reduction is 0.33.<sup>14</sup> With respect to the formulation of this quantity, the integrands are modulated by the factor  $C_{k_i} C_{k_j}$  which diminishes roughly the

result of the integration by one-half order of magnitude. This strong reducing effect is partly compensated by the prefactor of 4 entering in the definition of  $\mathcal{I}_i$ , thus leading to a smaller value. Now the other multi- $\mathbf{k}$  magnetic states are less and less tridimensional. The magnetic moments of the double- $\mathbf{k}$  state lie in a plane, whereas the single- $\mathbf{k}$  configuration is very similar to a canonical one-dimensional AF structure. This explains the differences between the estimations in Eqs. (45). Since the magnon energies are normalized to unity in the sense that  $\Omega(0,0,0)=1$ , we have strong reasons to believe that the contribution  $\langle \Omega^{(4)} \rangle_{II}$  is negligible at least for the triple- $\mathbf{k}$  structure.

Taking into account this approximation, the additional perturbative terms given by  $\mathcal{H}_4$  in Eq. (44) have two consequences. First, they renormalize the magnon energy by the prefactor  $1 - \bar{n}/S$ . As the temperature increases, the second term of this thermal average also increases, causing a decrease in the spin-wave frequencies. But interestingly enough, the overall shape of the magnon modes remains unaffected by the temperature at which they are measured. In other words, for this contribution, lowering the temperature increases the resolution of the frequency spectra. The Hamiltonian  $\mathcal{H}_2 + \mathcal{H}_4$  furnishes also an additive contribution behaving as  $\sqrt{1 - \bar{n}/S}$  and which partly jams the image given above. However, at least for the triple- $\mathbf{k}$  structure, a decrease in temperature of the spin-wave spectra is predicted. This is in agreement with what is called the softening phenomenon of the spin-wave frequencies.<sup>23</sup> This phenomenon is known to be common to most antiferromagnets.

The softening temperature  $T_s$ , the temperature below which the spin waves are stable, is in general greater than the Néel temperature. This is because  $T_N$  is the temperature limit above which the long-range order is destroyed. Indeed, when long-range AF order does not exist, the nearest-neighbor sites still have AF interactions, the correlation length remaining very long. In the present context, if the remaining additional terms which do not contribute to the temperature dependence,  $1 - \bar{n}/S$ , are lacking, the two critical temperatures are identical if, as usual,  $T_N$  is estimated to be the critical temperature at which the magnetization vanishes.

Finally, we make a rough comparison of Eq. (44) with the temperature dependence of the longitudinal and transverse modes in NpBi.<sup>3,24</sup> In fact, we choose the following parametrization for the energy of these magnetic excitations:

$$E = \lambda \sqrt{1 - \frac{\bar{n}}{S}} \sqrt{1 + r - \frac{\bar{n}}{S}}. \quad (46)$$

In addition to the prefactor  $\lambda$ , there is a second parameter in the expression of  $\bar{n}$ , Eq. (20), which measures the effective strength of the AF coupling in the Bose-Einstein distribution  $n_k^B = [\exp(\lambda_1 \omega/T) - 1]^{-1}$ . These parameters are chosen in order to reproduce the observed Néel temperature and the value of the excitation energy at  $T=0$ . Then, the behavior in temperature is given by the ratio  $r$ . The results (Fig. 1) emphasize two points. First a non-negligible contribution  $\langle \Omega^{(4)} \rangle_I$  is required in the expression (44) of the magnon energy. Indeed the dashed curve refers to Eq. (46) with  $r=0$ , whereas the solid curve reproducing the data on the

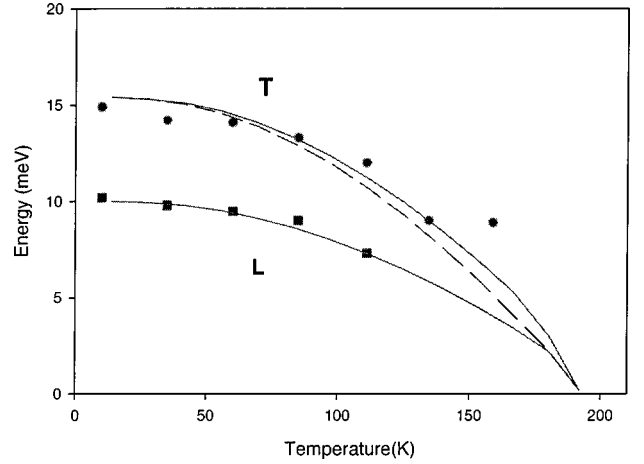


FIG. 1. The temperature dependence of the longitudinal and transverse modes in NpBi.

transverse modes ( $T$ ) is obtained with  $r=0.2$ . Thus, in spite of the freedom in the parameter choice, the necessity of  $r \neq 0$  is established. The parameter set is  $\lambda_T=280$  K, which corresponds to an effective coupling  $J=47$  K, and  $\lambda_1=555$  K ( $J_1=93$  K). Second, the temperature dependence of the data on the longitudinal modes ( $L$ ) can be straightforwardly obtained from the fit on the transverse ones by a constant rescaling, since now  $\lambda_S = \lambda_T/1.54$ , the remainder of the parameter set being unchanged. Note that this observation is qualitatively in agreement with the formulation presented here, Eq. (43), since the variation in temperature of the magnon energies is independent of the branch choice. However, we have to remember that this formulation concerns only the unrealistic description given by  $H_{nn}$ . In particular, the clear difference between the couplings  $\lambda$  and  $\lambda_1$  has to be explained by some model.

#### IV. HEISENBERG SYSTEMS WITH SUPPLEMENTARY INTERACTIONS

Now we discuss the temperature dependence given by the nonlinear spin-wave theory for the Heisenberg model with different types of anisotropy and interactions. First, we shall focus on stabilizing terms for which higher-order contributions can be evaluated easily. We accordingly restrict the discussion to the local single-ion anisotropy, the quadrupolar interaction, and the single-ion anisotropy given by the crystal electric field. These calculations are made for the triple- $\mathbf{k}$  state. A systematic derivation of higher-order properties of other stabilizing terms is a very complicated task. Hence, it seems worthwhile to wait for a particular physical application before attempting a careful analysis. However, it is possible to make some speculations about their behavior in temperature by considering only the “leading terms” giving for  $\mathcal{H}_2$  the prefactor  $1 - \bar{n}/S$ . This will be done for the fourth-order spin couplings.

##### A. Local anisotropy

First, let us discuss the local single-ion anisotropy in the triple- $\mathbf{k}$  configuration. The Hamiltonian which has to be added to  $\mathcal{H}_2$  in order to stabilize this type of configuration<sup>10</sup> has the general form

$$\delta\mathcal{H}_{an} = D_{an} \sum_i \sum_{l=1}^4 (\mathbf{S}_{il} \cdot \mathbf{d}_l)^2,$$

where the index  $i$  refers to the  $i$ th lattice site,  $l$  labeling the four sublattices. The unit vectors  $\mathbf{d}_l$  are defined along the direction of the trigonal axis as  $\mathbf{d}_1 = (1/\sqrt{3})(1,1,1)$ ,  $\mathbf{d}_2 = (1/\sqrt{3})(-1,1,-1)$ ,  $\mathbf{d}_3 = (1/\sqrt{3})(-1,-1,1)$ , and  $\mathbf{d}_4 = (1/\sqrt{3})(1,-1,-1)$ . Here the equilibrium conditions<sup>12</sup> do not depend on the order of the  $1/S$  expansion considered. The former Hamiltonian in the triple- $\mathbf{k}$  structure reduces to<sup>12</sup>

$$\delta\mathcal{H}_{an} = D_{an} S^2 \sum_i \sum_{l=1}^4 (S - n_l^I)^2,$$

resulting in the exact expression

$$\delta\mathcal{H}_{an}^{(2)} + \langle \delta\mathcal{H}_{an}^{(4)} \rangle = -2D_{an} S^3 \left(1 - \frac{\bar{n}}{S}\right) \sum_i \sum_{l=1}^4 n_l^I,$$

which corresponds to consider the leading term in  $S^3$  with the change  $D_{an} \Rightarrow D_{an}(1 - \bar{n}/S)$ . Thus there is no qualitative difference in the leading temperature behavior given by the expansion of the quartic term  $\mathcal{H}_2$ .

### B. Quadrupolar interaction

An analysis of the stability of antiferromagnetically ordered multi- $\mathbf{k}$  structures has shown the importance of the quadrupolar interactions.<sup>25</sup> A model including both crystal-field effects and anisotropic exchange originating from quadrupolar interactions was developed to explain the spin dynamics of several type-I antiferromagnets.<sup>1</sup> We only consider the case of diagonal and isotropic interactions with one effective quadrupolar coupling parameter  $K$ , given by the Hamiltonian

$$\delta\mathcal{H}_Q = K \sum_i \sum_{l=1}^4 (O_{2i}^0[I] O_{2i}^0[I] + 3O_{2i}^2[I] O_{2i}^2[I]),$$

where  $O_n^m$  are Stevens operators:<sup>26</sup>

$$O_2^0[I] = 3(\mathbf{S}_I^z)^2 - S(S+1),$$

$$O_2^2[I] = (\mathbf{S}_I^x)^2 - (\mathbf{S}_I^y)^2.$$

The resulting equilibrium conditions given by the linear term do not depend on the particular sublattice  $l=1, \dots, 4$  considered and are given by

$$6KS^2 \sin 2\theta (x_i + x_i^\dagger) (S+1 - 3S \cos^2 \theta) = 0.$$

Therefore, the quadrupolar interaction stabilizes the three fcc states potentially, the triple- $\mathbf{k}$  state being selected when the spin  $S$  is very large. In that case the strength  $K$  of the quadrupolar contribution is strongly enhanced by a factor  $S^3$ . Now concentrating on this last possibility we find the following expressions in the local coordinate system  $\xi \eta \zeta$ :

$$O_{2i}^0[I] O_{2i}^0[I] = [(\mathbf{S}_I^\xi \mp \sqrt{2}\mathbf{S}_I^\zeta) - S^2]^2,$$

$$3O_{2i}^2[I] O_{2i}^2[I] = [\mathbf{S}_I^\xi \cdot \mathbf{S}_I^\eta + \mathbf{S}_I^\eta \cdot \mathbf{S}_I^\xi \pm \sqrt{2}(\mathbf{S}_I^\xi \cdot \mathbf{S}_I^\eta + \mathbf{S}_I^\eta \cdot \mathbf{S}_I^\xi)]^2,$$

where the upper (lower) sign corresponds to the sublattices  $[1,3]$  ( $[2,4]$ ), respectively. From these expressions the three first quantum corrections are easily calculated. The term  $\delta\mathcal{H}_Q^{(3)}$ , third order in the Bose operators, vanishes. This result indicates the stability of the structure on the whole AF domain in temperature. Moreover, the interacting theory obtained to the fourth order gives similar results to the interacting  $\mathcal{H}_2$  at the same order:

$$\delta\mathcal{H}_Q^{(2)} + \langle \delta\mathcal{H}_Q^{(4)} \rangle = 8KS^3 \left(1 - \frac{\bar{n}}{S}\right) \sum_i \sum_{l=1}^4 n_l^I.$$

### C. Crystal electric field

The single-ion anisotropy can be also provided by the crystal electric field (CEF) in some special cases which we discuss now. For localized  $f$ -electron systems the corresponding Hamiltonian is given by

$$\begin{aligned} \delta\mathcal{H}_{CF} = & \sum_i \sum_{l=1}^4 \{B_4(O_{4i}^0[I] + 5O_{4i}^4[I]) \\ & + B_6(O_{6i}^0[I] - 21O_{6i}^4[I])\}, \end{aligned} \quad (47)$$

where  $B_4$  and  $B_6$  are effective crystal-field parameters. Such contribution has been used to calculate the excitation spectrum of USb by canonical spin-wave theory in the triple- $\mathbf{k}$  state.<sup>1,11</sup> For the  $U^{3+}$  ions the second term on the RHS of Eq. (47) vanishes and, then, the expression above can be approximated by the spin Hamiltonian<sup>11</sup>  $\delta\mathcal{H}_{CF} \approx \mathbf{S}_x^4 + \mathbf{S}_y^4 + \mathbf{S}_z^4$ . Within this approximation the classical stable states given by  $\delta\mathcal{H}_{CF}$  again are determined by the criterion of vanishing linear term,

$$\sin 2\theta (\mathbf{S}^\xi)^3 \mathbf{S}^\xi (\sin^2 \theta - 2 \cos^2 \theta) = 0. \quad (48)$$

They can then be the single- $\mathbf{k}$ , double- $\mathbf{k}$ , or triple- $\mathbf{k}$  state. We must now evaluate the  $1/S$  quantum correction to this result by taking into account  $\delta\mathcal{H}_{CF}^{(3)}$  by replacing two of the three operators in  $\delta\mathcal{H}_{CF}^{(3)}$  with their expectation value. Then the term, which is linear in Bose operators and of next order in  $1/S$ , is

$$\begin{aligned} \langle \delta\mathcal{H}_{CF}^{(3)} \rangle = & -2 \sin \theta \cos^3 \theta \langle (\mathbf{S}^\xi)^3 \mathbf{S}^\xi \rangle - 2 \sin^3 \theta \cos \theta \langle (\mathbf{S}^\xi)^3 \mathbf{S}^\xi \rangle \\ & + 6 \sin \theta \cos \theta \langle (\mathbf{S}^\eta)^2 \mathbf{S}^\xi \mathbf{S}^\xi \rangle, \end{aligned}$$

giving

$$\begin{aligned} \langle \delta\mathcal{H}_{CF}^{(3)} \rangle = & 6S^2 \sqrt{\frac{S}{2}} \sin 2\theta (2 \cos^2 \theta - \sin^2 \theta) \bar{n} \\ & \times (a_0 - b_0 + c_0 - d_0 + \text{H.c.}). \end{aligned}$$

Again the cancellation of this expression gives the same structures as Eq. (48).

Now restricting ourselves to the triple- $\mathbf{k}$  state and considering only the contribution of the magnetic moment  $A$  results in

$$\mathbf{S}_x^4 + \mathbf{S}_y^4 + \mathbf{S}_z^4 = \frac{1}{2} [(\mathbf{S}^\xi)^2 + (\mathbf{S}^\eta)^2]^2 + [\mathbf{S}^\xi]^2 [(\mathbf{S}^\xi)^2 + (\mathbf{S}^\eta)^2] \\ + [(\mathbf{S}^\xi)^2 + (\mathbf{S}^\eta)^2] (\mathbf{S}^\xi)^2 + \frac{1}{3} (\mathbf{S}^\xi)^4.$$

To see how this expression develops as a function of bosonic operators, we might note that  $(\mathbf{S}^\xi)^2 + (\mathbf{S}^\eta)^2 = S(2a^\dagger a + 1)$ . Hence we have the exact result at fourth order:

$$\mathbf{S}_x^4 + \mathbf{S}_y^4 + \mathbf{S}_z^4 = \left( \frac{8S^3}{3} - 2S^2 \right) a^\dagger a \\ - 4S(2S-1) a^\dagger a a^\dagger a + o(a^5).$$

In the limit of large  $S$ , the corresponding term which is bilinear in Bose operators is given by (the three others magnetic moments giving similar results)

$$\delta\mathcal{H}_{CF}^{(2)} + \langle \delta\mathcal{H}_{CF}^{(4)} \rangle \\ = \frac{8S^3}{3} \left( 1 - \frac{3\bar{n}}{S} \right) \sum_i (a_i^\dagger a_i + b_i^\dagger b_i + c_i^\dagger c_i + d_i^\dagger d_i).$$

Thus the temperature dependence given by the CEF is different from those given by  $H_{nn}$ .

#### D. Fourth-order spin couplings

The fourth-order spin couplings are the biquadratic exchange interaction  $(\mathbf{S}_A \cdot \mathbf{S}_B)^2$  (Ref. 7) and the four-spin exchange interaction  $(\mathbf{S}_A \cdot \mathbf{S}_B)(\mathbf{S}_C \cdot \mathbf{S}_D)$  (Ref. 8). The calculation of these interactions among localized spins is greatly simplified if we neglect the terms given by the integrals  $\mathcal{I}_\lambda$ ,  $\lambda = x, y, z$ . Indeed, let us consider for sake of illustration the following contribution:

$$\delta\mathcal{H}_4 \approx \sum_k C_{k_y} C_{k_z} a_k^\dagger b_k \sum_q C_{q_y} C_{q_z} c_q^\dagger d_q.$$

By imposing the Wick theorem, it is decoupled in the following way:

$$\langle \delta\mathcal{H}_4 \rangle \approx \sum_k C_{k_y} C_{k_z} \langle a_k^\dagger b_k \rangle \sum_q C_{q_y} C_{q_z} \langle c_q^\dagger d_q \rangle \\ + \sum_k C_{k_y} C_{k_z} a_k^\dagger b_k \sum_q C_{q_y} C_{q_z} \langle c_q^\dagger d_q \rangle \\ + \sum_k (C_{k_y} C_{k_z})^2 (\langle a_k^\dagger d_k \rangle b_k c_k^\dagger + a_k^\dagger c_k \langle b_k c_k^\dagger \rangle).$$

The first two terms of this decoupling vanish since one can neglect the two sums  $\sum_k C_{k_y} C_{k_z} \langle a_k^\dagger b_k \rangle$  and  $\sum_q C_{q_y} C_{q_z} \langle c_q^\dagger d_q \rangle$  when transformed into integrals. The last term disappears in the thermodynamic limit and we end up with  $\langle \delta\mathcal{H}_{IV} \rangle = 0$ . This result is easily extended to more general contributions. Indeed, the fourth-order terms of this kind of contributions can be roughly divided into two classes. The first one is provided by the product of two quadratic terms, whereas the second comes from the product of a fourth-order term with a

zero-order contribution. The latter one is given by expressions like  $\mathbf{S}^\xi \cdot \mathbf{S}^\xi \sim S^2$ , since then there is no structure factor canceling the integral in Fourier space. Thus it appears that this second class is the source of the  $1/S$  terms which build the prefactor  $1 - t\bar{n}/S$  giving the temperature dependence.

#### E. Four-spin interaction

The four-spin interaction likely to lift the degeneracy of  $H_{nn}$  (Ref. 12) has the form

$$\delta\mathcal{H}_{IV} = J_{IV} [(\mathbf{S}_1 \cdot \mathbf{S}_2)(\mathbf{S}_3 \cdot \mathbf{S}_4) + (\mathbf{S}_1 \cdot \mathbf{S}_4)(\mathbf{S}_2 \cdot \mathbf{S}_3) \\ + (\mathbf{S}_1 \cdot \mathbf{S}_3)(\mathbf{S}_2 \cdot \mathbf{S}_4)]. \quad (49)$$

This interaction gives a perturbative contribution at third order which can be separated into two real parts  $\text{Re } \mathcal{O}_A^{(3)}$  and  $\text{Re } \mathcal{O}_B^{(3)}$ , given, respectively, by  $(\mathbf{S}_1 \cdot \mathbf{S}_2)(\mathbf{S}_3 \cdot \mathbf{S}_4) + (\mathbf{S}_1 \cdot \mathbf{S}_4)(\mathbf{S}_2 \cdot \mathbf{S}_3)$  and  $(\mathbf{S}_1 \cdot \mathbf{S}_3)(\mathbf{S}_2 \cdot \mathbf{S}_4)$ :

$$\text{Re } \mathcal{O}_A^{(3)} \approx 2 \sin \theta \cos^3 \theta \Sigma, \quad \text{Re } \mathcal{O}_B^{(3)} \approx \sin 2\theta \cos 2\theta \Sigma,$$

with

$$\Sigma = \sum_{\langle i,j \rangle} \sum_{\langle i',j' \rangle} [(a_i + a_i^\dagger)(n_{i'}^b + n_{i'}^c + n_{i'}^d) \\ + (c_i + c_i^\dagger)(n_{i'}^a + n_{i'}^b + n_{i'}^d) - (b_i + b_i^\dagger)(n_{i'}^a + n_{i'}^c + n_{i'}^d) \\ - (d_i + d_i^\dagger)(n_{i'}^a + n_{i'}^b + n_{i'}^c) + Tl_i],$$

where

$$Tl_i = \frac{1}{4} (a_i^\dagger n_i^a + n_i^a a_i + c_i^\dagger n_i^c + n_i^c c_i - b_i^\dagger n_i^b \\ - n_i^b b_i - d_i^\dagger n_i^d - n_i^d d_i).$$

The suppression of this contribution requires

$$\sin 2\theta (\cos^2 \theta + \cos 2\theta) = 0, \quad (50)$$

and the stabilizing conditions remain unaffected by the third-order term in the Bose operators.

Concerning the temperature dependence, let us consider the first contribution on the RHS of Eq. (49). The calculation proceeds in several steps. The first one is to evaluate the term of second order in  $1/S$ :

$$[(\mathbf{S}_1 \cdot \mathbf{S}_2)(\mathbf{S}_3 \cdot \mathbf{S}_4)]^{(2)} = S^2 \cos^2 \theta [\cos \theta (\mathbf{S}_1^\xi \cdot \mathbf{S}_2^\eta + \mathbf{S}_1^\eta \cdot \mathbf{S}_2^\xi + \mathbf{S}_3^\xi \cdot \mathbf{S}_4^\eta + \mathbf{S}_3^\eta \cdot \mathbf{S}_4^\xi)^{(2)} - \sin^2 \theta (\mathbf{S}_1^\xi \cdot \mathbf{S}_2^\xi + \mathbf{S}_3^\xi \cdot \mathbf{S}_4^\xi)^{(2)} + \cos^2 \theta (\mathbf{S}_1^\zeta \cdot \mathbf{S}_2^\zeta + \mathbf{S}_3^\zeta \cdot \mathbf{S}_4^\zeta)^{(2)}].$$

Next we consider the fourth-order term

$$[(\mathbf{S}_1 \cdot \mathbf{S}_2)(\mathbf{S}_3 \cdot \mathbf{S}_4)]^{(4)} = S^2 \cos^2 \theta [\cos \theta (\mathbf{S}_1^\xi \cdot \mathbf{S}_2^\eta + \mathbf{S}_1^\eta \cdot \mathbf{S}_2^\xi + \mathbf{S}_3^\xi \cdot \mathbf{S}_4^\eta + \mathbf{S}_3^\eta \cdot \mathbf{S}_4^\xi)^{(4)} - \sin^2 \theta (\mathbf{S}_1^\xi \cdot \mathbf{S}_2^\xi + \mathbf{S}_3^\xi \cdot \mathbf{S}_4^\xi)^{(4)} + \cos^2 \theta (\mathbf{S}_1^\zeta \cdot \mathbf{S}_2^\zeta + \mathbf{S}_3^\zeta \cdot \mathbf{S}_4^\zeta)^{(4)}] + \cos^2 \theta [\cos \theta (\mathbf{S}_1^\xi \cdot \mathbf{S}_2^\eta + \mathbf{S}_1^\eta \cdot \mathbf{S}_2^\xi)^{(2)} - \sin^2 \theta (\mathbf{S}_1^\xi \cdot \mathbf{S}_2^\xi)^{(2)}] (\mathbf{S}_3^\zeta \cdot \mathbf{S}_4^\zeta)^{(2)} + \cos^2 \theta [\cos \theta (\mathbf{S}_3^\xi \cdot \mathbf{S}_4^\eta + \mathbf{S}_3^\eta \cdot \mathbf{S}_4^\xi)^{(2)} - \sin^2 \theta (\mathbf{S}_3^\xi \cdot \mathbf{S}_4^\xi)^{(2)}] (\mathbf{S}_1^\zeta \cdot \mathbf{S}_2^\zeta)^{(2)} + \cos^4 \theta (\mathbf{S}_1^\zeta \cdot \mathbf{S}_2^\zeta)^{(2)} (\mathbf{S}_3^\zeta \cdot \mathbf{S}_4^\zeta)^{(2)},$$

and finally, we compute the average of this fourth order term by using the general result  $\langle (\mathbf{S}_A \cdot \mathbf{S}_B)^{(4)} \rangle = -(\bar{n}/S)(\mathbf{S}_A \cdot \mathbf{S}_B)^{(2)}$ . It is now a matter of some algebra to find out how this contribution transforms,

$$\langle [(\mathbf{S}_1 \cdot \mathbf{S}_2)(\mathbf{S}_3 \cdot \mathbf{S}_4)]^{(4)} \rangle = -\frac{3\bar{n}}{S} [(\mathbf{S}_1 \cdot \mathbf{S}_2)(\mathbf{S}_3 \cdot \mathbf{S}_4)]^{(2)},$$

in such a way that this result can be generalized to the other terms in Eq. (49) so that

$$\delta\mathcal{H}_{IV}^{(2)} + \langle \delta\mathcal{H}_{IV}^{(4)} \rangle \simeq \left(1 - \frac{3\bar{n}}{S}\right) \delta\mathcal{H}_{IV}^{(2)}. \quad (51)$$

#### F. Biquadratic contribution

In the case of the biquadratic contribution

$$\delta\mathcal{H}_{BIQ} = J_{BIQ} [(\mathbf{S}_1 \cdot \mathbf{S}_2)^2 + (\mathbf{S}_3 \cdot \mathbf{S}_4)^2 + (\mathbf{S}_1 \cdot \mathbf{S}_4)^2 + (\mathbf{S}_2 \cdot \mathbf{S}_3)^2 + (\mathbf{S}_1 \cdot \mathbf{S}_3)^2 + (\mathbf{S}_2 \cdot \mathbf{S}_4)^2],$$

this supplements the linear terms by adding imaginary and real contributions. The calculation is quite lengthy and involves complicated expressions. However, the resulting stabilizing condition again remains identical to that found at first order of the expansion, i.e., the condition (50). Again we derive the  $T$  dependence given by the fourth-order term, following the same route as before. Taking the example of the first term we get successively

$$\begin{aligned} \frac{[(\mathbf{S}_1 \cdot \mathbf{S}_2)^2]^{(2)}}{2} &= S^2 \cos^2 \theta [\cos \theta (\mathbf{S}_1^\xi \cdot \mathbf{S}_2^\eta + \mathbf{S}_1^\eta \cdot \mathbf{S}_2^\xi)^{(2)} - \sin^2 \theta (\mathbf{S}_1^\xi \cdot \mathbf{S}_2^\xi)^{(2)} + \cos^2 \theta (\mathbf{S}_1^\zeta \cdot \mathbf{S}_2^\zeta)^{(2)}], \\ \frac{[(\mathbf{S}_1 \cdot \mathbf{S}_2)^2]^{(4)}}{2} &= S^2 \cos^2 \theta [\cos \theta (\mathbf{S}_1^\xi \cdot \mathbf{S}_2^\eta + \mathbf{S}_1^\eta \cdot \mathbf{S}_2^\xi)^{(4)} - \sin^2 \theta (\mathbf{S}_1^\xi \cdot \mathbf{S}_2^\xi)^{(4)} + \cos^2 \theta (\mathbf{S}_1^\zeta \cdot \mathbf{S}_2^\zeta)^{(4)}] \\ &+ \cos^2 \theta \left[ \cos \theta (\mathbf{S}_1^\xi \cdot \mathbf{S}_2^\eta + \mathbf{S}_1^\eta \cdot \mathbf{S}_2^\xi)^{(2)} - \sin^2 \theta \right. \\ &\left. \times (\mathbf{S}_1^\xi \cdot \mathbf{S}_2^\xi)^{(2)} + \frac{\cos^2 \theta}{2} (\mathbf{S}_1^\zeta \cdot \mathbf{S}_2^\zeta)^{(2)} \right] (\mathbf{S}_1^\zeta \cdot \mathbf{S}_2^\zeta)^{(2)}. \end{aligned}$$

These relations yield to  $\langle [(\mathbf{S}_1 \cdot \mathbf{S}_2)^2]^{(4)} \rangle = -(3\bar{n}/S)[(\mathbf{S}_1 \cdot \mathbf{S}_2)^2]^{(2)}$ . The other terms follow the same law. Hence the biquadratic contribution behaves like the four-spin contribution

$$\delta\mathcal{H}_{BIQ}^{(2)} + \langle \delta\mathcal{H}_{BIQ}^{(4)} \rangle \simeq \left(1 - \frac{3\bar{n}}{S}\right) \delta\mathcal{H}_{BIQ}^{(2)}.$$

#### V. CONCLUSION

In many cases the theory of interacting magnons requires a complicated formalism which is not only laborious to handle but where formal difficulties may hide the physics (for a review on the spin waves in magnetic insulators, see the work of Kaganov and Chubukov<sup>27</sup>). Clearly this is not the case for the frustrated AF Heisenberg model for the fcc type-I configuration. Let us recapitulate what we have learned about the behavior in temperature of the spin-wave frequencies. The perturbative treatment leads in great part to a renormalization of the magnetic characteristics at nonzero temperature. Indeed, the additional terms give, at least for the triple- $\mathbf{k}$  configuration, a temperature law of the generic form  $(\sqrt{1-t\bar{n}/S}\sqrt{a+b(1-t\bar{n}/S)})$  where  $\bar{n}$  is a Bose function and  $t$  a constant which is  $t_{nn} = 1$  for  $H_{nn}$ . In the case of systems experimentally realized, this Hamiltonian comes always with, at least, a supplementary contribution which lifts the extensive degeneracy of the ground state and specifies the AF spin state. We have been successful in proving that the leading form of the temperature dependence  $(1-\bar{n}/S)$  is not modified by the local anisotropy or the quadrupolar interactions. On the contrary, the CEF and the fourth-order spin couplings, four spin interaction, and biquadratic contributions behave in a very different way since now  $t_{CEF} = t_{IV} = t_{BIQ} = 3$ . These behaviors are in agreement with what is called the softening phenomenon of spin-wave frequencies: in most antiferromagnets the initial effect of the temperature is to decrease the spin-wave frequencies.

The discovery of invariances displayed by the magnon spectra has allowed us to carry out the calculations presented here. It is important to note that there is, at mean, one another model where the magnon spectra exhibit analog properties. It concerns the Heisenberg antiferromagnet where both the nearest-neighbor (NN) interactions and the next-nearest-neighbor (NNN) interactions are studied on a cubic

and square lattices.<sup>28</sup> In the latter case, one branch of the spin-wave spectra is given by

$$\omega_{1k} = 2S \left[ \{ 2\lambda (1 + \cos k_1 \cos k_2) + \cos k_1 + \cos k_2 \} \right. \\ \left. \times \{ 2\lambda (1 - \cos k_1 \cos k_2) + \cos \theta (\cos k_1 - \cos k_2) \} \right]^{1/2},$$

where the parameter  $\lambda$  represents the relative strength of the NNN interactions to the NN ones, the angle  $\theta$  characterizing the AF state considered. Starting from the expression above it is easy to see<sup>28</sup> that the three other branches are deduced by changing, respectively,  $\cos k_1 \Rightarrow -\cos k_1$ ,  $\cos k_2 \Rightarrow -\cos k_2$ , and  $\cos k_{1,2} \Rightarrow -\cos k_{1,2}$ . The three-dimensional spectra display similar properties.<sup>28</sup> It would be worthwhile to study the consequences of such invariances on the  $1/S$  expansion<sup>29</sup> of this model.

Measurements of low-frequency magnetic excitations of type-I compounds with an AF ordering of triple- $\mathbf{k}$  nature exist for USb (Refs. 1 and 30) and NpBi (Ref. 3) and are essentially limited to very low temperature.<sup>31</sup> They are, however, of great interest since an unambiguous phenomenological interpretation is still lacking.<sup>1,3,11,12</sup> Data on the temperature dependence of these excitations are also available for both the uranium antimonide<sup>2</sup> and the neptunium pnictide NpBi.<sup>3,24</sup> Contrary to NpBi, the low-frequency magnetic response of USb does not follow the expected temperature effect which, usually, decreases the spin-wave frequencies. In fact the initial change in the spin-wave spectrum upon raising the temperature up to  $\sim 80$  K is the *increasing* of the spin-wave frequencies. However, according to the model of Jensen and Bak<sup>11</sup> and some qualitative arguments given in

Ref. 1, the presence of more terms in the spin Hamiltonian than the minimal number given by the NN contribution along with a stabilizing term is expected. In particular, the crystal-field interaction is necessary to describe the spin-wave spectra of uranium antimonide at  $T=0$  K. It is therefore very amazing that a completely different behavior with temperature is predicted here, rendering this striking increase in the spin-wave frequencies of USb hardly understandable. There is, however, a general agreement between this analysis and the data. Indeed, the stability of the triple- $\mathbf{k}$  magnetic structure on the whole AF domain has been observed for USb,<sup>1,2</sup> NpBi,<sup>3</sup> and MnTe<sub>2</sub>.<sup>5</sup> This has to be compared with the remarkable permanence of the equilibrium conditions with respect to the temperature.

As a by-product of this analysis, the set of tools necessary to calculate the spin-wave frequencies when a stabilizing term is added to  $H_{nn}$  is given here and the road towards a meaningful and detailed analysis of experimental data is now actually open. This also asks for new experimental efforts to explore the magnetic properties of these compounds.

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<sup>1</sup>B. Halg and A. Furrer, Phys. Rev. B **34**, 6258 (1986).

<sup>2</sup>M. Hagen, W.G. Stirling, and G.H. Lander, Phys. Rev. B **37**, 1846 (1988).

<sup>3</sup>F. Bourdarot, J. Bossy, P. Burlet, B. Fak, P. Mochanesi, J. Rebizant, L.P. Regnault, J.C. Spirlet, and O. Vogt, Phys. Rev. B **56**, 14 029 (1997).

<sup>4</sup>P. Burlet, F. Bourdarot, J. Rossat-Mignod, J.P. Sanchez, J.C. Spirlet, J. Rebizant, and O. Vogt, Physica B **180-181**, 131 (1992).

<sup>5</sup>P. Burlet, E. Ressouche, B. Malaman, R. Welter, J.P. Sanchez, and P. Vulliet, Phys. Rev. B **56**, 14 013 (1997).

<sup>6</sup>N. Kasai, Y. Nishihara, and S. Ogawa, J. Phys. Soc. Jpn. **51**, 452 (1982).

<sup>7</sup>A. Yoshimori and S. Inagaki, J. Phys. Soc. Jpn. **44**, 101 (1978); **50**, 769 (1981).

<sup>8</sup>A. Yosida and S. Inagaki, J. Phys. Soc. Jpn. **50**, 3268 (1981).

<sup>9</sup>K. Hirai, J. Magn. Magn. Mater. **70**, 112 (1987).

<sup>10</sup>I. Hase, H. Fukuda, and A. Yoshimori, J. Phys. Soc. Jpn. **50**, 774 (1981).

<sup>11</sup>J. Jensen and P. Bak, Phys. Rev. B **23**, 6180 (1981).

<sup>12</sup>J-P. Ader, Phys. Rev. B **65**, 014411 (2002).

<sup>13</sup>T. Holstein and H. Primakoff, Phys. Rev. **58**, 1098 (1940).

<sup>14</sup>T. Oguchi, H. Nishimori, and Y. Taguchi, J. Phys. Soc. Jpn. **54**, 449 (1985).

<sup>15</sup>B.W. Char, K.O. Geddes, G.H. Gonnet, B.L. Leong, M.B. Mona-

gan, and S.M. Watt, *First Leaves: A Tutorial Introduction to Maple V* (Springer-Verlag, New York, 1992).

<sup>16</sup>M. Takahashi, Suppl. Theor. Phys. **87**, 233 (1986); Phys. Rev. B **36**, 3791 (1987).

<sup>17</sup>M. Takahashi, Phys. Rev. B **36**, 3791 (1987); J.E. Hirsch and S. Tang, *ibid.* **40**, 4769 (1989); S. Tang, M.E. Lazzouni, and J.E. Hirsch, *ibid.* **40**, 5000 (1987).

<sup>18</sup>A.E. Jacobs, T. Nikuni, and H. Shiba, J. Phys. Soc. Jpn. **62**, 4066 (1993).

<sup>19</sup>Q. Yuan, Phys. Lett. A **275**, 481 (2000).

<sup>20</sup>J. An, C-D. Gong, and H-Q. Lin, J. Phys.: Condens. Matter **13**, 115 (2001).

<sup>21</sup>D. Welz, J. Phys.: Condens. Matter **5**, 3643 (1993).

<sup>22</sup>I.S. Gradshteyn and I.M. Ryzhik, *Tables of Integrals, Series and Products* (Academic Press, New York, 1965).

<sup>23</sup>Y.K. Wu and Y. Song, Phys. Rev. B **49**, 9639 (1994).

<sup>24</sup>F. Bourdarot (private communication).

<sup>25</sup>M. Amara and P. Morin, Phys. Rev. B **205**, 379 (1995).

<sup>26</sup>K.W.H. Stevens, Proc. Phys. Soc., London, Sect. A **65**, 209 (1952).

<sup>27</sup>M.I. Kaganov and A.V. Chubukov, Sov. Phys. Usp. **30**, 1015 (1987).

<sup>28</sup>K. Kubo and T. Kishi, J. Phys. Soc. Jpn. **60**, 567 (1990).

<sup>29</sup>T. Oguchi and H. Kitatani, J. Phys. Soc. Jpn. **59**, 3322 (1990).

<sup>30</sup>G.H. Lander and W.G. Stirling, Phys. Rev. B **32**, 436 (1980).

<sup>31</sup>T. Chattopadhyay and H. Fjellvag, Phys. Lett. A **120**, 44 (1987).