PHYSICAL REVIEW B, VOLUME 65, 014411

Magnetic order in the frustrated Heisenberg model for the fcc type-I configuration

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(Received 21 May 2001; published 30 November 2001)

The Heisenberg model for the fcc type-I configuration with various types of interactions is studied with use of the noninteracting spin-wave theory. It is shown that local anisotropy, four-spin exchange interactions, and biquadratic interactions can lift the continuous degeneracy of the ground state through the stabilization of a noncollinear magnetic state. Moreover, in the particular case of a pyrite structure, a Dzyaloshinsky-Moriya term or a symmetric anisotropic exchange stabilizes the double-**k** configuration. The single-**k** state is also discussed. We prove, in particular, that quantum fluctuations favor such a case. Consequences for the magnetic moment reduction and for the spin-wave spectra of the presence of such stabilizing terms are calculated and compared with data. Experiments seem to indicate that the minimal model built from the nearest-neighbor Heisenberg interaction and a stabilizing term is not satisfactory.

DOI: 10.1103/PhysRevB.65.014411 PACS number(s): 75.10.Jm

I. INTRODUCTION

Physically realizable[1–3] type-I fcc antiferromagnets are a typical example 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 $K_1=(2\pi/a)(1,0,0)$, $K_2=(2\pi/a)(0,1,0)$, and $K_3=(2\pi/a)(0,0,1)$, where a is the fcc lattice constant. This structure can then be in single-**k**, double-**k**, or triple-**k** states. The triple-**k** structure is an extension of the 120° spin structure on the triangular lattice to the three-dimensional case. In the noncollinear double-**k** state, shown on Fig. 1, the spin moments are along a diagonal of two planes. In the collinear single-**k** structure the spins of the double-**k** structure align themselves antiferromagnetically in the planes. Finally, the canonical antiferromagnetic state is realized when the spin moments are antiferromagnetically aligned following one axis of the crystal.

Since the number of internal degrees of freedom for the ground state for a cluster of p,n component spins is known to be $n_0=p(n-1)-2n$, the ground state given by the simplest isotropic Heisenberg model including only nearest-neighbor interactions has a two-dimensional infinite degeneracy. This extensive degeneracy must be lifted by additional unknown terms in the Hamiltonian. No simple general principles are available to select these terms. For a recent review on this subject, see the work of Oja and Lounasmaa.[4]

The selection of the ground state by quantum fluctuations for isotropic interactions was already studied in the spin-wave calculation of Oguchi *et al.*[5] They showed numerically that quantum fluctuations favor a single-**k** structure. However this approach is questionable since it yields a unique spin-wave dispersion relation, whereas a true calculation would exhibit spin-wave dispersion relations containing, as they should as many branches in the spin-wave spectrum as there are magnetic sublattices in the unit cell. The present work confirms and gives an analytical proof that the zero-point oscillations of the spin waves stabilize the state with a collinear ordering corresponding to the single-**k** state. This proof is borrowed from the paper of Kubo and Kishi[6] assuming ordering due to quantum fluctuations in the Heisenberg antiferromagnet with both nearest-neighbor and next-nearest neighbor interactions.

Concerning the terms to be added to the isotropic Heisenberg model to lift the degeneracy, the great number of studies and the fact that, nevertheless, nothing is well established reflect the difficulty of the problem. First of all, an isotropic next-nearest-neighbor interaction does not classically induce any angular dependence, since it connects two sublattices of the same species. Such a contribution was already considered in Ref. 6, where it was shown that a ‘‘strip’’ state with collinear spin ordering is stabilized by zero-point spin-wave fluctuations. Now let us discuss the early work of Heinilä and Oja.[7] They studied the general form of an anisotropic nearest-neighbor spin-spin exchange consistent with a pyrite structure. In this structure, which has a cubic symmetry, the magnetic ions are cast in a fcc lattice. The symmetry around each magnetic ion in the unit cell is trigonal. The suggestion

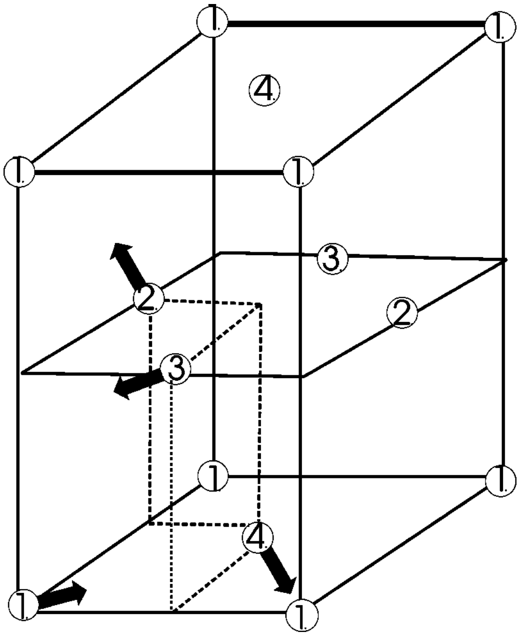

FIG. 1. The double-**k** magnetic structure in the fcc compound $MnTe_2$.

of Ref. 7 is that the Dzyaloshinskii-Moriya (DM) interaction[8,9] can stabilize the triple-**k** structure observed in $MnTe_2$ (Ref. 10) when the temperature T is slightly below the Néel temperature T_N. First we determine the kind of DM interaction compatible with the pyrite structure, and then show that this form is unable to stabilize a triple-**k** state at $T=0$, in contradiction with the antiferromagnetic order determined through Mössbauer investigations as well as neutron experiments.[10] But this term remains a candidate to explain double-**k** ordering, as well as anisotropic nearest-neighbor coupling.

Effects due to biquadratic and four-spin exchange interactions in fcc spin lattices were first discussed, to our knowledge, by Yoshimori and Inagaki[11] and Yosida and Inagaki[12] in a classical treatment of the Heisenberg Hamiltonian with particular reference to the compound NiS_2. The same model was used by Hirai[13] to explain the systematic change of magnetic structures among the intermetallic compounds MnS_2, $MnSe_2$ and $MnTe_2$. These effects are studied here by employing the spin-wave theory.

Finally we discuss the alternative possibilities given by the anisotropy energies. The problem of dipolar anisotropy was considered by Jensen and Bak,[14] who employed spin-wave theory to study the USb compound (uranium antimonide). In Ref. 15 some particular anisotropy energy of the form $\delta\mathcal{H}=\Sigma_{i=1,4}(\mathbf{S}_\nu\cdot\mathbf{d}_\nu)^2$ was considered in a mean-field framework. The magnetic ions $\mathbf{S}_\nu$ being located at δ_ν in the unit cell $\delta_1=(0,0,0)$, $\delta_2=(0,1/2,1/2)$, $\delta_3=(1/2,0,1/2)$, and $\delta_4=(1/2,1/2,0)$, we define the unit vectors $\mathbf{d}_\nu$ 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 $\mathbf{d}_4=(1/\sqrt{3})(1,-1,-1)$. Again corresponding spin-wave theories are developed, and the role of these contributions in order to stabilize noncollinear structures is clarified.

This paper is organized as follows. In Sec. II we give a general formalism for this kind of frustrated magnet to study the dynamical quantities at $T=0$. This formulation is used in Sec. III to show that quantum fluctuations select the single-**k** configuration. We consider (Sec. IV) anisotropic nearest-neighbor exchange interactions in the pyrite structure. Only the anticollinear double-**k** state can be stabilized. Section V is dedicated to a study of the four-spin exchange interactions and biquadratic couplings. In Sec. VI we treat the case of local single-ion anisotropy. To discuss neutron-scattering experiments, in Sec. VII we calculate the magnetic moment reduction in the ground state, and compare our numerical results with experiment. In Sec. VIII the general characteristics of the spin-wave spectra are calculated and discussed. A rough comparison with experimental magnetic spectra is given. We compare also our results with the theoretical work of Jensen and Bak.[14] Finally Sec. IX gives a summary of the present work and some insight for future investigations.

II. LINEAR SPIN-WAVE THEORY

A. Hamiltonian

In order to consider single-**k**, double-**k**, and triple-**k** states simultaneously, we choose in full generality the direction of the magnetizations on sublattices $l=1,2,3,4$ located on the unit cell at δ_l, as (α,β,γ), $(-\alpha,\beta,-\gamma)$, $(-\alpha,-\beta,\gamma)$, 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-**k** state is given by $\alpha=\beta=\gamma=1/\sqrt{3}$, and corresponds to the antiferromagnetic state observed in the $MnTe_2$ compound.[10] Fixing, for instance, $\theta\equiv\pi/2$, in order to obtain coplanar structure, the double-**k** model is recovered by putting $\varphi=\pi/4$. Finally the single-**k** model is obtained from $\theta=0$.

The Hamiltonian we start from reads

$$H_{nn}=J\sum_{\langle i,j\rangle}\mathbf{S}_i\cdot\mathbf{S}_j, \tag{1}$$

with $J>0$, and where $\mathbf{S}_i$ represents a spin located at the ith site. The summation $\langle i,j\rangle$ is taken over all nearest-neighbor pairs. Let us recall the crystal structure of fcc. Each site has 12 nearest-neighbor sites which belong four by four to the other three sublattices. Following Ref. 5 we denote the interaction between a and b sites by the ab bond. The total number of bonds is $6N$, where N is the total number of sites. Sometimes this structure is called a tetrahedral arrangement. Thus H_{nn} can be written as

$$\begin{aligned}H_{nn}=J\sum_{\langle i,j\rangle}&(\mathbf{S}_{i1}\cdot\mathbf{S}_{j2}+\mathbf{S}_{i1}\cdot\mathbf{S}_{j3}+\mathbf{S}_{i1}\cdot\mathbf{S}_{j4}\\&+\mathbf{S}_{i2}\cdot\mathbf{S}_{j3}+\mathbf{S}_{i2}\cdot\mathbf{S}_{j4}+\mathbf{S}_{i3}\cdot\mathbf{S}_{j4}).\end{aligned}$$

We follow the standard calculation within the linear spin-wave theory based on the Holstein-Primakoff representation.[16] First we rewrite the Hamiltonian in the local coordinate system. Taking a particular spin $\mathbf{S}_l$, we denote its equilibrium direction by ζ, the direction perpendicular to this and the z axis by ξ, and the third direction by η. Thus we define a local coordinate system $\xi\eta\zeta$ at each site of the lattice. Then we express the resulting Hamiltonian in terms of the spin-wave operators by introducing four kinds of Holstein-Primakoff operators a, b, c, and d for sublattices 1, 2, 3, and 4 respectively. Neglecting terms of more than second order of operators, we obtain the following expression of the Hamiltonian in terms of the Fourier transforms of the Bose operators:

$$\begin{aligned}\frac{H}{JS}=&-2NS+\frac{1}{4}\sum_k(n_k^a+n_k^b+n_k^c+n_k^d)\\&+\sum_k[\mathcal{A}_{yz}C_yC_z(a_kb_{-k}+c_kd_{-k})+\mathcal{A}_{xy}C_xC_y\\&\times(b_kc_{-k}+a_kd_{-k})+\mathcal{A}_{xz}C_xC_z(a_kc_{-k}+b_kd_{-k})+\text{H.c}]\\&+\sum_k[\mathcal{B}_{yz}C_yC_z(a_k^\dagger b_k+c_k^\dagger d_k)+\mathcal{B}_{xy}C_xC_y(b_k^\dagger c_k+a_k^\dagger d_k)\\&-\mathcal{B}_{xz}C_xC_z(a_k^\dagger c_k+b_k^\dagger d_k)+\text{H.c.}],\end{aligned} \tag{2}$$

where $\mathcal{A}_{yz}=\frac{1}{2}(1-2\cos^2\theta\sin^2\varphi+\cos 2\varphi)+i\cos\theta\sin 2\varphi$, $\mathcal{A}_{xy}=\frac{1}{2}(1-2\cos^2\theta\cos^2\varphi-\cos 2\varphi)-i\cos\theta\sin 2\varphi$, $\mathcal{A}_{xz}$

$=\sin^2\theta$, $\mathcal{B}_{yz}=\sin^2\theta\sin^2\varphi$, $\mathcal{B}_{xy}=\sin^2\theta\cos^2\varphi$, $\mathcal{B}_{xz}=\cos^2\theta$, $C_i=\cos[a(k_i/2)]$ $(i=x,y,z)$, and $n_k^a=a_k^\dagger a_k$. The summation of the wave vector **k** is done over the first Brillouin zone of fcc. Note that we have neglected the linear terms of the operators, since they will disappear automatically when the classical spin orientations are in equilibrium.

B. Exact spectrum

To actually calculate the spin wave spectrum we have to find a transformation of Bose operators that would diagonalize Hamiltonian (2). This transformation is a Bogoliubov-Valatin transformation involving a column vector of eight operators $V_k^+\equiv(a_k^\dagger\, b_k^\dagger\, c_k^\dagger\, d_k^\dagger\, a_{-k}\, b_{-k}\, c_{-k}\, d_{-k})$. To ensure the conservation of Bose commutation relations, we have to diagonalize the matrix $\mathbf{g}M$, where $\mathbf{g}$ is the diagonal matrix

$$\begin{pmatrix} I & \\ & -I \end{pmatrix},$$

I being the 4×4 unit matrix, and where the matrix $\mathcal{M}$ is

$$\mathcal{M}=\begin{pmatrix} \mathcal{M}_1 & \mathcal{M}_2 \\ \mathcal{M}_2^* & \mathcal{M}_1 \end{pmatrix}.$$

The expression of the real matrix $\mathcal{M}_1$ is

$$\mathcal{M}_1=\begin{pmatrix} 1 & \mathcal{B}_{yz}C_yC_z & -\mathcal{B}_{xz}C_xC_z & \mathcal{B}_{xy}C_xC_y \\ \mathcal{B}_{yz}C_yC_z & 1 & \mathcal{B}_{xy}C_xC_y & -\mathcal{B}_{xz}C_xC_z \\ -\mathcal{B}_{xz}C_xC_z & \mathcal{B}_{xy}C_xC_y & 1 & \mathcal{B}_{yz}C_yC_z \\ \mathcal{B}_{xy}C_xC_y & -\mathcal{B}_{xz}C_xC_z & \mathcal{B}_{yz}C_yC_z & 1 \end{pmatrix},$$

whereas the complex matrix $\mathcal{M}_2$ looks like

$$\mathcal{M}_2=\begin{pmatrix} 0 & \mathcal{A}_{yz}^*C_yC_z & \mathcal{A}_{xz}C_xC_z & \mathcal{A}_{xy}^*C_xC_y \\ \mathcal{A}_{yz}^*C_yC_z & 0 & \mathcal{A}_{xy}^*C_xC_y & \mathcal{A}_{xz}C_xC_z \\ \mathcal{A}_{xz}C_xC_z & \mathcal{A}_{xy}^*C_xC_y & 0 & \mathcal{A}_{yz}^*C_yC_z \\ \mathcal{A}_{xy}^*C_xC_y & \mathcal{A}_{xz}C_xC_z & \mathcal{A}_{yz}^*C_yC_z & 0 \end{pmatrix}.$$

The eigenvalues are doubly degenerate solutions of a very messy fourth-order equation. Nevertheless, and although we were unable to calculate the exact form of the Bogoliubov transformation, the explicit expressions of the solutions are obtained analytically through the MAPLE mathematical manipulation language[17] for the three structures. They are quite simple, nicely reflecting the symmetry of the problem. Indeed, our calculation yields to spin-wave dispersion relations containing four branches for the two noncollinear states and two branches for the single-**k** state—as it should, since there are as many branches in the spin-wave spectrum as there are magnetic sublattices. The eigenvalue which is symmetric with respect to permutations of the cosines can be written as

$$\begin{aligned}\omega_1(k)&\equiv\Omega(C_x,C_y,C_z)\\ &=(\gamma^2[(1-C_xC_z)^2-C_y^2(C_x-C_z)^2]\\ &\quad+\alpha^2[(1-C_xC_y)^2-C_z^2(C_x-C_y)^2]\\ &\quad+\beta^2[(1-C_yC_z)^2-C_x^2(C_y-C_z)^2])^{1/2}. \qquad (3)\end{aligned}$$

The other spectrum branches are straightforwardly deduced from the expression above by changing the signs of C_x, C_y, and C_z respectively:

$$\omega_2(k)=\Omega(-C_x,C_y,C_z),\quad \omega_3(k)=\Omega(C_x,-C_y,C_z),$$
$$\omega_4(k)=\Omega(C_x,C_y,-C_z). \qquad (4)$$

The energies are measured in units of the spin-wave velocity, i.e., the prefactor $v=4JS$ is understood. As expected from the original classical spin configuration, it is satisfactory that this set of spin waves displays the full symmetry between x, y, and z in the triple-**k** direction $(1/\sqrt{3},1/\sqrt{3},1/\sqrt{3})$. Obviously, the above energies obey the usual invariance under the changes $k_i\Rightarrow -k_i$.

Around the point Γ, $k=(0,0,0)$, located at the center of the Brillouin zone, the explicit expressions of the magnon energies above are

$$\omega_1(k)=\frac{1}{4}\sqrt{(\alpha k_xk_y)^2+(\beta k_yk_z)^2+(\gamma k_xk_z)^2},$$

$$\omega_2(k)=\sqrt{(\alpha k_z)^2+(\gamma k_y)^2},$$

$$\omega_3(k)=\sqrt{(\alpha k_z)^2+(\beta k_x)^2},\qquad \omega_4(k)=\sqrt{(\beta k_x)^2+(\gamma k_y)^2}.$$

These results show the presence of both ferromagnetic and antiferromagnetic aspects in the present system, the quadratic behavior with wave vector k indicating the ferromagnetic character of the $\omega_1(k)$ mode.

C. Analytical way

Now, based on these results, we develop an analytical approach in order to tackle more complicated expressions of the Hamiltonian. The starting approximation[5] is to take the sublattices to be identical. Although we have four magnetic ions in the basic cell, we do not distinguish them when we make the Fourier transformation. The resulting effective spin-wave Hamiltonian H_{eff} is expressed in term of the Fourier transforms of two Bose operators α_k and $\alpha_k^\dagger$ as

$$H_{eff}=-2JS^2+4SJ \times\sum_k\left(P_k\alpha_k^{\dagger}\alpha_k+\frac{1}{2}Q_k\alpha_k\alpha_{-k}+\frac{1}{2}Q_k^{\dagger}\alpha_k^{\dagger}\alpha_{-k}^{\dagger}\right), \tag{5}$$

where

$$P_k=\mathcal{B}_{yz}C_yC_z+\mathcal{B}_{xy}C_xC_y-\mathcal{B}_{xz}C_xC_z+1 \tag{6}$$

and

$$Q_k=\mathcal{A}_{yz}C_yC_z+\mathcal{A}_{xy}C_xC_y+\mathcal{A}_{xz}C_xC_z\,. \tag{7}$$

The spin-wave operators ξ_k and $\xi_k^{\dagger}$ are defined through the Bogoliubov transformations[5]

$$\alpha_k=\frac{1}{\sqrt{2}}[\mathcal{C}_k(\xi_k+\xi_{-k})+\mathcal{S}_k(\xi_k^{\dagger}-\xi_{-k}^{\dagger})]\exp(-i\Phi_k),\quad k>0,$$

$$\alpha_{-k}=\frac{1}{\sqrt{2}}[\mathcal{C}_k(\xi_k-\xi_{-k})+\mathcal{S}_k(\xi_k^{\dagger}+\xi_{-k}^{\dagger})]\exp(-i\Phi_k),\quad k>0,$$

$$\alpha_0=(\mathcal{C}_0\xi_0+\mathcal{S}_0\xi_0^{\dagger})\exp(-i\Phi_0),$$

where $\mathcal{C}_k=\cosh\Theta_k$ and $\mathcal{S}_k=\sinh\Theta_k$, and

$$[\xi_k,\xi_{k'}^{\dagger}]=\delta_{k,k'}\,,\quad [\xi_k,\xi_{k'}]=[\xi_k^{\dagger},\xi_{k'}^{\dagger}]=0.$$

The coefficients $\mathcal{C}_k,\mathcal{S}_k$, and Φ_k are chosen so that the off-diagonal terms vanish:

$$\mathcal{C}_k^2=\frac{1}{2}\left(\frac{P_k}{\sqrt{P_k^2-|Q_k|^2}}+1\right),\quad \mathcal{S}_k^2=\frac{1}{2}\left(\frac{P_k}{\sqrt{P_k^2-|Q_k|^2}}-1\right),$$

$$\cos^2\Phi_k=\frac{1}{2}\left(1\pm\frac{\operatorname{Re}Q_k}{|Q_k|}\right),\quad \sin^2\Phi_k=\frac{1}{2}\left(1\mp\frac{\operatorname{Re}Q_k}{|Q_k|}\right).$$

The diagonal Hamiltonian reads

$$H_D=\sum_k\left(-\frac{1}{2}P_k+\sqrt{P_k^2-|Q_k|^2}+\sqrt{P_k^2-|Q_k|^2}\,\xi_k^{\dagger}\xi_k\right).$$

The spin-wave dispersion can then be evaluated to give

$$\omega_3(k)\equiv\Omega(C_x,-C_y,C_z)=\{\gamma^2[(1-C_xC_z)^2-C_y^2\times(C_x-C_z)^2]+\alpha^2[(1+C_xC_y)^2-C_z^2(C_x+C_y)^2]+\beta^2[(1+C_yC_z)^2-C_x^2(C_y+C_z)^2]\}^{1/2}. \tag{8}$$

We note that the solution of the approximated method coincides with $\omega_3(k)$. Thus by this method we can recover the exact result when it is realized that, from the underlying (three-dimensional) symmetry, the complete set of solutions can be obtained from a particular solution by some substitutions of the cosines C_i. Indeed, to obtain $\omega_1(k)$ one has to substitute $C_y\Rightarrow -C_y$ in the expression above, whereas $\omega_2(k)$ and $\omega_4(k)$ are deduced from $\omega_3(k)$ by changing the signs of C_z and C_x respectively. Thus the complete set of solutions is obtained from the approximate result straightforwardly. This provides us with a systematic way to build up the spin-wave theory when more complicated Heisenberg Hamiltonians having the general expression of Eq. (5) are considered.

D. Self-consistency of the calculation

In general, the integrand occuring in a spin-wave calculation based on the framework described before depends on the integration variables through the cosines C_i in a very specific way. Indeed, the spin-wave dispersions [Eqs. (3) and (4)] as well as the parameters of the Bogoliubov transformation [Eqs. (6) and (7)] are invariant with respect to the changes $C_i\Rightarrow -C_i$, $i=x,y,z$. This invariance, which we now assume to be displayed by the integrand $f(C_x,C_y,C_z)$, has two important consequences: first a considerable simplification of the integration on the Brillouin zone; and, second, a very satisfactory self-consistent description of the spin-wave physics. Indeed, the main applications of a spin-wave calculation 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, there are several sublattices, different equations lead to different solutions, thus rendering the overall framework inconclusive.

Now we proceed to prove that, within the method of calculation we have adopted here, these solutions are in fact identical, a very nice characteristic of this framework. Let us state the problem very precisely. In fact, we have as many possibilities as the number of sublattices to manage the calculation. Indeed, we have just presented a solution corresponding assuming the four sublattices to be identical, and which leads to the $\omega_3(k)$ spin-wave dispersion and the Bogoliubov parameters [Eqs. (6) and (7)]. However it is also possible to work with one of the three remaining dispersions, say $\omega_2(k)$, the corresponding Bogoliubov parameters being deduced from Eqs. (6) and (7) by the same changes allowing us to obtain $\omega_2(k)$ from $\omega_3(k)$. Then the expression of a given integral on the Brillouin zone, when the dispersion chosen is $\omega_2(k)$, looks like

$$\mathcal{I}(\omega_2)=\int\int\int_{BZ}dk_xdk_ydk_zf(-C_x,C_y,C_z),$$

knowing that the choice $\omega_1(k)$ gives

$$\mathcal{I}(\omega_1)=\int\int\int_{BZ}dk_xdk_ydk_zf(C_x,C_y,C_z).$$

The Brillouin zone of the fcc lattice is described in general by a set of ten integrals. The invariance of the integrand, under the usual changes $k_i\Rightarrow -k_i$ allows us to reduce this set to the three integrals

$$I_1(f)=\int_0^{\pi/a}dk_z\int_0^{(\pi/a)-k_z}dk_x\int_0^{2\pi/a}dk_yf(-C_x,C_y,C_z),$$

$$I_2(f)=\int_0^{\pi/a}dk_z\int_{(\pi/a)-k_z}^{2\pi/a}dk_x\int_0^{3\pi/a-k_x-k_z}\times dk_y f(-C_x,C_y,C_z),$$

$$I_3(f)=\int_{\pi/a}^{2\pi/a}dk_z\int_0^{(3\pi/a)-k_z}dk_x\int_0^{(3\pi/a)-k_x-k_z}\times dk_y f(-C_x,C_y,C_z),$$

and the final expression is

$$\mathcal{I}(\omega_2)=8[I_1(f)+I_2(f)+I_3(f)].$$

Using the invariance of the integrand,

$$f(C_x,C_y,C_z)\equiv f(-C_x,-C_y,-C_z), \tag{9}$$

it is possible to rewrite these integrals as

$$I_1(f)=\int_0^{\pi/a}dk_z\int_{(\pi/a)+k_z}^{2\pi/a}dk_x\int_0^{2\pi/a}dk_y f(C_x,C_y,C_z),$$

$$I_2(f)=\int_0^{\pi/a}dk_z\int_0^{(\pi/a)+k_z}dk_x\int_0^{(\pi/a)+k_x-k_z}dk_y f(C_x,C_y,C_z),$$

$$I_3(f)=\int_0^{\pi/a}dk_z\int_0^{(\pi/a)+k_z}dk_x\int_{(\pi/a)+k_x-k_z}^{2\pi/a}dk_y f(C_x,C_y,C_z).$$

Consequently the full integration reads

$$\mathcal{I}(\omega_2)=8\int_0^{\pi/a}dk_z\int_0^{2\pi/a}dk_x\int_0^{2\pi/a}dk_y f(C_x,C_y,C_z). \tag{10}$$

Now for the choice $i=3$, the integration on the Brillouin zone is

$$\mathcal{I}(\omega_3)=\int\int\int_{BZ}dk_xdk_ydk_z f(C_x,-C_y,C_z)$$
$$=8[I_1(f)+I_2(f)+I_3(f)].$$

Again, using Eq. (9) we rewrite the first two integrals by changing $C_y\Rightarrow -C_y$. The third integral is modified by the changes $C_x\Rightarrow -C_x$ and $C_z\Rightarrow -C_z$, and the overall result is identical to Eq. (10). Using the same tools it is easy to obtain

$$\mathcal{I}(\omega_1)=\mathcal{I}(\omega_2)=\mathcal{I}(\omega_3)=\mathcal{I}(\omega_4)$$
$$=8\int_0^{\pi/a}dk_z\int_0^{2\pi/a}dk_x\int_0^{2\pi/a}dk_y f(C_x,C_y,C_z). \tag{11}$$

This proves the self-consistency claimed at the beginning of this subsection. Moreover, in the present framework, we can replace the cumbersome numerical integration on the fcc Brillouin zone by a simple cubic integration. Thus we finally obtain a powerful tool to build the interacting spin-wave theory of such spin systems.

E. $\mathcal{C}$-parity invariance

There is a set of transformations on the boson operators leaving Hamiltonian (2) unchanged. Indeed, let us consider a pair of sublattices (a,b). The transformations

$$\mathcal{C}a_k\mathcal{C}^{-1}=-d_k,\quad \mathcal{C}b_k\mathcal{C}^{-1}=-c_k$$

amount to interchanging pairs of sublattices (a,b) and (d,c) while making a π rotation of the ion spins about the x axis. The invariance properties of the cell unit under rotations of the crystal axis allow us to obtain two other transformations of the same kind. By exchanging the sublattices (a,b) for (c,d) and making a π rotation of the spins about the z axis, we obtain

$$\mathcal{C}a_k\mathcal{C}^{-1}=c_k,\quad \mathcal{C}b_k\mathcal{C}^{-1}=d_k,$$

whereas

$$\mathcal{C}a_k\mathcal{C}^{-1}=-b_k,\quad \mathcal{C}c_k\mathcal{C}^{-1}=-d_k$$

corresponds to changing (a,c) to (b,d) and to executing a rotation of the spins about the y axis. These laws are straightforwardly readable in the expressions for the sublattices in the local coordinate system $\xi\eta\zeta$. They allow us to discard eventual terms which would be incompatible with the rotational symmetry of the unit cell of the crystal. For instance, in a general way, they forbid a spin Hamiltonian which would not be sign invariant under these transformations. They cease to be good selection rules when the environment of each metal atom is not compatible with the assumed invariant interchanges. This is the case of the mineral pyrite structure, which can be considered as a fcc grouping of metal atoms and anion pairs. The important changes in the coupling between the magnetic ions brought by the presence of these four anion pairs were analyzed in Ref. 7. The $\mathcal{C}$-parity invariance was already introduced in the context of double perovskite materials.[18] Such compounds, the physics of which are governed by electrons which hop among or are localized on metal ions, are described by a Hamiltonian combining both antiferromagnetic exchange and double exchange. In order to take all of these effects into account at the same time, $\mathcal{C}$-parity invariance seems a very useful tool. We think that this invariance is a first-step toward a first-principles calculation for selecting terms in the Hamiltonian.

III. ORDERING DUE TO QUANTUM FLUCTUATIONS

Taking into account the four branches of spin waves, the shift of the energy ΔE of the ground state due to quantum effects is given by

$$\frac{\Delta E}{4SJ}=\sum_{l=1,4}\sum_k\left(-\frac{1}{2}P_k^l+\sqrt{(P_k^l)^2-|Q_k^l|^2}\right).$$

The l index refers to the branch of spin waves. The expressions of P_k^3 and Q_k^3 are given in Eqs. (6) and (7). As noted in Sec. II, the other quantities P_k^l and Q_k^l, for $l=2$ and 4, are deduced from P_k^3 and Q_k^3 by changing the sign of C_z and C_x

respectively, whereas P_k^1 and Q_k^1 are obtained from Eqs. (6) and (7) by changing the sign of C_y. The shift ΔE becomes

$$\frac{\Delta E}{4SJ}=\sum_k \left(-2+\sum_{l=1,4}\omega_l(k)\right). \tag{12}$$

Let us point out that the strength of the interaction and the spin of the metallic ions involved appear as a prefactor. We now show that this contribution stabilizes the state with a collinear ordering, i.e., the single-**k** state. The calculation parallels that of Kubo and Kishi on quantum fluctuations in the frustrated Heisenberg model with next-nearest-neighbor interactions.[6] We first define two independent variables $\tau=(\alpha^2+\beta^2)/2$ and $\sigma=(\alpha^2-\beta^2)/2$. Then by exchanging the summing variables k_x and k_z, we keep a formally invariant τ whereas σ becomes $-\sigma$. It follows readily that

$$\sum_k \omega_3(k,\sigma,\tau)=\sum_k \omega_3(k,-\sigma,\tau).$$

It is then straightforward to deduce

$$\sigma\frac{\partial}{\partial\sigma}\sum_k \omega_3(k)<0.$$

for $\sigma\neq 0$.

In the same way, we easily see that

$$\sum_k \omega_1(k,\sigma,\tau)=\sum_k \omega_1(k,-\sigma,\tau),$$

$$\sum_k [\omega_2(k,\sigma,\tau)+\omega_4(k,\sigma,\tau)]$$

$$=\sum_k [\omega_2(k,-\sigma,\tau)+\omega_4(k,-\sigma,\tau)].$$

Finally we have

$$\sigma\frac{\partial}{\partial\sigma}\sum_k \Delta E<0.$$

This means that ΔE takes its minimum at the minimum or maximum of $\alpha^2-\beta^2$ for a fixed value of $\alpha^2+\beta^2$. From the overall symmetry of the set of energy spectra, the same result holds if we consider (β^2,γ^2) or (γ^2,α^2) as independent variables instead of (α^2,β^2). Thus the minimum of ΔE can occur only at $(\alpha^2,\beta^2,\gamma^2)=(1,0,0)$ or $(0,1,0)$ or $(0,0,1)$. These three states correspond to three equivalent two-sublattice structures with collinear ordering, i.e., the canonical antiferromagnetic states. For instance the choice $(0,0,1)$ leads to the following spectra

$$\omega_a(k)=\sqrt{(1-C_xC_z)^2-C_y^2(C_x-C_z)^2},$$

$$\omega_b(k)=\sqrt{(1+C_xC_z)^2-C_y^2(C_x+C_z)^2}. \tag{13}$$

This quantum effect can also be estimated numerically by computing the shift in energy of the ground state, as a function of the angle θ. For the sake of simplicity, we have set $\varphi=\pi/4$ in the calculations. Obviously the actual conclusion is confirmed, the zero-point energy favoring the angle of magnetization $\theta=0$, an already known result.[5]

IV. PYRITE STRUCTURE, DM TERM, AND SYMMETRIC ANISOTROPIC EXCHANGES

A. Pyrite structure

In this section we consider that the spin-spin interaction Hamiltonian contains, in addition to a nearest-neighbor exchange interaction, an antisymmmetric part, i.e., the DM exchange

$$\delta H=\sum_{\langle i,j\rangle}\mathbf{D}'_{ij}\cdot(\mathbf{S}_i\times\mathbf{S}_j),$$

where the components of the $\mathbf{D}'_{ij}$ vectors (18 constant parameters introduced *ab initio*) are restricted by the symmetry of the crystal. The most general forms compatible with the symmetry transformations allowed under the pyrite structure were studied in Ref. 7. Each bond ij is characterized by an orthonormal basis (s_{ij},t_{ij},u_{ij}) such that the basis for the bond qr is $(s_{qr},t_{qr},u_{qr})=(Rs_{ij},Rt_{ij},Ru_{ij})$, where the matrix R transforms the two nearest-neighbor bonds into one another. This basis is locally well defined through the geometry of the pyrite structure given by the environment of Te atoms which are grouped in pairs with the magnetic ions in the center. In general, the magnetic ions have six nearest neighbors that form an octahedron with the metal atom at its center.

For instance let us discuss the case of the bond 12. In the general frame the two nearby anion pairs of the Mn^{2+} ions 1 and 2 have the locations $(\pm u,\mp u,1\mp u)$ and $(\pm u,1\pm u,\mp u)$ respectively. This induces the local frame

$$s_{12}=\frac{1}{\sqrt{2}}(0,-1,1),\quad t_{12}=(-1,0,0),\quad u_{12}=\frac{1}{\sqrt{2}}(0,1,1).$$

At the end we obtain the parametrization

$$A_{ij}^{DM}=(r_{ij}\cdot u_{ij})[D'_1(u_{ij}s_{ij}-s_{ij}u_{ij})$$
$$+D'_2(u_{ij}t_{ij}-t_{ij}u_{ij})+D'_3(s_{ij}t_{ij}-t_{ij}s_{ij})],$$

where

$$r_{ij}=\frac{r_j-r_i}{|r_j-r_i|},$$

r_i (r_j), being the positions of the ions i and j respectively.

Finally, the resulting DM vectors associated with the set of bonds are

$$\mathbf{D}_{12}=\begin{pmatrix} D_1 \\ -D_2 \\ D_3 \end{pmatrix}, \quad \mathbf{D}_{13}=\begin{pmatrix} -D_3 \\ -D_1 \\ D_2 \end{pmatrix}, \quad \mathbf{D}_{14}=\begin{pmatrix} -D_2 \\ D_3 \\ D_1 \end{pmatrix}, \tag{14}$$

$$\mathbf{D}_{23}=\begin{pmatrix} D_2 \\ D_3 \\ -D_1 \end{pmatrix}, \quad \mathbf{D}_{24}=\begin{pmatrix} -D_3 \\ D_1 \\ -D_2 \end{pmatrix}, \quad \mathbf{D}_{34}=\begin{pmatrix} -D_1 \\ D_2 \\ D_3 \end{pmatrix}, \tag{15}$$

where $D_2=(D_2'+D_3')/\sqrt{2}$ and $D_3=(D_2'-D_3')/\sqrt{2}$. These vectors can be gathered by pairs $(\mathbf{D}_{12},\mathbf{D}_{34})$, $(\mathbf{D}_{13},\mathbf{D}_{24})$, and $(\mathbf{D}_{14},\mathbf{D}_{23})$. Each member of the pair is deduced from the other by letting component z, x, or y, respectively, remain fixed and changing the signs of the two other components. This is the particular formulation required by the symmetry of the pyrite structure.

B. Spin-wave analysis

Linear terms in the boson operators appear in the Hamiltonian when it is written in the Holstein-Primakoff representation. The spin configurations which are kept in balance by this Hamiltonian have to annihilate the coefficients of these linear terms. With the parametrization above, these coefficients are of the form $C_1D_1+C_2D_2+C_3D_3$, where

$$\begin{aligned} C_1=&-(\mathbf{S}_1^\eta+\mathbf{S}_2^\eta+\mathbf{S}_3^\eta+\mathbf{S}_4^\eta)\cos\theta\sin\varphi+(\mathbf{S}_1^\xi+\mathbf{S}_2^\xi+\mathbf{S}_3^\xi+\mathbf{S}_4^\xi)\\ &\times\cos 2\theta\cos\varphi-(\mathbf{S}_1^\eta-\mathbf{S}_2^\eta+\mathbf{S}_3^\eta-\mathbf{S}_4^\eta)\\ &\times(\sin\theta\cos 2\varphi+\cos\theta\cos\varphi)-(\mathbf{S}_1^\xi+\mathbf{S}_3^\xi-\mathbf{S}_2^\xi-\mathbf{S}_4^\xi)\\ &\times(\cos 2\theta\sin\varphi+\sin 2\theta\sin\varphi\cos\varphi),\end{aligned}$$

$$\begin{aligned} C_2=&-(\mathbf{S}_1^\xi+\mathbf{S}_2^\xi+\mathbf{S}_3^\xi+\mathbf{S}_4^\xi)\cos\varphi+(\mathbf{S}_1^\xi+\mathbf{S}_4^\xi-\mathbf{S}_2^\xi-\mathbf{S}_3^\xi)\\ &\times\sin\varphi+(\mathbf{S}_1^\eta+\mathbf{S}_2^\eta+\mathbf{S}_3^\eta+\mathbf{S}_4^\eta)\cos\theta\sin\varphi\\ &+(\mathbf{S}_1^\eta-\mathbf{S}_2^\eta-\mathbf{S}_3^\eta+\mathbf{S}_4^\eta)(\sin\theta+\cos\theta\cos\varphi),\end{aligned}$$

$$\begin{aligned} C_3=&-(\mathbf{S}_1^\eta+\mathbf{S}_2^\eta+\mathbf{S}_3^\eta+\mathbf{S}_4^\eta)\cos\theta\cos\varphi-(\mathbf{S}_1^\xi+\mathbf{S}_2^\xi+\mathbf{S}_3^\xi+\mathbf{S}_4^\xi)\\ &\times\cos 2\theta\sin\varphi+(\mathbf{S}_1^\eta-\mathbf{S}_2^\eta+\mathbf{S}_3^\eta-\mathbf{S}_4^\eta)\\ &\times(\sin\theta\cos 2\varphi-\cos\theta\cos\varphi)+(\mathbf{S}_1^\xi+\mathbf{S}_3^\xi-\mathbf{S}_2^\xi-\mathbf{S}_4^\xi)\\ &\times(\cos 2\theta\cos\varphi+\sin 2\theta\sin\varphi\cos\varphi).\end{aligned}$$

The choices $\theta=\pi/2$ and $\varphi=\pi/4$ give $C_1=-C_3=-\sqrt{2}(\mathbf{S}_2^\xi+\mathbf{S}_4^\xi)$ and $C_2=-\sqrt{2}(\mathbf{S}_2^\xi+\mathbf{S}_3^\xi)+\mathbf{S}_1^\eta-\mathbf{S}_2^\eta-\mathbf{S}_3^\eta+\mathbf{S}_4^\eta$. They correspond to the double-**k** state, which is stabilized if $D_1=D_3\equiv D$ and $D_2=0$. Expressions (6) and (7) are now completed by

$$\delta_{DM}P_k=\frac{D}{2J}(C_yC_z-C_xC_y),$$

$$\delta_{DM}Q_k=\frac{D}{2J}[C_xC_y-C_yC_z+i\sqrt{2}(C_yC_z+C_xC_y)],$$

and the excitation energy [Eq. (8)] becomes

$$\omega_3(k)=\sqrt{(1-C_xC_z+rC_yC_z-rC_xC_y)(1+C_yC_z+C_xC_y+C_xC_z)},$$

where the ratio $r=D/2J$ is constrained to obey $|r|\leqslant 1$. A careful examination of the resulting full spectrum shows that the set of excitation energies is an even function of r.

C. Symmetric anisotropic exchanges

There are other ways which have been proposed to stabilize the pyrite structure,[7] in particular the Hamiltonian

$$\delta H=\sum_{\langle i,j\rangle}\mathbf{S}_i\cdot\mathbf{A}_{ij}\cdot\mathbf{S}_j, \tag{16}$$

where the symmetric interaction matrix of the nearest-neighbor interaction is

$$\begin{aligned}\mathbf{A}_{ij}=&P_1s_{ij}\cdot s_{ij}+P_2t_{ij}\cdot t_{ij}+P_3u_{ij}\cdot u_{ij}+S_{12}(s_{ij}t_{ij}+t_{ij}s_{ij})\\ &+S_{23}(u_{ij}t_{ij}+t_{ij}u_{ij})+S_{31}(s_{ij}u_{ij}+u_{ij}s_{ij}).\end{aligned}$$

The isotropic nearest-neighbor coupling constant J can be defined as $J=P_1/3=P_2/3=P_3/3$. In fcc systems, P_1, P_2, and P_3 completely specify the nearest-neighbor coupling. From the linear terms of the spin-wave theory, we conclude that neither P_1+P_3 nor P_2 lifts the degeneracy of the ground state, whereas the extrema of the contribution given by P_1-P_3 are determined by

$$\begin{aligned}&\sin\varphi(\mathbf{S}_1^\xi+\mathbf{S}_2^\xi+\mathbf{S}_3^\xi+\mathbf{S}_4^\xi)-\cos\varphi(\mathbf{S}_1^\xi+\mathbf{S}_2^\xi-\mathbf{S}_3^\xi-\mathbf{S}_4^\xi)-\sin\theta(\mathbf{S}_1^\eta\\ &\quad-\mathbf{S}_2^\eta-\mathbf{S}_3^\eta+\mathbf{S}_4^\eta)+\cos\theta\cos\varphi(\mathbf{S}_1^\eta+\mathbf{S}_2^\eta+\mathbf{S}_3^\eta+\mathbf{S}_4^\eta)\\ &\quad+\cos\theta\sin\varphi(\mathbf{S}_1^\eta+\mathbf{S}_2^\eta-\mathbf{S}_3^\eta-\mathbf{S}_4^\eta)=0.\end{aligned}$$

Obviously this kind of contribution has nothing to do with the fcc type-I configuration.

Concerning the anisotropic symmetric interactions S_{ij}, the terms S_{31} and $S^{(-)}=(S_{12}-S_{23})/\sqrt{2}$ lead to conditions which also cannot be fulfilled by this configuration. The condition given by the contribution $S^{(+)}=(S_{12}+S_{23})/\sqrt{2}$ looks like

$$
\begin{aligned}
&(\sin\theta\cos 2\varphi-\cos\theta\sin\varphi)(\mathbf{S}_1^{\eta}-\mathbf{S}_2^{\eta}+\mathbf{S}_3^{\eta}-\mathbf{S}_4^{\eta})\\
&\quad-\cos\theta\cos\varphi(\mathbf{S}_1^{\eta}+\mathbf{S}_2^{\eta}-\mathbf{S}_3^{\eta}-\mathbf{S}_4^{\eta})+(\mathbf{S}_1^{\xi}-\mathbf{S}_2^{\xi}+\mathbf{S}_3^{\xi}-\mathbf{S}_4^{\xi})\\
&\quad\times[(\sin\varphi+\cos\varphi)\cos 2\theta+\sin 2\theta\sin\varphi\cos\varphi]=0,
\end{aligned}
$$

showing that this kind of interaction selects a double-**k** state with the choice $\cos\theta=0$ and $\sin\varphi=-\cos\varphi$. Expressions (6) and (7) now possess the adding terms

$$\delta_S P_k = r_S\left(1+\frac{C_xC_z}{2}\right),$$

$$\delta_S Q_k = \frac{r_S}{2}[-C_xC_z+i\sqrt{2}(C_yC_z+C_xC_y)],$$

where r_S is the ratio between the antiferromagnetic coupling constant and the strength of the stabilizing term. The explicit expressions of the corresponding magnons energies are rather messy, and we omit them here.

D. Mapping the DM interaction into an anisotropic exchange coupling

It was argued by Kaplan[19] and by Shekhtman, Entin-Wohlman, and Aharony[20] that, in the case of two sublattice magnetizations, the DM interaction can be eliminated by a gauge transformation of the spin variables. In fact the DM interaction is mapped into an anisotropic exchange coupling. This anisotropy is canceled by the superexchange anisotropy under some assumptions.[20] Indeed, as a concrete example, let us consider the following Hamiltonian which couples two spins with a DM interaction pointing in the z direction:

$$H_2 = J\sum_{\langle i,j\rangle}\mathbf{S}_{i1}\cdot\mathbf{S}_{j2}+D\sum_{\langle i,j\rangle}(\mathbf{S}_{i1}^{x}\mathbf{S}_{j2}^{y}-\mathbf{S}_{i1}^{y}\mathbf{S}_{j2}^{x}).$$

By a rotation about the z axis by an alternating angle,

$$\mathbf{S}_{i1}^{\pm}\to\mathbf{S}_{i1}^{\pm}\exp\left(\pm i\frac{\alpha}{2}\right),\quad \mathbf{S}_{j2}^{\mp}\to\mathbf{S}_{j2}^{\mp}\exp\left(\pm i\frac{\alpha}{2}\right),\tag{17}$$

where $\tan\alpha=D/J$, this Hamiltonian is transformed to

$$H_2=\frac{1}{2}\sum_{\langle i,j\rangle}|\mathcal{J}|(\mathbf{S}_{i1}^{+}\mathbf{S}_{j2}^{-}+\mathbf{S}_{i1}^{-}\mathbf{S}_{j2}^{+})+J\sum_{\langle i,j\rangle}\mathbf{S}_{i1}^{z}\mathbf{S}_{j2}^{z},$$

where $\mathbf{S}^{\pm}=\mathbf{S}^{x}\pm i\mathbf{S}^{y}$ and $\mathcal{J}=J+iD$.

The extension of this kind of result to three-dimensional frustrated systems is not possible in full generality. Indeed, when the DM interaction does not point in a definite direction, it is impossible to write this interaction under a form which does not display imaginary terms. However, an exception to this is the classical version of the DM interaction stabilizing the double-**k** structure, found in Sec. IV B above. Indeed, by taking Eqs. (14) and (15) with the conditions $D_1=D_3\equiv D$ and $D_2=0$, we obtain

$$
\begin{aligned}
H_{dm}=D\sum_{\langle i,j\rangle}&(\mathbf{S}_{i1}^{x}\mathbf{S}_{j2}^{y}-\mathbf{S}_{i1}^{y}\mathbf{S}_{j2}^{x}+\mathbf{S}_{i1}^{x}\mathbf{S}_{j4}^{y}-\mathbf{S}_{i1}^{y}\mathbf{S}_{j4}^{x}\\
&-\mathbf{S}_{i2}^{x}\mathbf{S}_{j3}^{y}+\mathbf{S}_{i2}^{y}\mathbf{S}_{j3}^{x}+\mathbf{S}_{i3}^{x}\mathbf{S}_{j4}^{y}-\mathbf{S}_{i4}^{y}\mathbf{S}_{j3}^{x}).
\end{aligned}
$$

Then the mappings $\mathbf{S}_{i3}^{\pm}\to\mathbf{S}_{i3}^{\pm}\exp[\pm i(\alpha/2)]$ and $\mathbf{S}_{j4}^{\mp}\to\mathbf{S}_{j4}^{\mp}\exp[\pm i(\alpha/2)]$, together with Eq. (17), transform the Hamiltonian $H_{nn}+H_{dm}$ to

$$
\begin{aligned}
H_{nn}+H_{dm}=\frac{J}{2}\sum_{\langle i,j\rangle}&(\mathbf{S}_{i1}^{+}\mathbf{S}_{j3}^{-}+\mathbf{S}_{i1}^{-}\mathbf{S}_{j3}^{+}+\mathbf{S}_{i2}^{+}\mathbf{S}_{j4}^{-}+\mathbf{S}_{i2}^{-}\mathbf{S}_{j4}^{+})\\
&+\frac{|\mathcal{J}|}{2}\sum_{\langle i,j\rangle}(\mathbf{S}_{i1}^{+}\mathbf{S}_{j2}^{-}+\mathbf{S}_{i1}^{-}\mathbf{S}_{j2}^{+}+\mathbf{S}_{i1}^{+}\mathbf{S}_{j4}^{-}+\mathbf{S}_{i1}^{-}\mathbf{S}_{j4}^{+}\\
&+\mathbf{S}_{i2}^{+}\mathbf{S}_{j3}^{-}+\mathbf{S}_{i2}^{-}\mathbf{S}_{j3}^{+}+\mathbf{S}_{i3}^{+}\mathbf{S}_{j4}^{-}+\mathbf{S}_{i3}^{-}\mathbf{S}_{j4}^{+}).
\end{aligned}
$$

This anisotropy is different from the symmetric anisotropy interaction studied Sec. IV C, which also leads to a double-**k** state.

V. FOUR-SPIN EXCHANGE INTERACTIONS AND BIQUADRATIC COUPLINGS

Effects of the fourth-order interactions among localized spins have been considered in a fcc single-band Hubbard model near the insulator limit.[21] This kind of interaction was shown to lift the degeneracy of the structure with equivalent wave numbers to make them noncollinear.

The fourth-order perturbation theory gives rise to two- and four-spin interactions and also biquadratic and three-spin interactions for S larger than one-half. The four-spin interaction has the form

$$
\begin{aligned}
\delta\mathcal{H}_{IV}^{(\mp)}=K[&(\mathbf{S}_1\cdot\mathbf{S}_2)(\mathbf{S}_3\cdot\mathbf{S}_4)+(\mathbf{S}_1\cdot\mathbf{S}_4)(\mathbf{S}_2\cdot\mathbf{S}_3)\\
&\mp(\mathbf{S}_1\cdot\mathbf{S}_3)(\mathbf{S}_2\cdot\mathbf{S}_4)],
\end{aligned}\tag{18}
$$

and gives the conditions

$$
\begin{aligned}
&\sin 2\theta[-1+2\sin^2\theta(\sin^4\varphi+\cos^4\varphi)\\
&\quad\pm\cos 2\theta](\mathbf{S}_1^{\xi}-\mathbf{S}_2^{\xi}+\mathbf{S}_3^{\xi}-\mathbf{S}_4^{\xi})=0\\
&\sin^2\theta\sin 4\varphi(\mathbf{S}_1^{\eta}-\mathbf{S}_2^{\eta}+\mathbf{S}_3^{\eta}-\mathbf{S}_4^{\eta})=0.
\end{aligned}
$$

The solutions of this system can be divided into four classes: (1) $\sin\theta=0$, (2) $\cos\theta=0$ and $\sin^2\varphi=\cos^2\varphi$, (3) $\sin^2\varphi=\cos^2\varphi$ and $\cos^2\theta+\cos 2\theta=0$, and (4) $\sin^2\varphi=\cos^2\varphi$ and $-\cos^2\theta+\cos 2\theta=0$. The two first solutions are common to $\delta\mathcal{H}_{IV}^{(+)}$ and $\delta\mathcal{H}_{IV}^{(-)}$. They correspond to the single-**k** and double-**k** states, respectively. The third and fourth possibilities are provided by $\delta\mathcal{H}_{IV}^{(+)}$ and $\delta\mathcal{H}_{IV}^{(-)}$, respectively. The third case corresponds to a triple-**k** state. The final configuration is again a single-**k** state. Therefore, $\delta\mathcal{H}_{IV}^{(+)}$ stabilizes the three fcc states potentially, whereas $\delta\mathcal{H}_{IV}^{(-)}$ cannot put the triple-**k** state into equilibrium. The classical energies given by $\delta\mathcal{H}_{IV}^{(+)}$ are $K/2$, K, and $K/3$ for the single-, double-, and triple-**k** states, respectively. Therefore, a positive coupling constant K favors a triple-**k** structure, whereas a negative K stabilizes a double-**k** state. The other Hamiltonian $\delta\mathcal{H}_{IV}^{(-)}$ gives K (single-**k**) and $-K$ (double-**k**) as classical energies, thus putting the canonical antiferromagnetic state into equilibrium with a negative coupling and the double-**k** state into equilibrium with a positive coupling.

The spin-wave energies are easily obtained from the expressions

$$P_k = 1 - \frac{4KS^2}{9J} + \left(1 - \frac{2KS^2}{3J}\right) \times (\mathcal{B}_{yz} C_y C_z + \mathcal{B}_{xy} C_x C_y - \mathcal{B}_{xz} C_x C_z), \tag{19}$$

$$Q_k = \left(1 - \frac{2KS^2}{3J}\right)(\mathcal{A}_{yz} C_y C_z + \mathcal{A}_{xy} C_x C_y + \mathcal{A}_{xz} C_x C_z). \tag{20}$$

Introducing the parameter

$$\mathcal{B} = \frac{\left(1 - \dfrac{2KS^2}{3J}\right)}{\left(1 - \dfrac{4KS^2}{9J}\right)},$$

we obtain

$$\begin{aligned} \omega_1(k) = \{ & \gamma^2[(1-\mathcal{B}C_x C_z)^2 - \mathcal{B}^2 C_y^2 (C_x - C_z)^2] \\ & + \alpha^2[(1-\mathcal{B}C_x C_y)^2 - \mathcal{B}^2 C_z^2 (C_x - C_y)^2] \\ & + \beta^2[(1-\mathcal{B}C_y C_z)^2 - \mathcal{B}^2 C_x^2 (C_y - C_z)^2]\}^{1/2}. \end{aligned} \tag{21}$$

The three other energies are obtained as explained above. From this spectrum we conclude that, in order to ensure that the argument in the square root is always positive in the whole Brillouin zone, the parameter $\mathcal{B}$ has to obey to $0 \leqslant \mathcal{B} \leqslant 1$. It is easy to verify that this condition is satisfied by the ratio K/J if $0 \leqslant K/J \leqslant 3/2S^2$.

The suppression of the linear terms given by the biquadratic term

$$\delta\mathcal{H}_{biq} = j \sum_{\langle i,j \rangle} (\mathbf{S}_i \cdot \mathbf{S}_j)^2$$

requires the same conditions as those corresponding to the four-spin interaction $\delta\mathcal{H}_{IV}^{(+)}$. However the classical energies are now $6j$, $2j$, and $2j/3$ for the single-, double-, and triple-**k** states, respectively. Again a positive constant j favors the triple-**k** structure, whereas the single-**k** structure is stabilized by a negative contribution. The dispersion energies are given by the same expressions as those given for the four-spin interaction, with a trivial change of coupling:

$$\frac{4K}{9J} \Rightarrow \frac{2j}{3J}.$$

The antiferromagnetic substance MnO is known to have a critical temperature ($T_N = 116$ K), a Curie temperature ($\theta = -610$ K), a transverse susceptibility, and a temperature dependence on the sublattice magnetization consistent with the idea of biquadratic exchange.[22] The compound $\mathrm{MnTe_2}$ shows similar behaviors ($T_N = 86$ K and $\theta = -528$ K). Moreover, in both compounds, the variation of the magnetic moment as a function of the temperature is sharper than the Brillouin function $B_{5/2}$ expected in a mean-field approximation. Thus it is plausible that the magnetic structure corresponding to $\mathrm{MnTe_2}$ is also stabilized by the biquadratic term.

VI. EFFECT OF THE LOCAL ANISOTROPY

In this section we consider the case of local single-ion anisotropy, and use a Hamiltonian[15] which has to be added to $\mathcal{H}_2$,

$$\delta\mathcal{H}_{an} = D_{an} \sum_{i=1}^{4} (\mathbf{S}_i \cdot \mathbf{d}_i)^2, \tag{22}$$

where D_{an} is the anisotropy coupling. The resulting equilibrium conditions given by the linear terms do not depend on the particular sublattice $l = 1, \ldots, 4$ considered. They read

$$\frac{D_{an}}{\sqrt{3}} a (b\mathbf{S}_l^{\xi} + c\mathbf{S}_l^{\eta}) = 0,$$

where

$$a = (\sin\varphi + \cos\varphi)\sin\theta + \cos\theta,$$

$$b = (\sin\varphi + \cos\varphi)\cos\theta - \sin\theta, \quad c = \cos\varphi - \sin\varphi.$$

Thus the stable structure is the one that fulfills $a = 0$ (I) or $b = c = 0$ (II). The first condition is satisfied by a double-**k** structure with $\sin\varphi = -\cos\varphi$ and $\cos\theta = 0$. The second condition is fulfilled with the choices $\sin\varphi = \cos\varphi$ and $\tan 2\theta = -2\sqrt{2}$, and corresponds to a triple-**k** structure. The classical energy given by such a contribution is $\delta\mathcal{E}_{an} = 4D_{an}S^4$ whereas $\delta\mathcal{E}_{an} = 8D_{an}S^4/3$ in the double-**k** case. Thus the triple-**k** state is stabilized by the anisotropy energy with a negative coupling.

Accordingly, expression (3) is modified as

$$\begin{aligned} \omega_1(k) = \Bigg(& \gamma^2\left[\left(1 + \frac{\mathcal{A}}{2} - C_x C_z\right)^2 - C_y^2 (C_x - C_z)^2\right] \\ & + \alpha^2\left[\left(1 + \frac{\mathcal{A}}{2} - C_x C_y\right)^2 - C_z^2 (C_x - C_y)^2\right] \\ & + \beta^2\left[\left(1 + \frac{\mathcal{A}}{2} - C_y C_z\right)^2 - C_x^2 (C_y - C_z)^2\right]\Bigg)^{1/2} \end{aligned} \tag{23}$$

where $\mathcal{A} = -D_{an}S^2/J$ (triple-**k** case) or $-2D_{an}S^2/3J$ (double-**k** case). The four branches of the spectrum are gapped antiferromagnetic modes with a common gap Δ given by

$$\Delta = \mathcal{A}\left(\frac{\mathcal{A}}{4} + 1\right).$$

In order that the spectrum deduced from Eq. (23) be well defined in the whole Brillouin zone, it is necessary that the coupling has to remain positive. Interestingly enough, this constraint discards the possibility of putting a triple-**k** state into equilibrium.

Expression (22) is not the only possible form of the anisotropy energy; it might in general also include terms with spin projections on vectors not necessarily identical to the spin directions. For instance, we can consider a cyclic permutation of the indices

$$\delta\mathcal{H}_{an}^{(1)}=D_{an}[(\mathbf{S}_1\cdot\mathbf{d}_2)^2+(\mathbf{S}_2\cdot\mathbf{d}_3)^2+(\mathbf{S}_3\cdot\mathbf{d}_4)^2+(\mathbf{S}_4\cdot\mathbf{d}_1)^2].$$

However, let us point out that this kind of contribution is discarded by the symmetry criterion formulated by the $\mathcal{C}$-parity operator (see Sec. II).

VII. MAGNETIC MOMENT REDUCTION IN THE GROUND STATE

The interpretation of a magnetic moment value measured on an antiferromagnetic compound is faced with the task of separating covalency reduction from zero-point quantum fluctuation effects. According to the spin-wave analysis developed here, the quantum reduction $\langle\Delta S\rangle$ in the ground state can be formulated as

$$\langle\Delta S\rangle=\langle\Delta S\rangle_1=\langle\Delta S\rangle_2=\langle\Delta S\rangle_3=\langle\Delta S\rangle_4$$
$$=\frac{1}{8N}\sum_{l=1,4}\sum_{k}\left(\frac{P_k^l}{\sqrt{(P_k^l)^2-|Q_k^l|^2}}-1\right),$$

where

$$\langle\Delta S\rangle_l=\frac{1}{2N}\sum_{k}\left(\frac{P_k^l}{\sqrt{(P_k^l)^2-|Q_k^l|^2}}-1\right). \tag{24}$$

These expressions are independent of both the spin S and the antiferromagnetic coupling J, and give identical results as discussed above. Replacing the sum above by a Watson-like integral, one obtains a deviation of magnetic moments from their nominal value S at $T=0$. Assuming $\mathcal{V}=a^3$, where $\mathcal{V}$ is the volume of the portion of crystal used in defining the boundary conditions, we have found $\langle\Delta S\rangle_{II}=1.016\mu_B$ and $\langle\Delta S\rangle_{III}=0.794\mu_B$ for the double- and triple-**k** configurations, respectively. Such large spin reductions are expected from frustration effects.

Now we discuss the eventual modifications coming from the various types of anisotropy and interactions likely to stabilize these configurations. First we present an analytical argument predicting the effect of these interactions following the sign of the ratio of the antiferromagnetic coupling J and the strength C_S of the stabilizing term. Indeed, it follows from Eqs. (19) and (20) that the parameters defining the Bogoliubov transformation diagonalizing $\mathcal{H}_{nn}+\mathcal{H}_S$ ($\mathcal{H}_S$ being a biquadratic, four-spin, or anisotropic contribution) can be expressed in full generality as $P_k^S=\Lambda_1+\Lambda_0 P_k$ and $Q_k^S=\Lambda_0 Q_k$, where we recall that P_k and Q_k are the parameters which diagonalize $\mathcal{H}_{nn}$. Let us assume that the coupling constant of the stabilizing contribution $\mathcal{H}_S$ is small compared to J. Consequently we can take the constant Λ_1 as a perturbative parameter and expand Eq. (24) to first order. This results in (keeping one branch of the spectrum)

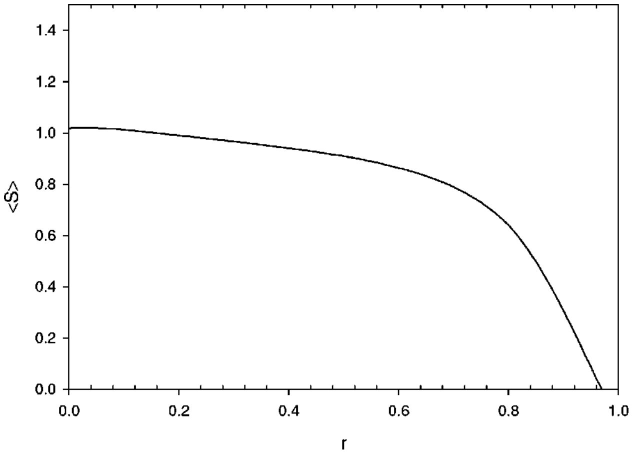

FIG. 2. Calculated spin-reduction curve given by a DM term for the double-**k** state.

$$\langle\Delta S\rangle\simeq\frac{\mathcal{V}}{2(2\pi)^3}\int_{BZ}\left[\frac{P_k}{\sqrt{P_k^2-|Q_k|^2}}\right.$$
$$\left.\times\left(1-\frac{\Lambda_1}{\Lambda_0}\frac{|Q_k|^2}{P_k(P_k^2-|Q_k|^2)}\right)-1\right]. \tag{25}$$

Since we know from Eq. (11) that the integral is definite positive, a careful examination of the signs of the constants Λ_i leads to the conclusion that the stabilizing contribution increases (decreases) the spin reduction $\langle\Delta S\rangle$ with a negative (positive) C_S. In fact, as we now discuss, a numerical study extends this conclusion to every value of the coupling ratio.

In Figs. 2–4 we plot some spin reductions given by different stabilizing terms as a function of the ratio $r=C_S/J$. The curve given by the local anisotropic term for the triple-**k** state is nearly identical to the result shown for the double-**k** state. These plots reveal very pertinent general trends of all stabilizing terms of noncollinear states. Indeed, as expected, $\langle\Delta S\rangle$ shrinks for positive r as this ratio begans to increase. Moreover, with the exception of the DM term, the spin reduction continuously decreases up to the regime of an ex-

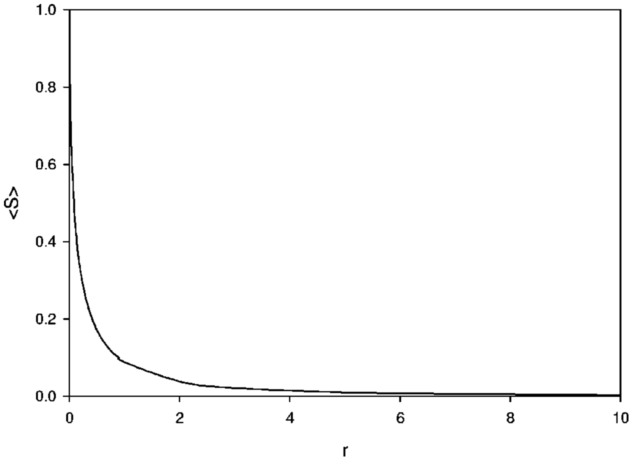

FIG. 3. Calculated spin-reduction curve given by the local anisotropic term for the double-**k** state.

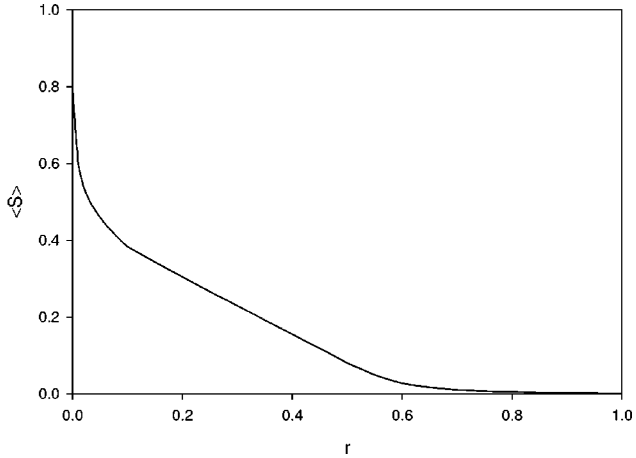

FIG. 4. Calculated spin-reduction curve given by the biquadratic term for the triple-**k** state.

tremely strong stabilizing term. This suggests that this quantity vanishes smoothly in the $1/r \Rightarrow 0$ limit, rather than maintaining a limiting value or becoming negative. Also we have observed that, at fixed r, the spin reduction sizes given by the biquadratic and anisotropic terms are nearly the same, when the triple-**k** configuration is considered. Finally, let us emphasize that for the anisotropic contribution the ratio r has to remain positive for both noncollinear states, whereas, with regard to the biquadratic term, a positive r is again required by the stabilization of the triple-**k** state. Thus we have shown in full generality that, in this context, the existence of this state results in a sizable diminution of the spin reduction, at mean. Now we briefly discuss the DM term. Let us remember that it only can stabilize the double-**k** state, that $|r| \leqslant 1$, and that $\langle \Delta S(r) \rangle = \langle \Delta S(-r) \rangle$ Thus, again in this case a shrinking of the spin reduction induced by the stabilizing term is predicted. Obviously these conclusions are not true if the spin Hamiltonian contains supplementary terms.

The triple-**k** type-I structure is common to many compounds which sometimes display similar properties. In particular USb, NpSb, and NpBi have very similar Néel temperatures (212, 199, and 192.5 K, respectively; see, for instance Ref. 23) and ordered magnetic moments of the Np^{3+} ions ($2.8\mu_B$, $2.5 \pm 0.05\mu_B$, and $2.63\mu_B$, respectively). On the other hand, it is now well experimentally established[10] that the triple-**k** magnetic structure of the MnTe_2 compound is stable down to 0 K with a large moment reduction of $0.72\mu_B$. In such fcc compounds, the unit cell on which the boundary conditions are enforced is one-eight of the nuclear fcc lattice on which the Brillouin zone is defined. Thus, in that case, the magnetic moment reduction predicted by the canonical Heisenberg exchange Hamiltonian [Eq. (1)] is $0.2\mu_B$ when a triple-**k** structure is assumed. For Mn^{2+} ions the measure is roughly larger than three times this prediction. Let us remember that the effect of a stabilizing term would be to worsen this large discrepancy. For the other compounds the comparison is difficult since the spins of the uranium and neptunium are not well known.

VIII. STABILIZING EXCHANGES AND SPIN DYNAMICS

A. Theoretical predictions on the spectrum

In general a magnetic excitation spectrum consisting of branches with longitudinal and transverse polarizations is a characteristic of noncollinear systems.[14] Indeed, in a collinear structure all the excitations are transverse. Below we give both kinds of spin-wave dispersions along various directions of high symmetry in the Brillouin zone. Our main interest is twofold. First, from the results given by the simplest Hamiltonian [Eq. (1)], we extract the essential theoretical predictions, then, investigating the effects of stabilizing terms, we show that these predictions remain essentially unaffected. Second, these predictions are compared following the type of noncollinear structure (double **k** or triple **k**) considered.

The spin-wave energies [Eqs. (3) and (4)] have the following expressions along the symmetry direction $(0,0,\zeta)$:

$$\omega_1(0,0,q) = \omega_4(0,0,q) = 0,$$

$$\omega_2(0,0,q) = \omega_3(0,0,q) = 2\alpha\sqrt{1-q^2}, \tag{26}$$

where $q = \cos(ka/2)$, whereas for the direction $(\zeta,\zeta,0)$ we obtain [now denoting $\cos(ka/2\sqrt{2})$ by q]

$$\begin{aligned}\omega_1(q,q,0) &= \omega_4(-q,-q,0)\\ &= \sqrt{(1-q)(1-q^2)(1-q+2q\alpha^2)},\\ \omega_2(q,q,0) &= \omega_3(-q,-q,0)\\ &\quad\times\sqrt{(1-q^2)[1-2q(\beta^2-\gamma^2)+q^2(1-2\alpha^2)]}.\end{aligned} \tag{27}$$

Finally along (ζ,ζ,ζ) the spin-wave energies become

$$\omega_1(q,q,q) = (1-q^2),$$

$$\omega_2(q,q,q) = \sqrt{(1-q^2)(1+3q^2-4q^2\beta^2)}$$

where now $q = \cos(ka/2\sqrt{3})$. The two remaining components of the spin-wave spectra $\omega_3(q,q,q)$ and $\omega_4(q,q,q)$ are deduced from $\omega_2(q,q,q)$ by replacing $\beta \rightarrow \gamma$ and $\beta \rightarrow \alpha$, respectively. These expressions display the following remarkable features.

(i) For wave vectors along the ΓX direction $(0,0,\zeta)$ the ω_2 and ω_3 modes are different from zero for the double- and triple-**k** states, and disappear in the collinear state. These modes are thus identified as the longitudinal branches missing in the single-**k** structure. This observation is corroborated by the spin-wave spectrum corresponding to this structure [i.e., Eq. (3)] since $\omega_a \equiv \omega_1$ and ω_b can be identified with ω_4.

(ii) The transverse and longitudinal contributions are independently $q \rightarrow -q$ invariant. As we shall discuss next, this property infers very important phenomenological consequences.

(iii) The longitudinal and transverse modes are degenerate at the point Γ in both the $(0,0,\zeta)$ and the $(\zeta,\zeta,0)$ directions with a zero value, and at the two points L [$\mathbf{k} = \sqrt{2}(\pi/a,\pi/a,0)$] and [$\mathbf{k} = \sqrt{3}(\pi/a,\pi/a,\pi/a)$], where their common value is 1. These results do not depend upon the specific noncollinear state chosen.

(iv) At the points Γ and X the longitudinal modes reach their minima or maxima. It is also worthwhile to underline that these modes are alone to survive in the ΓX direction $(0,0,\zeta)$.

(v) The dispersions of the longitudinal modes [Eqs. (26)] around $k=0$ are $\omega_2 \equiv \omega_3 = (\alpha k/\sqrt{2}) + o(k^3)$, whereas for the same value in the direction $(\zeta,\zeta,0)$ from Eq. (27) we obtain $\omega_2 = (\gamma k/\sqrt{2}) + o(k^3)$ and $\omega_3 = (\beta k/\sqrt{2}) + o(k^3)$ when the points in this direction are plotted in $2\sqrt{2}\pi/a$ units. As a consequence we see that these dispersions are isotropic near the Γ point when $\alpha=\beta=\gamma$, i.e., for the triple-**k** state. This difference of behavior between the double- and triple-**k** states is a way to distinguish them in compounds where they would be both potentially present. This is a consequence of the fact that, unlike the two other structures, the triple-**k** structure does not violate cubic symmetry.

When the model is completed by a local anisotropy interaction $(\mathcal{A})$ or biquadratic terms $(\mathcal{B})$, using Eqs. (23) and (21) the expressions of the spin-wave energies are found as follows: (i) In the direction $(0,0,\zeta)$,

$$\omega_1(0,0,q) = \omega_4(0,0,-q) = \sqrt{\left(\frac{\mathcal{A}}{2}\right)^2 + \mathcal{A}(1-\alpha^2)(1-q)},$$

$$\omega_2(0,0,q) = \omega_3(0,0,-q) = \sqrt{4\alpha^2(1-q^2) + \left(\frac{\mathcal{A}}{2}\right)^2 + \mathcal{A}(1+\alpha^2) + \mathcal{A}q(\gamma^2-\beta^2)},$$

$$\omega_1(0,0,q) = \omega_4(0,0,-q) = \sqrt{1-\mathcal{B}}\sqrt{1+\mathcal{B}-2q\mathcal{B}-2\alpha^2\mathcal{B}(1-q)},$$

$$\omega_2(0,0,q) = \omega_3(0,0,-q) = \sqrt{\alpha^2(1+\mathcal{B}-2q\mathcal{B})(1+\mathcal{B}+2q\mathcal{B}) + (1-\mathcal{B})[\beta^2(1+\mathcal{B}+2q\mathcal{B}) + \gamma^2(1+\mathcal{B}-2q\mathcal{B})]};$$

(ii) in the direction $(\zeta,\zeta,0)$,

$$\omega_1(q,q,0) = \omega_4(-q,-q,0) = \sqrt{(1-q)(1-q^2)(1-q+2q\alpha^2) + (\mathcal{A}/2)^2 + \mathcal{A}(1-q)(1+\alpha^2 q)},$$

$$\omega_2(q,q,0) = \omega_3(-q,-q,0) = \sqrt{(1-q^2)[1-2q(\beta^2-\gamma^2)+q^2(1-2\alpha^2)] + (\mathcal{A}/2)^2 + \mathcal{A}[1+\alpha^2 q^2 - q(\beta^2-\gamma^2)]},$$

$$\omega_1(q,q,0) = \omega_4(-q,-q,0) = \sqrt{1-\mathcal{B}q^2}\sqrt{1+\mathcal{B}q^2-2q\mathcal{B}+2\alpha^2\mathcal{B}q(1-q)},$$

$$\begin{aligned}\omega_2(q,q,0) &= \omega_3(-q,-q,0)\\ &= \{\alpha^2(1+\mathcal{B}q^2-2q\mathcal{B})(1+\mathcal{B}q^2+2q\mathcal{B}) + (1-\mathcal{B}q^2)[\beta^2(1+\mathcal{B}q^2-2q\mathcal{B}) + \gamma^2(1+\mathcal{B}q^2+2q\mathcal{B})]\}^{1/2};\end{aligned}$$

and (iii) in the direction (ζ,ζ,ζ),

$$\omega_1(q,q,q) = 1 + \frac{\mathcal{A}}{2} - q^2,$$

$$\omega_2(q,q,q) = \sqrt{(1-q^2)(1+3q^2-4q^2\beta^2) + (\mathcal{A}/2)^2 + \mathcal{A}(1+q^2-2q^2\beta^2)},$$

$$\omega_1(q,q,q) = 1 - \mathcal{B}q^2,$$

$$\omega_2(q,q,q) = \sqrt{(1-\mathcal{B}q^2)(1+3\mathcal{B}q^2-4\mathcal{B}q^2\beta^2)}.$$

The two other components of the spin-wave spectra $\omega_3(q,q,q)$ and $\omega_4(q,q,q)$ are obtained from $\omega_2(q,q,q)$ by replacing $\beta \rightarrow \gamma$ and $\beta \rightarrow \alpha$, respectively. From these expressions it is easy to state that the stabilizing terms added to Hamiltonian (1) break the degeneracy between longitudinal and transverse modes at the points X and Γ. However, these modes remain separately equal at these points.

B. Comparison with observed magnetic excitations

These parametrizations are now compared with some experimental spectra from compounds where the dispersion curves and the intensities of the phonons are well known, thus insuring a precise identification of the magnetic scattering. We first discuss the results for USb.[1,24] In fact there is a qualitative feature of the data which the conventional spin-wave theory presented here is not able to capture. This is the striking fact that the energy of the longitudinal mode observed at the Γ point is about four times higher than at the X point. Spin-wave theory predicts that the two frequencies of the longitudinal modes at these points should be equal. It is worthwhile to emphasize that this strong disagreement with the observed dispersions is actually a consequence of the symmetry of the unit cell of this kind of crystal. Therefore, it should survive any peculiar parametrization and all higher-order corrections. Moreover, the theory presented here should produce a splitting of the spin wave into different modes along (ζ,ζ,ζ) which is not experimentally observed. Note that, whatever the model used or the noncollinear state considered, the longitudinal and transverse modes remain degenerate at the L points in both the $(\zeta,\zeta,0)$ and the (ζ,ζ,ζ) directions, in agreement with the spin-wave data on USb.

Jensen and Bak[14] calculated the excitation spectrum of USb by using a Hamiltonian including, in addition to the antiferromagnetic exchange coupling between nearest neighbors, the pseudodipolar interaction [Eq. (16)] with the parameters $P_1=P_3$ and $P_2=S_{12}=S_{23}=S_{31}=0$, and a crystal-field term with cubic symmetry. Written in the operator framework introduced by Stevens,[25] this last term is classically $H_{cr}\sim S_x^4+S_y^4+S_z^4$. In spite of some criticisms,[1] this model reproduces quite well the excitations observed in the compound USb with crystal-field parameters, in rough agreement with experiment and with a very large anisotropic coupling, twice as larger as the antiferromagnetic coupling. However, as explained in Sec. VII, the chosen pseudodipolar interaction is unable to stabilize a noncollinear state. Moreover, performing a spin-wave calculation of H_{cr}, it is easy to convince oneself that the resulting expression does not contain linear terms. Thus the spin Hamiltonian used by Jensen and Bak cannot be selected by our analysis. Finally, these authors were able to reproduce the excitation spectrum of USb because their formulation contained only one branch of that spectrum, thus breaking the $q\rightarrow -q$ invariance and consequently the degeneracy of the longitudinal modes at the points X and Γ.

NpBi is a compound which orders antiferromagnetically below $T=192.5$ K in a triple-**k** type-I structure.[23] At $T=10$ K, dispersions along $(0,0,\zeta)$ of the longitudinal and transverse modes were measured[2] by inelastic neutron-scattering experiments. They are completely different from that of USb, since the energy of the transverse mode observed at Γ is about $\frac{3}{2}$ times higher than that of the longitudinal mode at the same ordering wave vector. These two branches remain distinct in the range of ζ between 0 and approximately 0.4. The measurements along the $(\zeta,\zeta,0)$ direction exhibit roughly the same behavior, with a degeneracy of the two modes beginning at $\zeta\sim 0.3$.

The observed marked difference between the dispersions of the two compounds can be predicted by the spin-wave theory if the multi-**k** antiferromagnetic structure of NpBi at low temperature is assumed to be a double-**k** structure. Indeed, first consider the effect of a local anisotropy term when the state $\alpha=0$ and $\beta^2=\gamma^2=\frac{1}{2}$ is selected. The longitudinal (L) and transverse modes (T) are then degenerate at the point $\mathbf{k}=(0,0,\pi/a)$ since $E_L=E_T=\sqrt{\mathcal{A}^2+4\mathcal{A}}$ and at the point $\mathbf{k}=\sqrt{2}(\pi/a,\pi/a,0)$ where $E_L=E_T=\mathcal{A}+2$. Moreover the model predicts the same value of the energies of the transverse mode at $\mathbf{k}=(0,0,\pi/a)$ and at $\mathbf{k}=(0,0,0)$, a fact which is, on the whole, experimentally verified. The most serious drawback of this parametrization, prohibiting us from doing a more refined analysis, is the impossibility to explain the value of the longitudinal energy at $\mathbf{k}=(0,0,2\pi/a)$ which is predicted the same than those at $\mathbf{k}=(0,0,0)$, in clear contradiction with the data. The biquadratic contribution gives very similar results

C. A peculiar case: The γ-Mn alloys

The multi-**k** magnetic ordering is also encountered frequently in the γ-Mn alloys together with some lattice distortion. Whereas moderately doped alloys become fct with $c<a$, the more heavily doped compounds remain cubic with $c=a$. For a given impurity range $17\%<x<22\%$, MnNi alloys become fct at low temperature with $c>a$. These three crystal structures may be identified with single-, triple-, and double-**k** magnetic phases, respectively (for a recent work on this subject, see Ref. 26). It is quite simple to understand in the present framework why the magnetic phases which violate cubic symmetry cannot be in the triple-**k** state. Indeed, the breaking of the cubic symmetry induces a stronger coupling of bonds (13) and (24), resulting in a small anisotropy of nearest neighbor Heisenberg Hamiltonian [Eq. (1)],

$$H_{nn}=J\sum_{\langle i,j\rangle}(\mathbf{S}_{i1}\cdot\mathbf{S}_{j2}+\mathbf{S}_{i1}\cdot\mathbf{S}_{j4}+\mathbf{S}_{i2}\cdot\mathbf{S}_{j3}+\mathbf{S}_{i3}\cdot\mathbf{S}_{j4})$$

$$+J'\sum_{\langle i,j\rangle}(\mathbf{S}_{i1}\cdot\mathbf{S}_{j3}+\mathbf{S}_{i2}\cdot\mathbf{S}_{j4})$$

where the constant coupling J' is larger than J. It is easy to see that the condition to eliminate the linear terms of the linear spin wave formulation is

$$\sin 2\theta=0.$$

The degeneracy is only partially lifted. It remains to add a term to stabilize the angle φ. For instance, the anisotropy energy considered in Sec. VIII would select the double-**k** state.

IX. CONCLUSION

We have investigated the noncollinear magnetic structures of type I on a fcc lattice, trying to learn how the continuous degeneracy, which is present in the classical theory, is removed. We selected according to the kind of contribution the most stable state. Summarizing our results, the four spin interactions $\delta\mathcal{H}_{IV}^{(+)}$ and the biquadratic term put the triple-**k** state into equilibrium when the sign of their coupling strength is positive. Since the spin reduction is diminished by the stabilizing term, we arrive at the general result that, in this case, the stabilizing procedure increases the value of the magnetic moment. The double-**k** state is selected by $\delta\mathcal{H}_{IV}^{(+)}$ and $\delta\mathcal{H}_{IV}^{(-)}$, and we have local single-ion anisotropies with negative and positive couplings, respectively. When the structure of the crystal is pyritelike, the double-**k** antiferromagnetic order is also given by a DM term or a symmetric anisotropic exchange.

Concerning the spin-wave spectra, the two results depend neither on the particular choice of the phenomenological Hamiltonian describing the spin state nor on the peculiar parametrization used to describe the data. They are rather a consequence of the overall symmetry of the framework in which the spin-wave theory is formulated. First, the large difference observed at low temperature between the longitudinal and transverse energies $(E_T/E_L{\sim}5)$, at the X point in USb, is in strong disagreement with the prediction of the spin-wave calculation. One might also emphasize a common feature of spin-wave data which is of great phenomenological importance. This is the fact that the energies of spin waves almost reach their maximum values at the points Γ, in strong contradiction with the Heisenberg antiferromagnetic Hamiltonian [Eq. (1)] which predicts zero values. Thus the data on the energies at these points would settle the order of magnitude of the stabilizing terms. If this is true, they could not be considered perturbative terms, since their coupling would be at least of the same order as the strength J of the assumed leading contribution. This is in contrast to the majority of spontaneous magnetized materials displaying canted spin arrangements, where a weak perturbation superimposed on the dominant antiferromagnetic interaction produces weak ferromagnetism.

However, this straightforward comparison between theoretical spin-wave spectra and results of neutron-scattering experiments is perhaps misleading and has to be considered, at best, as an indicator. Indeed, when several sublattices enter the game one has to perform an exact calculation of the dynamical correlation functions which are observed experimentally. It would be an all-important progress to know to what extent the discrepancies we have just pointed out would be deleted in a more rigorous treatment.

In fact, although it is obvious that no clear signal emerges from the data, we believe the way is now open to very refined comparisons. The magnetic moment reduction of the manganese measured in the compound MnTe_2 is in clear contradiction with the simplest model proposed here. On the other hand, in the theoretical explanation of the compound Usb the crystal-field effect seems to play an important role. These two examples show the necessity of taking more terms in the spin Hamiltonian into account than the minimal model built from the nearest-neighbor contribution and a stabilizing term. Moreover, these supplementary terms are eventually apt to remove the strong obstacle raised by the sign of the stabilizing contribution, which leads to a diminishing of the spin reduction given by the basic Hamiltonian when the triple-**k** states are considered. This fact is in contradiction with data (at least for the compound MnTe_2). An additive contribution breaking the energy hierarchy between the double- and the triple-**k** states would be welcome.

ACKNOWLEDGMENTS

This work was supported by the ESF ''Vortex'' Program and the CEA (Accord-Cadre N° 12M). I am particularly indebted to H. Kachkachi for generously introducing me to the subject. I am also very glad to acknowledge F. Biet for his participation at the early stage of this work. I have also benefitted from patient explanations and pertinent suggestions from A. Vediayev. I would like to thank A. I. Buzdin, D. Foerster, and C. Lacroix for valuable discussions. Finally it is a great pleasure to thank J. P. Sanchez for introducing me to the experimental aspect of this subject.

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