Spin waves in several Heisenberg systems: Three-sublattice with different exchange constants ($J_{ab}=J_{bc}\neq J_{ca}$) and a superlattice with the elementary unit of four or three different layers

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(Received 5 February 1996; revised manuscript received 3 February 1997)

The quadratic Holstein-Primakoff spin-wave Hamiltonian for three-sublattice Heisenberg systems with different exchange constants ($J_{ab}=J_{bc}\neq J_{ca}$) was diagonalized by a three-step procedure. The classical ground state was examined and the effects of quantum fluctuation on the ground state were discussed. The 0 K spin reorientation angles due to quantum fluctuations were found to originate from the asymmetry of the system. As an extended application of the spin-wave study of the multisublattice systems, the spin-wave spectra of a Heisenberg superlattice, which has four or three magnetic atoms or spins, in each magnetic unit cell in the x direction, were solved analytically in terms of creation and annihilation operators. It has been found that the spin-wave spectra of the present superlattice systems depend on the exchange constants J and J', and that the degeneracy of the spin-wave spectra remains for the superlattice of four atomic layers and is partially removed for the superlattice of three atomic layers mainly due to the asymmetry of the system. Another type of splitting of the energy level occurs which differs for those levels with different degeneracies. The mixing of the interface and the bulk modes were found for the spin-wave dispersion of the superlattices. It was predicted that even for different superlattices having the same exchange constants (either J or J'), i.e., without the oscillation of the exchange coupling, the asymmetry can lead to the oscillation of magnetic properties, such as magnetization and giant magnetoresistance.
[S0163-1829(97)04018-6]

I. INTRODUCTION

Since the early work of Bloch,[1] Holstein and Primakoff,[2] Anderson,[3] and Kubo[4] spin waves have attracted considerable interest.[5,6] To our knowledge, few authors have dealt with the spin-wave excitations in the systems with multiple structurally ordered magnetic sublattices.[7–17] It has been commonly accepted that if the lattice is complicated, i.e., the cases when the number of spins per magnetic unit cell is more than two, the problem cannot be solved explicitly in terms of the elements of the matrices for each **k**. Generally, it is necessary to use numerical methods to solve the problem[9–11] and contradictory results were obtained by these authors because of differing methods.[7,12–17] Recently, the spin-wave spectra at low temperatures of a four-sublattice Heisenberg ferrimagnet or ferromagnet with different exchange constants ($J_{ab}=J_{cd}\neq J_{bc}=J_{da}$) were studied in terms of creation and annihilation operators.[18] For diagonalizing the Hamiltonians, two extended Bogoliubov transformations were developed so that the spin-wave spectra of the four-sublattice ferromagnet and ferrimagnet could be calculated explicitly. It seems that the method, used for the multisublattice systems,[18] can be extended to be appropriate for deriving analytical solutions of the spin-wave spectra of magnetic superlattices.

Recently, layered composite materials such as multilayers and superlattices have become of great interest since the magnetic properties of these composite materials may be distinctly different from those of their bulk counterparts.[19–21] The spin waves of magnetic superlattices in the exchange limit have been investigated by use of various quantum microscopic theories.[22–34]

In this paper we first present in Sec. II calculations to investigate the spin waves at low temperatures of three-sublattice Heisenberg systems with different exchange constants ($J_{ab}=J_{bc}\neq J_{ca}$). In Sec. II A, for studying all possible spin configurations, a rotation transformation will be applied to deal with the different quantization axes of the three sublattices. The Hamiltonian for the present system will be reduced by use of the Holstein-Primakoff transform[2] and the linear spin-wave approximation.[3,4] In Sec. III B, the classical ground state of the three-sublattice systems will be examined. In Sec. III C, for avoiding the loss of generality, the general theoretical outline will be represented for treating the problem for the whole present three-sublattice system. The diagonalizing procedure consists of three coupled Cullen transformations,[35] an extended Bogoliubov transformation, and three independent Bogoliubov transformations.[36] The extended Bogoliubov transformation requires us to solve an equation group consisting of 24 equations and 24 unknowns.

To show an extended application of the spin-wave study of the four-sublattice (in the author's previous work[18]) and the three-sublattice (in Sec. II of the present work) systems, attention will be paid on the analytical solutions of the spin-wave spectra of Heisenberg superlattices, formed from two ferromagnetic materials which couple antiferromagnetically at the interfaces, in terms of creation and annihilation operators. The problem for a special case, i.e., a superlattice which has four or three magnetic atoms or spins in each magnetic unit cell in the x direction, will be solved analytically. In the

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present work, for simplicity, we shall only deal with the same amplitudes for the whole system. The Hamiltonian for the superlattice system will be established and reduced by use of the Holstein-Primakoff transform[2] and the linear spin-wave approximation[3,4] in Sec. III A. The Hamiltonian will be rewritten by introducing the Fourier transforms of the boson operators in the reduced Brillouin zone, which will be performed in both the y-z plane and the x direct. After performing the Fourier transforms, one reduces the Hamiltonian for the x direction of the present system to that for a four-sublattice Heisenberg system with different exchange constants ($J_{ab}=J_{cd}=J\neq J_{bc}=J_{da}=J'$)[18] or a three-sublattice Heisenberg system with different exchange constants ($J_{ab}=J_{bc}=J'\neq J_{ca}=J$). In Secs. III B 1 and III B 2, two different extended Bogoliubov transformations[18,36] will be developed for diagonalizing the Hamiltonians in the x direction, for the superlattices with four and three layers, respectively, by establishing the equation group consisting of eight equations and eight unknowns. The spin wave spectra will be derived by solving the equation group (shown in Appendix C for the four-layer superlattice and in Appendixes D and E for the three-layer superlattice) and consequently by performing the transformation. The final forms of the spin-wave spectra of the superlattices are represented in Sec. III C. Discussions and concluding remarks will be represented in Sec. IV.

II. THREE-SUBLATTICE SYSTEM

In this section, we study the three-sublattice Heisenberg system with different isotropic exchange constants between spins in the different sublattices.

A. Hamiltonian and rotate transformation

The three-sublattice system is modeled by the following Hamiltonian:

$$\begin{aligned}\mathbf{H}&=-\sum_{\langle l,i\rangle} J_{l,i;l,i+\delta}\mathbf{S}_{l,i}\cdot\mathbf{S}_{l,i+\delta}\\&=-\sum_{i,\delta} J_{ab}\mathbf{S}_{a,i}\cdot\mathbf{S}_{b,i+\delta}-\sum_{j,\delta} J_{bc}\mathbf{S}_{b,j}\cdot\mathbf{S}_{c,j+\delta}\\&\quad-\sum_{m,\delta} J_{ca}\mathbf{S}_{c,m}\cdot\mathbf{S}_{a,m+\delta}\quad (l=a,b,c),\end{aligned}\tag{2.1}$$

where l denotes three sublattices a, b, and c. δ represents that only the exchanges between the nearest neighbors are taken into account. The number of the nearest neighbors is Z. The sign of the exchange constants can be positive or negative, depending on whether ferromagnetic or ferrimagnetic coupling between them. $S_i=\langle S_i^x, S_i^y, S_i^z\rangle$ are operators belonging to the spin-S representation.[18]

In this work, we shall study the case of $J_{ab}=J_{bc}\neq J_{ca}$ and deal with the case of the same spin amplitudes for the different sublattices. In a low-temperature limit ($T\ll T_c$), usually the spin-wave approximation naturally assumes small spin deviations from the quantization axis. For the present system, the natural quantization axis is the averaged magnetization direction. If the system was near the easy-axis configuration, it would be reasonable to assume that the original ferromagnetic state or ferrimagnetic state is easy axis, in which all spins couple parallel or antiparallel along the z axis. But the initial state in the three-sublattice system can be in various spin configurations, i.e., the quantization axes of the spins in different sublattices may vary on their angles with respect to the z axis. As proposed by del Moral,[7] one needs to rotate the quantization axis frame by Eulerian angles θ and ψ. The following transformation of spin vector coordinates is performed:[7,37,38]

$$\begin{aligned}S_i^x&=S_i^{x'}\cos\theta_i\cos\psi_i+S_i^{z'}\sin\theta_i\cos\psi_i-S_i^{y'}\sin\psi_i\\S_i^y&=S_i^{x'}\cos\theta_i\sin\psi_i+S_i^{z'}\sin\theta_i\sin\psi_i+S_i^{y'}\cos\psi_i\\S_i^z&=S_i^{z'}\cos\theta_i-S_i^{x'}\sin\theta_i\quad (i=a,b,c).\end{aligned}\tag{2.2}$$

Here the (x,y,z) indices refer to the crystal axis frame and the primed ones to the rotated magnetization ones. The commutation relations of the operators $S_i'=\langle S_i^{x'}, S_i^{y'}, S_i^{y'}\rangle$ are the same as those above.[18] By use of the Holstein-Primakoff transform[2] and the linear spin-wave approximation,[3,4] retaining terms up to the second order in the boson operators a_i^+, a_i; b_j^+, b_j; c_m^+, c_m, we have

$$\begin{aligned}\mathbf{H}=\mathbf{H}_0^0&+\sum_l A_l\sum_i \mathbf{a}_{l,i}^+\mathbf{a}_{l,i}\\&+\sum_l \frac{C_{l,l+1}}{Z}\sum_{i\delta}(\mathbf{a}_{l,i}\mathbf{a}_{l+1,i+\delta}+\mathbf{a}_{l,i}^+\mathbf{a}_{l+1,i+\delta}^+)\\&+\sum_l \frac{D_{l,l+1}}{Z}\sum_{i\delta}(\mathbf{a}_{l,i}\mathbf{a}_{l+1,i+\delta}^+\mathbf{a}_{l,i}^+\mathbf{a}_{l+1,i+\delta})\\&+\sum_l \frac{C'_{l,l+1}}{Z}\sum_{i\delta}(\mathbf{a}_{l,i}\mathbf{a}_{l+1,i+\delta}-\mathbf{a}_{l,i}^+\mathbf{a}_{l+1,i+\delta}^+)\\&+\sum_l \frac{D'_{l,l+1}}{Z}\sum_{i\delta}(\mathbf{a}_{l,i}\mathbf{a}_{l+1,i+\delta}^+-\mathbf{a}_{l,i}^+\mathbf{a}_{l+1,i+\delta})\\&+\sum_l \frac{B_l}{\sqrt{N}}\sum_i(\mathbf{a}_{l,i}+\mathbf{a}_{l,i}^+)+\sum_l \frac{B'_l}{\sqrt{N}}\sum_i(\mathbf{a}_{l,i}-\mathbf{a}_{l,i}^+)\\&(l=a,b,c),\end{aligned}\tag{2.3}$$

where

$$\begin{aligned}\mathbf{H}_0^0=-NZS^2\sum_l J_{l,l+1}[&\cos\theta_l\cos\theta_{l+1}\\&+\sin\theta_l\sin\theta_{l+1}\cos(\psi_l-\psi_{l+1})],\end{aligned}\tag{2.4}$$

$$\begin{aligned}A_l=ZS\sum_{j=1}^{2} J_{l,l+j}[&\cos\theta_l\cos\theta_{l+j}\\&+\sin\theta_l\sin\theta_{l+j}\cos(\psi_l-\psi_{l+j})],\end{aligned}\tag{2.5}$$

$$C_{l,l+1}=-\frac{ZS}{2}J_{l,l+1}[\sin\theta_l\sin\theta_{l+1}+(\cos\theta_l\cos\theta_{l+1}-1)\cos(\psi_l-\psi_{l+1})], \quad (2.6)$$

$$D_{l,l+1}=-\frac{ZS}{2}J_{l,l+1}[\sin\theta_l\sin\theta_{l+1}+(\cos\theta_l\cos\theta_{l+1}+1)\cos(\psi_l-\psi_{l+1})] \quad (2.7)$$

$$C'_{l,l+1}=i\frac{ZS}{2}J_{l,l+1}(\cos\theta_l-\cos\theta_{l+1})\sin(\psi_l-\psi_{l+1}), \quad (2.8)$$

$$D'_{l,l+1}=-i\frac{ZS}{2}J_{l,l+1}(\cos\theta_l+\cos\theta_{l+1})\sin(\psi_l-\psi_{l+1}), \quad (2.9)$$

$$B_l=\frac{\sqrt{2NS}S}{2}\sum_{j=1}^{2}J_{l,l+j}[\sin\theta_l\cos\ \theta_{l+j}-\cos\theta_l\sin\theta_{l+j}\cos(\psi_l-\psi_{l+j})], \quad (2.10)$$

$$B'_l=-i\frac{\sqrt{2NS}S}{2}\sum_{j=1}^{2}J_{l,l+j}\sin\theta_{l+j}\sin(\psi_l-\psi_{l+j}). \quad (2.11)$$

Here the parameters $C'_{l,l+1}$, $D'_{l,l+1}$ and B'_l $(l=a,b,c)$ are imaginary, which must be zero in a certain condition (see discussion in Sec. II B).

The Hamiltonian is rewritten by introducing the Fourier transforms of the boson operators in the reduced Brillouin zone:

$$\begin{aligned}\mathbf{H}=\mathbf{H}_0^0&+\sum_l A_l\sum_k \mathbf{a}^+_{l,k}\mathbf{a}_{l,k}\\&+\sum_l C_{l,l+1}\sum_k \gamma_k(\mathbf{a}_{l,k}\mathbf{a}_{l+1,k}+\mathbf{a}^+_{l,k}\mathbf{a}^+_{l+1,k})\\&+\sum_l D_{l,l+1}\sum_k \gamma_k(\mathbf{a}_{l,k}\mathbf{a}^+_{l+1,k}+\mathbf{a}^+_{l,k}\mathbf{a}_{l+1,k})\\&+\sum_l C'_{l,l+1}\sum_k \gamma_k(\mathbf{a}_{l,k}\mathbf{a}_{l+1,k}-\mathbf{a}^+_{l,k}\mathbf{a}^+_{l+1,k})\\&+\sum_l D'_{l,l+1}\sum_k \gamma_k(\mathbf{a}_{l,k}\mathbf{a}^+_{l+1,k}-\mathbf{a}^+_{l,k}\mathbf{a}_{l+1,k})\\&+\sum_l B_l(\mathbf{a}_{l,0}+\mathbf{a}^+_{l,0})+\sum_l B'_l(\mathbf{a}_{l,0}-\mathbf{a}^+_{l,0})\end{aligned} \quad (l=a,b,c), \quad (2.12)$$

with

$$\gamma_k=\frac{1}{Z}\sum_\delta e^{i\mathbf{k}\cdot\boldsymbol{\delta}}. \quad (2.13)$$

B. Classical ground state

Before studying the spin waves of a magnetic system, one needs to examine carefully the classical ground state of the system. Therefore, in this section, we shall investigate the classical ground state of the present three-sublattice system.

The classical energy E_0 of the present system is described by the Hamiltonian $\mathbf{H}_0^0$ shown in Eq. (2.4). The equilibrium state is found by minimizing Eq. (2.4) with respect to the angles θ_l and ψ_l $(l=a,b,c)$. This involves the nonlinear equations

$$\frac{\partial E_0}{\partial\theta_l}=NZS^2\sum_{j=1}^{2}J_{l,l+j}[\sin\theta_l\cos\theta_{l+j}-\cos\theta_l\sin\theta_{l+j}\cos(\psi_l-\psi_{l+j})]. \quad (2.14)$$

$$\frac{\partial E_0}{\partial\psi_l}=NZS^2\sum_{j=1}^{2}J_{l,l+j}\sin\theta_l\sin\theta_{l+j}\sin(\psi_l-\psi_{l+j}) \quad (2.15)$$

and the second derivatives of the free energy with respect to the angles θ_l and ψ_l $(l=a,b,c)$:

$$\frac{\partial^2 E_0}{\partial\theta_l^2}=NZS^2\sum_{j=1}^{2}J_{l,l+j}[\cos\theta_l\cos\theta_{l+j}+\sin\theta_l\sin\theta_{l+j}\cos(\psi_l-\psi_{l+j})], \quad (2.16)$$

$$\frac{\partial^2 E_0}{\partial\theta_l\partial\theta_{l+j}}=-NZS^2J_{l,l+j}[\sin\theta_l\sin\theta_{l+j}+\cos\ \theta_l\ \cos\ \theta_{l+j}\ \cos(\psi_l-\psi_{l+j})] \quad (j=1,2), \quad (2.17)$$

$$\frac{\partial^2 E_0}{\partial\psi_l^2}=NZS^2\sum_{j=1}^{2}J_{l,l+j}\sin\theta_l\sin\theta_{l+j}\cos(\psi_l-\psi_{l+j}), \quad (2.18)$$

$$\frac{\partial^2 E_0}{\partial\psi_l\partial\psi_{l+j}}=-NZS^2J_{l,l+j}\sin\theta_l\sin\theta_{l+j}\cos(\psi_l-\psi_{l+j}) \quad (j=1,2), \quad (2.19)$$

$$\frac{\partial^2 E_0}{\partial\theta_l\partial\psi_l}=NZS^2\sum_{j=1}^{2}J_{l,l+j}\cos\theta_l\sin\theta_{l+j}\sin(\psi_l-\psi_{l+j}), \quad (2.20)$$

$$\frac{\partial^2 E_0}{\partial\theta_l\partial\psi_{l+j}}=-NZS^2J_{l,l+j}\cos\theta_l\sin\theta_{l+j}\sin(\psi_l-\psi_{l+j}) \quad (j=1,2), \quad (2.21)$$

From Eqs. (2.14) and (2.15), one finds a simple condition for the equilibrium state:

$$\sin(\psi_a-\psi_b)=\sin(\psi_b-\psi_c)=\sin(\psi_c-\psi_a)=0. \quad (2.22)$$

This equals

$$\psi_a-\psi_b=\psi_b-\psi_c=\psi_c-\psi_a=0\ \text{ or }\ \pi. \quad (2.23)$$

As soon as Eqs. (2.22) and (2.23) are satisfied, one has

$$\sin(\theta_a \mp \theta_b) = \sin(\theta_b \mp \theta_c) = \sin(\theta_c \mp \theta_a) = 0, \quad (2.24)$$

where the negative or positive sign correspond to 0 or π in Eq. (2.23).

One finds another trivial condition from Eqs. (2.14) and (2.15):

$$\sin\theta_a = \sin\theta_b = \sin\theta_c = 0, \quad (2.25)$$

namely,

$$\theta_a = \theta_b = \theta_c = 0 \text{ or } \pi. \quad (2.26)$$

Actually, the condition of Eqs. (2.25) and (2.26) coincides with that of Eqs. (2.22) and (2.23).

To investigate whether such solutions indeed correspond to energy minima one has to evaluate the second derivatives Eqs. (2.16)–(2.21) and consider the expression

$$D = \left(h_a \frac{\partial}{\partial \theta_a} + h_b \frac{\partial}{\partial \theta_b} + h_c \frac{\partial}{\partial \theta_c} + g_a \frac{\partial}{\partial \psi_a} + g_b \frac{\partial}{\partial \psi_b} + g_c \frac{\partial}{\partial \psi_c} \right)^2 E_0. \quad (2.27)$$

The criterion $D>0$, as

$$\sqrt{h_a^2+h_b^2+h_c^2+g_a^2+g_b^2+g_c^2}$$

is very small, ensures the existence of a minimum. After some manipulation, one can get an expression for D:

$$\begin{aligned} D = NZS^2 \sum_{l=a,b,c} J_{l,l+1} \{ & \cos\theta_l \cos\theta_{l+1} [h_l^2 + h_{l+1}^2 - 2h_l h_{l+1} \cos(\psi_l - \psi_{l+1})] \\ & + \sin\theta_l \sin\theta_{l+1} [(h_l^2 + h_{l+1}^2)\cos(\psi_l - \psi_{l+1}) - 2h_l h_{l+1}] \\ & + \sin\theta_l \sin\theta_{l+1} \cos(\psi_l - \psi_{l+1})(g_l - g_{l+1})^2 \\ & + 2\sin(\psi_l - \psi_{l+1})(g_l - g_{l+1})(h_l \cos\theta_l \sin\theta_{l+1} \\ & + h_{l+1} \sin\theta_l \cos\theta_{l+1}) \}. \end{aligned} \quad (2.28)$$

Using the condition of Eq. (2.22), one finds

$$D = NZS^2 \sum_{l=a,b,c} J_{l,l+1} [\cos(\theta_l \mp \theta_{l+1})(h_l \mp h_{l+1})^2 \pm \sin\theta_l \sin\theta_{l+1} (g_l - g_{l+1})^2]. \quad (2.29)$$

Here the upper or lower sign corresponds to the solution of 0 or π in Eq. (2.23). For a minimum, the values of D must be always positive, being independent of the values of h_l and g_l $(l=a,b,c)$. It is easy to verify that a minimum can be found only if (i) $\theta_l = \theta_{l+1}$ and $\psi_l = \psi_{l+1}$ when $J_{l,l+1}>0$ and (ii) $\theta_l - \theta_{l+1} = \pi$ and $\psi_l = \psi_{l+1}$ when $J_{l,l+1}<0$. One may note that the solutions found above are degeneracy for all possible values of θ_l and ψ_l being valid for the above conditions. This degeneracy can be removed by adding the terms of the anisotropy and/or the external field into the Hamiltonian (2.1). To simplify, in Sec. III, we shall not take the terms of the anisotropy and the external field into account, but only assume that the spins orient preferentially in their particular directions.

One finds the nontrivial solutions of Eqs. (2.14) and (2.15) as

$$\cos(\psi_l - \psi_{l+1}) = \frac{J_{l+1,l+2} J_{l+2,l} \sin^2\theta_{l+2}}{2J_{l,l+1}^2 \sin\theta_l \sin\theta_{l+1}} - \frac{J_{l+1,l+2}\sin\theta_{l+1}}{2J_{l+2,l}\sin\theta_l} - \frac{J_{l+2,l}\sin\theta_l}{2J_{l+1,l+2}\sin\theta_{l+1}}, \quad (2.30)$$

where the angles θ_l $(l=a,b,c)$ are limited by

$$\sum_{l=a,b,c} J_{l,l+1} J_{l+2,l} \cos\theta_l = 0. \quad (2.31)$$

Then, for the nontrivial solutions, the dependence of the equilibrium orientations can be studied with the aid of computer. According to Eq. (2.31), actually, the nontrivial solutions are degeneracy also.

On the other hand, it is reasonable to remove all imaginary terms in the Hamiltonians (2.3) and (2.12). It is easy to verify that all the imaginary terms can be eliminated only if either the trivial condition (2.22) or the nontrivial condition (2.30) with $\cos\theta_l = 0$ of the equilibrium orientations are satisfied. The condition of $\cos\theta_l = 0$ means that the noncollinear spin configurations are limited inside the basal plane. This limitation is attributed to one of the quantum effects of the spin waves on the classical ground state. Then the Hamiltonian (2.12) can be written as

$$\begin{aligned} \mathbf{H} = \mathbf{H}_0^0 & + A_a \sum_k \mathbf{a}_k^+ \mathbf{a}_k + A_b \sum_k \mathbf{b}_k^+ \mathbf{b}_k + A_c \sum_k \mathbf{c}_k^+ \mathbf{c}_k \\ & + C_{ab} \sum_k \gamma_k (\mathbf{a}_k^+ \mathbf{b}_k^+ + \mathbf{a}_k \mathbf{b}_k) + D_{ab} \sum_k \gamma_k (\mathbf{a}_k^+ \mathbf{b}_k + \mathbf{a}_k \mathbf{b}_k^+) \\ & + C_{bc} \sum_k \gamma_k (\mathbf{b}_k^+ \mathbf{c}_k^+ + \mathbf{b}_k \mathbf{c}_k) + D_{bc} \sum_k \gamma_k (\mathbf{b}_k^+ \mathbf{c}_k + \mathbf{b}_k \mathbf{c}_k^+) \\ & + C_{ca} \sum_k \gamma_k (\mathbf{c}_k^+ \mathbf{a}_k^+ + \mathbf{c}_k \mathbf{a}_k) + D_{ca} \sum_k \gamma_k (\mathbf{c}_k^+ \mathbf{a}_k + \mathbf{c}_k \mathbf{a}_k^+) \\ & + B_a (\mathbf{a}_0^+ + \mathbf{a}_0) + B_b (\mathbf{b}_0^+ + \mathbf{b}_0) + B_c (\mathbf{c}_0^+ + \mathbf{c}_0). \end{aligned} \quad (2.32)$$

C. General theoretical outline for diagonalizing the Hamiltonian

In this section, we represent the general theoretical outline for diagonalizing the Hamiltonian (2.32). For the three-sublattice system, in order to eliminate the nondiagonal terns, a three-step diagonalizing procedure should be developed, which consists of three coupled Cullen transformations,[7,35] an extended Bogoliubov transformation and three independent Bogoliubov transformations.[36]

1. The first step: three coupled Cullen transformations

The linear terms in $\mathbf{a}_0^+$, $\mathbf{a}_0$; $\mathbf{b}_0^+$, $\mathbf{b}_0$; $\mathbf{c}_0^+$, $\mathbf{c}_0$ in Eq. (2.32) can be eliminated by using three coupled Cullen diagonalization transformations,[7,35]

$$\mathbf{a}_k=\mathbf{d}_k+\mathbf{h}_k\delta_{k,0}, \tag{2.33a}$$

$$\mathbf{a}_k^+=\mathbf{d}_k^++\mathbf{h}_k^+\delta_{k,0}, \tag{2.33b}$$

$$\mathbf{b}_k=\mathbf{f}_k+\mathbf{p}_k\delta_{k,0}, \tag{2.34a}$$

$$\mathbf{b}_k^+=\mathbf{f}_k^++\mathbf{p}_k^+\delta_{k,0}, \tag{2.34b}$$

and

$$\mathbf{c}_k=\mathbf{g}_k+\mathbf{q}_k\delta_{k,0}, \tag{2.35a}$$

$$\mathbf{c}_k^+=\mathbf{g}_k^++\mathbf{q}_k^+\delta_{k,0}, \tag{2.35b}$$

where the Fourier transforms $\mathbf{h}_0$, $\mathbf{p}_0$, and $\mathbf{q}_0$ represent frozen-in uniform spin deviations. The Hamiltonian (2.32) will be diagonalized only if the following conditions are satisfied:

$$A_a\mathbf{h}_0+C_{ab}\mathbf{p}_0^++D_{ab}\mathbf{p}_0+C_{ca}\mathbf{q}_0^++D_{ca}\mathbf{q}_0+B_a=0, \tag{2.36a}$$

$$A_b\mathbf{p}_0+C_{ab}\mathbf{h}_0^++D_{ab}\mathbf{h}_0+C_{bc}\mathbf{q}_0^++D_{bc}\mathbf{q}_0+B_b=0, \tag{2.36b}$$

$$A_c\mathbf{q}_0+C_{bc}\mathbf{p}_0^++D_{bc}\mathbf{p}_0+C_{ca}\mathbf{h}_0^++D_{ca}\mathbf{h}_0+B_c=0. \tag{2.36c}$$

Values for $\mathbf{h}_0^+$, $\mathbf{h}_0$; $\mathbf{p}_0^+$, $\mathbf{p}_0$; $\mathbf{q}_0^+$, $\mathbf{q}_0$ can be derived by solving the above linear equation group, consisting of Eqs. (2.36a)–(2.36c) which can be separated into six equations. The new diagonalized Hamiltonian contains $\mathbf{k}\neq 0$ terms identical to Eq. (2.32), but with $\mathbf{a}_k^+$, $\mathbf{a}_k$; $\mathbf{b}_k^+$, $\mathbf{b}_k$; $\mathbf{c}_k^+$, $\mathbf{c}_k$ substituted by the $\mathbf{d}_k^+$, $\mathbf{d}_k$; $\mathbf{f}_k^+$, $\mathbf{f}_k$; $\mathbf{g}_k^+$, $\mathbf{g}_k$ operators and a new $\mathbf{k}=0$ term

$$\begin{aligned}\mathbf{H}_0^1=&A_a|\mathbf{h}_0|^2+A_b|\mathbf{p}_0|^2+A_c|\mathbf{q}_0|^2+C_{ab}(\mathbf{h}_0^+\mathbf{p}_0^++\mathbf{h}_0\mathbf{p}_0)\\&+D_{ab}(\mathbf{h}_0^+\mathbf{p}_0+\mathbf{h}_0\mathbf{p}_0^+)+C_{bc}(\mathbf{p}_0^+\mathbf{q}_0^++\mathbf{p}_0\mathbf{q}_0)\\&+D_{bc}(\mathbf{p}_0^+\mathbf{q}_0+\mathbf{p}_0\mathbf{q}_0^+)+C_{ca}(\mathbf{q}_0^+\mathbf{h}_0^++\mathbf{q}_0\mathbf{h}_0)\\&+D_{ca}(\mathbf{q}_0^+\mathbf{h}_0+\mathbf{q}_0\mathbf{h}_0^+)+B_a(\mathbf{h}_0^++\mathbf{h}_0)+B_b(\mathbf{p}_0^++\mathbf{p}_0)\\&+B_c(\mathbf{q}_0^++\mathbf{q}_0)\end{aligned} \tag{2.37a}$$

which represents the uniform ($k=0$) quantum spin-canting fluctuations. Since $\mathbf{h}_0^+=\mathbf{h}_0$, $\mathbf{p}_0^+=\mathbf{p}_0$, and $\mathbf{q}_0^+=\mathbf{q}_0$, Eq. (2.37a) can actually be written in a simpler form:

$$\begin{aligned}\mathbf{H}_0^1=&A_a\mathbf{h}_0^2+A_b\mathbf{p}_0^2+A_c\mathbf{q}_0^2+2(C_{ab}+D_{ab})\mathbf{h}_0\mathbf{p}_0\\&+2(C_{bc}+D_{bc})\mathbf{p}_0\mathbf{q}_0+2(C_{ca}+D_{ca})\mathbf{q}_0\mathbf{h}_0+2B_a\mathbf{h}_0\\&+2B_b\mathbf{p}_0+2B_c\mathbf{q}_0.\end{aligned} \tag{2.37b}$$

2. The second step: an extended Bogoliubov transformation

To remove the nondiagonal terms $\mathbf{d}_k^+\mathbf{f}_k^++\mathbf{d}_k\mathbf{f}_k$, $\mathbf{d}_k^+\mathbf{f}_k+\mathbf{d}_k\mathbf{f}_k^+$, $\mathbf{f}_k^+\mathbf{g}_k^++\mathbf{f}_k\mathbf{g}_k$, $\mathbf{f}_k^+\mathbf{g}_k+\mathbf{f}_k\mathbf{g}_k^+$, $\mathbf{g}_k^+\mathbf{d}_k^++\mathbf{g}_k\mathbf{d}_k$, $\mathbf{g}_k^+\mathbf{d}_k+\mathbf{g}_k\mathbf{d}_k^+$, an extended Bogoliubov transformation should be developed as the following matrix:

$$\begin{pmatrix}\alpha_k^+\\ \alpha_k\\ \beta_k^+\\ \beta_k\\ \xi_k^+\\ \xi_k\end{pmatrix}=\begin{pmatrix}a_{1k}&a_{2k}&a_{3k}&a_{4k}&a_{5k}&a_{6k}\\ a_{2k}&a_{1k}&a_{4k}&a_{3k}&a_{6k}&a_{5k}\\ a_{3k}&a_{4k}&a_{7k}&a_{8k}&a_{9k}&a_{10k}\\ a_{4k}&a_{3k}&a_{8k}&a_{7k}&a_{10k}&a_{9k}\\ a_{5k}&a_{6k}&a_{9k}&a_{10k}&a_{11k}&a_{12k}\\ a_{6k}&a_{5k}&a_{10k}&a_{9k}&a_{12k}&a_{11k}\end{pmatrix}\begin{pmatrix}d_k^+\\ d_k\\ f_k^+\\ f_k\\ g_k^+\\ g_k\end{pmatrix} \tag{2.38}$$

and consequently its reversed transformation[18] with parameters A_{jk} ($j=1,2,\ldots,12$). The relationship between the parameters a_{ik} and A_{jk} ($i=1,2,\ldots,12$ and $j=1,2,\ldots,12$) gives 12 equations which are omitted here for simplicity. The commutation relations of the new operators lead to six equations. For instance,

$$[\alpha_k,\alpha_{k'}^+]=\delta_{kk'},$$

$$[\alpha_k,\beta_{k'}^+]=0,$$

respectively, lead to

$$a_{1k}^2+a_{3k}^2+a_{5k}^2-a_{2k}^2-a_{4k}^2-a_{6k}^2=1, \tag{2.39}$$

$$a_{1k}a_{3k}+a_{3k}a_{7k}+a_{5k}a_{9k}-a_{2k}a_{4k}-a_{4k}a_{8k}-a_{6k}a_{10k}=0. \tag{2.40}$$

For eliminating the nondiagonal terms, i.e., $\alpha_k^+\beta_k^++\alpha_k\beta_k$, $\alpha_k^+\beta_k+\alpha_k\beta_k^+$, $\alpha_k^+\xi_k^++\alpha_k\xi_k$, $\alpha_k^+\xi_k+\alpha_k\xi_k^+$, $\beta_k^+\xi_k^++\beta_k\xi_k$, and $\beta_k^+\xi_k+\beta_k\xi_k^+$, one needs to establish six equations (omitted also for simplicity). Then the problem becomes to solve the equation group, mentioned above, consisting of 24 equations and 24 unknowns. After solving this equation group and performing the transformation, the Hamiltonian may be written in the following form:

$$\begin{aligned}\mathbf{H}=&\mathbf{H}_0^0+\mathbf{H}_0^1+\mathbf{H}_0^2+\sum_k A_{k1}\alpha_k^+\alpha_k+\sum_k B_{k1}\beta_k^+\beta_k\\&+\sum_k C_{k1}\xi_k^+\xi_k+\sum_k A_{k2}(\alpha_k^+\alpha_k^++\alpha_k\alpha_k)\\&+\sum_k B_{k2}(\beta_k^+\beta_k^++\beta_k\beta_k)+\sum_k C_{k2}(\xi_k^+\xi_k^++\xi_k\xi_k).\end{aligned} \tag{2.41}$$

The parameters in Hamiltonian (2.41) are described in Appendix A.

For the present system with exchange constants $J_{ab}=J_{bc}\neq J_{ca}$, the conditions of Eq. (2.22) and $\cos(\theta_a-\theta_b)=\cos(\theta_b-\theta_c)$ are commonly satisfied. Then one has $A_a=A_c$, $C_{ab}=C_{bc}$, and $D_{ab}=D_{bc}$ and thus the transformation matrix (2.38) and consequently the equation group can be simplified to be as those represented in Appendix B.

3. The third step: three independent Bogoliubov transformations

In this step, one needs to remove the nondiagonal terms in Eq. (2.41) by a further diagonalizing procedure, consisting of three independent Bogoliubov transformations,[36] one of which is described as

$$\alpha_k^+ = l_{1k}\zeta_k^+ + l_{2k}\zeta_k , \tag{2.42a}$$

$$\alpha_k = l_{1k}\zeta_k + l_{2k}\zeta_k^+ , \tag{2.42b}$$

with

$$l_{1k} = \left(\frac{1+\epsilon_k}{2\epsilon_k} \right)^{1/2} \tag{2.43a}$$

$$l_{2k} = \left(\frac{1-\epsilon_k}{2\epsilon_k} \right)^{1/2} \tag{2.43b}$$

$$\epsilon_k = \left[1 - \left(\frac{2A_{k2}}{A_{k1}} \right)^2 \right]^{1/2} \tag{2.43c}$$

The other two (with the parameters $m_{1k}, m_{2k}; n_{1k}, n_{2k}$) are similar to these.

After performing the three Bogoliubov transformations, one obtains

$$\mathbf{H} = \mathbf{H}_0^0 + \mathbf{H}_0^1 + \mathbf{H}_0^2 + \mathbf{H}_0^3 + \sum_k \hbar\omega_{k,1}\zeta_k^+\zeta_k + \sum_k \hbar\omega_{k,2}\eta_k^+\eta_k + \sum_k \hbar\omega_{k,3}\nu_k^+\nu_k . \tag{2.44}$$

Here

$$\mathbf{H}_0^3 = \frac{1}{2}\sum_k \left[\sqrt{A_{k1}^2 - 4A_{k2}^2} - A_{k1} + \sqrt{B_{k1}^2 - 4B_{k2}^2} - B_{k1} + \sqrt{C_{k1}^2 - 4C_{k2}^2} - C_{k1} \right], \tag{2.45}$$

$$\hbar\omega_{k,1} = \sqrt{A_{k1}^2 - 4A_{k2}^2}, \tag{2.46}$$

$$\hbar\omega_{k,2} = \sqrt{B_{k1}^2 - 4B_{k2}^2}, \tag{2.47}$$

$$\hbar\omega_{k,3} = \sqrt{C_{k1}^2 - 4C_{k2}^2}. \tag{2.48}$$

It should be noticed that if the condition of Eq. (2.22) and $\cos(\theta_a - \theta_b) = \cos(\theta_b - \theta_c)$ is satisfied, one will have $A_a = A_c$, $C_{ab} = C_{bc}$, and $D_{ab} = D_{bc}$ and thus $A_{k1} = C_{k1}$ and $A_{k2} = C_{k2}$ which result in $n_{1k} = l_{1k}$ and $n_{2k} = l_{2k}$, respectively. Then one has

$$\hbar\omega_{k,1} = \hbar\omega_{k,3} \tag{2.49}$$

and consequently the Hamiltonian (2.44) and (2.45) can be rewritten as

$$\mathbf{H} = \mathbf{H}_0^0 + \mathbf{H}_0^1 + \mathbf{H}_0^2 + \mathbf{H}_0^3 + \sum_k \hbar\omega_{k,1}(\zeta_k^+\zeta_k + \nu_k^+\nu_k) + \sum_k \hbar\omega_{k,2}\eta_k^+\eta_k \tag{2.50}$$

and

$$\mathbf{H}_0^3 = \frac{1}{2}\sum_k \left[2\sqrt{A_{k1}^2 - 4A_{k2}^2} - 2A_{k1} + \sqrt{B_{k1}^2 - 4B_{k2}^2} - B_{k1} \right]. \tag{2.51}$$

Generally speaking, the Hamiltonian of the present three-sublattice Heisenberg system can be diagonalized by the three-step diagonalizing procedure, developed above. However, in most cases, such a procedure is very complex since at least 16 equations and 16 unknowns are involved in the equation group.

D. Quantum fluctuations on the ground state

Besides the quantum effect on the classical ground state, found in Sec. II B, which limits the noncollinear spin configurations to be within the basal plane, one may discuss some quantum fluctuations on the classical ground state. The quantum fluctuations on the ground state originate from Hamiltonians $\mathbf{H}_0^1$, $\mathbf{H}_0^2$, and $\mathbf{H}_0^3$. One needs to repeat the procedure in Sec. II B, discussing the energy minima of the system. In this case, the energies of the system include not only the Hamiltonian $\mathbf{H}_0^0$ but also the quantum fluctuations $\mathbf{H}_0^1$, $\mathbf{H}_0^2$, and $\mathbf{H}_0^3$. A numerical method needs to be employed for finding the energy minima.

If the initial state was assumed to be of the completely ordered state, in which all spins couple parallel or antiparallel along the z axis, the diagonalizing procedures in Sec. II C could be simplified so that only one step is necessary for diagonalizing. The uniform ($k=0$) quantum spin-canting fluctuations disappear. The remaining quantum fluctuation on the ground state is the zero-point vibrating of the spin waves.

III. SUPERLATTICES

The infinite Heisenberg superlattices, studied in this work, are formed from two ferromagnetic materials with the simple cubic lattice, which couple antiferromagnetically at the interfaces. The layered structure in which an elementary unit consisting of q (100) atomic planes is repeated periodically along the stacking direction parallel to the x axis of the coordinate system.

A. Hamiltonian

The superlattices can be described by the Hamiltonian

$$\mathbf{H} = -\frac{1}{2}\sum_{l=1}^{q}\sum_{\rho,\delta} J_{l,\rho;l,\rho+\delta}\mathbf{S}_{l,\rho}\cdot\mathbf{S}_{l,\rho+\delta} = -\frac{1}{2}\sum_{l=1}^{q}\sum_{\rho}\sum_{\delta_\parallel} J_l\mathbf{S}_{l,\rho}\cdot\mathbf{S}_{l,\rho+\delta_\parallel} - \sum_{l=1}^{q}\sum_{\rho} J_{l,l+1}\mathbf{S}_{l,\rho}\cdot\mathbf{S}_{l+1,\rho}, \tag{3.1}$$

where l denotes the spins in the elementary unit in the x direction. δ represents that only the exchanges between the nearest neighbors are taken into account while $\delta_\parallel$ denotes the nearest neighbors within one plane parallel to the y-z plane of the coordinate system.

When the number q of the planes in the magnetic unit cell is chosen to be 4, the Hamiltonian (3.1) is reduced to

$$\mathbf{H}=-\frac{1}{2}\sum_{l=1}^{4}\sum_{\rho,\delta_{\parallel}}J_l\mathbf{S}_{l,\rho}\cdot\mathbf{S}_{l,\rho+\delta_{\parallel}}-\sum_{i,\delta_x}J_{12}\mathbf{S}_{1,i}\cdot\mathbf{S}_{2,i+\delta_x}$$
$$-\sum_{j,\delta_x}J_{23}\mathbf{S}_{2,j}\cdot\mathbf{S}_{3,j+\delta_x}-\sum_{m,\delta_x}J_{34}\mathbf{S}_{3,m}\cdot\mathbf{S}_{4,m+\delta_x}$$
$$-\sum_{n,\delta_x}J_{41}\mathbf{S}_{4,n}\cdot\mathbf{S}_{1,n+\delta_x}. \tag{3.2a}$$

For the superlattice with three different atomic planes, $q=3$ and the Hamiltonian (3.1) becomes

$$\mathbf{H}=-\frac{1}{2}\sum_{l=1}^{3}\sum_{\rho,\delta_{\parallel}}J_l\mathbf{S}_{l,\rho}\cdot\mathbf{S}_{l,\rho+\delta_{\parallel}}-\sum_{i,\delta_x}J_{12}\mathbf{S}_{1,i}\cdot\mathbf{S}_{2,i+\delta_x}$$
$$-\sum_{j,\delta_x}J_{23}\mathbf{S}_{2,j}\cdot\mathbf{S}_{3,j+\delta_x}-\sum_{m,\delta_x}J_{31}\mathbf{S}_{3,m}\cdot\mathbf{S}_{1,m+\delta_x}. \tag{3.2b}$$

The first term on the right-hand side of Eq. (3.2a) or Eq. (3.2b) is the Hamiltonian for the exchanges between the spins with the neighbors within the y-z planes. Others are for the exchanges between the spins in different planes in the x direction. δ_x denotes the nearest neighbor in the x direction.

If one assumes the following conditions: (i) All exchange constants J_1 for the exchanges within the y-z planes are equal, i.e., $J_1=J_2=J_3=J_4=J>0$ and (ii) For the exchange constants between the different planes and for the four-layer superlattice, one has $J_{12}=J_{34}=J>0$ and $J_{23}=J_{41}=J'<0$, the Hamiltonian Eq. (3.2a) may be written as

$$\mathbf{H}=-\frac{1}{2}J\sum_{l=1}^{4}\sum_{\rho,\delta_{\parallel}}\mathbf{S}_{l,\rho}\cdot\mathbf{S}_{l,\rho+\delta_{\parallel}}-J\sum_{i,\delta_x}\mathbf{S}_{1,i}\cdot\mathbf{S}_{2,i+\delta_x}$$
$$-J'\sum_{j,\delta_x}\mathbf{S}_{2,j}\cdot\mathbf{S}_{3,j+\delta_x}-J\sum_{m,\delta_x}\mathbf{S}_{3,m}\cdot\mathbf{S}_{4,m+\delta_x}$$
$$-J'\sum_{n,\delta_x}\mathbf{S}_{4,n}\cdot\mathbf{S}_{1,n+\delta_x}. \tag{3.3a}$$

For the three-layer superlattice, one has $J_{12}=J_{23}=J'<0$ and $J_{31}=J>0$, the Hamiltonian Eq. (3.2b) may be written as

$$\mathbf{H}=-\frac{1}{2}J\sum_{l=1}^{3}\sum_{\rho,\delta_{\parallel}}\mathbf{S}_{l,\rho}\cdot\mathbf{S}_{l,\rho+\delta_{\parallel}}-J'\sum_{i,\delta_x}\mathbf{S}_{1,i}\cdot\mathbf{S}_{2,i+\delta_x}$$
$$-J'\sum_{j,\delta_x}\mathbf{S}_{2,j}\cdot\mathbf{S}_{3,j+\delta_x}-J\sum_{m,\delta_x}\mathbf{S}_{3,m}\cdot\mathbf{S}_{1,m+\delta_x}. \tag{3.3b}$$

In this section, we shall still restrict ourselves to the low temperature region of $T\ll T_c$. The initial state is assumed to be of the completely ordered state, in which all spins couple parallel or antiparallel along the z axis, in accordance with the signs of the exchange constants J and J'. The linearized Holstein-Primakoff transformation[2–4] enables us to rewrite the Hamiltonians in the following forms:

$$\mathbf{H}=\mathbf{H}_0+S(ZJ+Z_x|J'|)\sum_{l=1}^{4}\sum_{\rho}\mathbf{b}_{l,\rho}^{+}\mathbf{b}_{l,\rho}$$
$$-\frac{1}{2}SJ\sum_{l=1}^{4}\sum_{\rho,\delta_{\parallel}}[\mathbf{b}_{l,\rho}^{+}\mathbf{b}_{l,\rho+\delta_{\parallel}}+\mathbf{b}_{l,\rho}\mathbf{b}_{l,\rho+\delta_{\parallel}}^{+}]$$
$$-S\Big[J\sum_{i\delta_x}(\mathbf{b}_{1,i}^{+}\mathbf{b}_{2,i+\delta_x}+\mathbf{b}_{1,i}\mathbf{b}_{2,i+\delta_x}^{+})$$
$$+J'\sum_{j\delta_x}(\mathbf{b}_{2,j}^{+},\mathbf{b}_{3,j+\delta_x}^{+}+\mathbf{b}_{2,j}\mathbf{b}_{3,j+\delta_x})$$
$$+J\sum_{m\delta_x}(\mathbf{b}_{3,m}^{+}\mathbf{b}_{4,m+\delta_x}+\mathbf{b}_{3,m}\mathbf{b}_{4,m+\delta_x}^{+})$$
$$+J'\sum_{n\delta_x}(\mathbf{b}_{4,n}^{+}\mathbf{b}_{1,n+\delta_x}+\mathbf{b}_{4,n}\mathbf{b}_{1,n+\delta_x})\Big] \tag{3.4a}$$

with

$$\mathbf{H}_0=-2NS^2(ZJ+Z_x|J'|) \tag{3.5a}$$

for the four-layer superlattice and

$$\mathbf{H}=\mathbf{H}_0+S(ZJ+Z_x|J'|)\sum_{l=1}^{3}\sum_{\rho}\mathbf{b}_{l,\rho}^{+}\mathbf{b}_{l,\rho}$$
$$-\frac{1}{2}SJ\sum_{l=1}^{3}\sum_{\rho,\delta_{\parallel}}[\mathbf{b}_{l,\rho}^{+}\mathbf{b}_{l,\rho+\delta_{\parallel}}+\mathbf{b}_{l,\rho}\mathbf{b}_{l,\rho+\delta_{\parallel}}^{+}]$$
$$-S\Big[J'\sum_{i\delta_x}(\mathbf{b}_{1,i}^{+}\mathbf{b}_{2,i+\delta_x}^{+}+\mathbf{b}_{1,i}\mathbf{b}_{2,i+\delta_x})$$
$$+J'\sum_{j\delta_x}(\mathbf{b}_{2,j}^{+}\mathbf{b}_{3,j+\delta_x}^{+}+\mathbf{b}_{2,j}\mathbf{b}_{3,j+\delta_x})$$
$$+J\sum_{m\delta_x}(\mathbf{b}_{3,m}^{+}\mathbf{b}_{1,m+\delta_x}+\mathbf{b}_{3,m}\mathbf{b}_{1,m+\delta_x}^{+})\Big] \tag{3.4b}$$

with

$$\mathbf{H}_0=-NS^2[(Z+\tfrac{1}{2}Z_{yz})J+2Z_x|J'|] \tag{3.5b}$$

for the three-layer superlattice, respectively. Here Z_x or Z_{yz} is the number of the nearest neighbors in the x direction or the y-z plane for a spin.

The superlattice is periodic not only in the y and z directions, but also in the x direction. The superlattice has a larger periodicity in the x direction perpendicular to the slabs. Thus one expects to have solutions of the Bloch form for the three directions. The Hamiltonians (3.4a) and (3.4b) can be rewritten by introducing the Fourier transforms of the boson operators in the reduced Brillouin zone:

$$\mathbf{H}=\mathbf{H}_0+S\sum_{l=1}^{4}\sum_{k}[Z_x|J'|+ZJ(1-\gamma_{k_{\parallel}})]\mathbf{b}_{1,k}^{+}\mathbf{b}_{1,k}$$
$$-ZS\sum_{k}\gamma_{k_x}[J(\mathbf{b}_{1,k}^{+}\mathbf{b}_{2,k}+\mathbf{b}_{1,k}\mathbf{b}_{2,k}^{+})+J'(\mathbf{b}_{2,k}^{+}\mathbf{b}_{3,k}^{+}+\mathbf{b}_{2,k}\mathbf{b}_{3,k})$$
$$+J(\mathbf{b}_{3,k}^{+}\mathbf{b}_{4,k}+\mathbf{b}_{3,k}\mathbf{b}_{4,k}^{+})+J'(\mathbf{b}_{4,k}^{+}\mathbf{b}_{1,k}^{+}+\mathbf{b}_{4,k}\mathbf{b}_{1,k})] \tag{3.6a}$$

for the four-layer superlattice and

$$\mathbf{H}=\mathbf{H}_0+S\sum_{l=1,3}\sum_k\ [Z_x|J'|+ZJ(1-\gamma_{k_\parallel})]\mathbf{b}^+_{1,k}\mathbf{b}_{1,k}$$
$$+S\sum_k\ [2Z_x|J'|+J(Z_{yz}-Z\gamma_{k_\parallel})]\mathbf{b}^+_{2,k}\mathbf{b}_{2,k}$$
$$-ZS\sum_k\ \gamma_{k_x}[J'(\mathbf{b}^+_{1,k}\mathbf{b}^+_{2,k}+\mathbf{b}_{1,k}\mathbf{b}_{2,k})$$
$$+J'(\mathbf{b}^+_{2,k}\mathbf{b}^+_{3,k}+\mathbf{b}_{2,k}\mathbf{b}_{3,k})+J(\mathbf{b}^+_{3,k}\mathbf{b}_{1,k}+\mathbf{b}_{3,k}\mathbf{b}^+_{1,k})] \tag{3.6b}$$

for the three-layer superlattice, respectively, with

$$\gamma_{k_\parallel}=\frac{1}{Z}\sum_{\delta_\parallel}\ e^{i\mathbf{k}_\parallel\cdot\delta_\parallel}, \tag{3.7}$$

$$\gamma_{k_x}=\frac{1}{Z}\sum_{\delta_x}\ e^{i\mathbf{k}_x\cdot\delta_x}, \tag{3.8}$$

where $k_\parallel$ is a two-dimensional wave vector parallel to the interfaces and k_x is that along the x direction.

The form of the Hamiltonian (3.6a) is similar to that obtained for the four-sublattice systems,[18] with the exception of the parameters in the Hamiltonians. It is evident that a procedure, similar to that developed in the previous work,[18] is applicable for diagonalizing the Hamiltonian of the present superlattice system. Meanwhile, the form of the Hamiltonian (3.6b) is much simpler than that of the Hamiltonian (2.13) or (2.32). Actually, it can be treated as a special example of the three-sublattice system, i.e., the spin configuration with the angles of $(0,\pi,0)$ for the sublattice moments. The condition of $J_{ab}=J_{bc}<0$ and $J_{ca}>0$ in the three-sublattice systems is the necessary condition for the existence of this spin configuration. Nevertheless, the problem for the superlattice with the Hamiltonian (3.6b) can be solved also by finding out the simplest form of the transformation matrices. The procedure of diagonalizing the Hamiltonians (3.6a) and (3.6b) will be performed in next section.

B. Diagonalizing procedures

We shall perform an extended Bogoliubov transformation[36] for eliminating the nondiagonal terms in the Hamiltonian (3.6). The diagonalizing procedures for the four- and three-layer superlattices will be shown in Secs. III B 1 and III B 2, respectively.

1. Four-layer superlattice

Since the nondiagonal terms in Eq. (3.6a) differ from those in the previous paper,[18] it is reasonable that the transformation matrixes as well as the equation group of the parameters are also different. For the present system, the transformation may be written as the following matrix:

$$\begin{pmatrix}\xi^+_{1,k}\\ \xi^+_{2,k}\\ \xi_{3,k}\\ \xi_{4,k}\end{pmatrix}=\begin{pmatrix}a_{1k}&a_{2k}&a_{3k}&a_{4k}\\ a_{2k}&a_{1k}&a_{4k}&a_{3k}\\ a_{3k}&a_{4k}&a_{1k}&a_{2k}\\ a_{4k}&a_{3k}&a_{2k}&a_{1k}\end{pmatrix}\begin{pmatrix}\mathbf{b}^+_{1,k}\\ \mathbf{b}^+_{2,k}\\ \mathbf{b}_{3,k}\\ \mathbf{b}_{4,k}\end{pmatrix} \tag{3.9a}$$

with its reversed matrix having parameters A_{jk} ($j=1$, 2, 3, and 4) (omitted here for simplicity). The commutation relation of the new operators

$$[\xi_{\tau,k},\xi_{\tau',k'}]=\delta_{\tau\tau'}\delta_{kk'}, \tag{3.10a}$$

leads to one equation:

$$a^2_{1k}+a^2_{2k}-a^2_{3k}-a^2_{4k}=1. \tag{3.11a}$$

It should be noted that in Eqs. (3.9) and (3.10), an index τ is needed for labeling the perpendicular standing component of the spin waves and for diagonalizing the spin-wave Hamiltonian (3.6a). For eliminating the nondiagonal terms of the new operators, one needs to establish the following three equations, respectively,

$$2[ZJ(1-\gamma_{k_\parallel})-Z_xJ'](A_{1k}A_{2k}+A_{3k}A_{4k})$$
$$-Z\gamma_{k_x}[J(A^2_{1k}+A^2_{2k}+A^2_{3k}+A^2_{4k})$$
$$+2J'(A_{1k}A_{3k}+A_{2k}A_{4k})]=0, \tag{3.12a}$$

$$[ZJ(1-\gamma_{k_\parallel})-Z_xJ'](A_{1k}A_{3k}+A_{2k}A_{4k})-Z\gamma_{k_x}[J(A_{1k}A_{4k}$$
$$+A_{2k}A_{3k})+J'(A_{1k}A_{2k}+A_{3k}A_{4k})]=0, \tag{3.13a}$$

$$2[ZJ(1-\gamma_{k_\parallel})-Z_xJ'](A_{1k}A_{4k}+A_{2k}A_{3k})-Z\gamma_{k_x}[2J(A_{1k}A_{3k}$$
$$+A_{2k}A_{4k})+J'(A^2_{1k}+A^2_{2k}+A^2_{3k}+A^2_{4k})]=0, \tag{3.14a}$$

The relations between the parameters a_{ik} and A_{jk} are the same as in Eqs. (27)–(31) in Ref. 18. The problem now becomes solving the equation group of the parameters a_{ik} and A_{jk}, which consists of eight equations and eight unknowns. This equation group (denoted as E_1) is solved by the procedure represented in Appendix C so that the transformation can be carried out.

2. Three-layer superlattice

The transformation matrix for the three-layer superlattice is

$$\begin{pmatrix}\xi^+_{1,k}\\ \xi_{2,k}\\ \xi^+_{3,k}\end{pmatrix}=\begin{pmatrix}b_{1k}&b_{2k}&b_{3k}\\ b_{2k}&b_{4k}&b_{2k}\\ b_{3k}&b_{2k}&b_{1k}\end{pmatrix}\begin{pmatrix}\mathbf{b}^+_{1,k}\\ \mathbf{b}_{2,k}\\ \mathbf{b}^+_{3,k}\end{pmatrix}, \tag{3.9b}$$

with its reversed matrix having parameters B_{jk} ($j=1$, 2, 3, and 4) (omitted here also). Comparing the matrix (3.9b) with the matrix (B1), one has the relations between parameters $a_{1k}=b_{1k}$, $a_{2k}=a_{3k}=a_{6k}=a_{8k}=0$, $a_{4k}=b_{2k}$, $a_{5k}=a_{3k}$, and $a_{7k}=b_{4k}$. The same is true between the parameters A_{jk} and B_{jk}.

The commutation relations of the new operators become

$$b_{1k}^2+b_{3k}^2-b_{2k}^2=1, \tag{3.10b}$$

$$b_{4k}^2-2b_{2k}^2=1. \tag{3.11b}$$

For eliminating the nondiagonal terms, one only needs to establish

$$\begin{aligned}&[ZJ(1-\gamma_{k_\parallel})-Z_xJ']B_{2k}(B_{1k}+B_{3k})\\&+[(Z_{yz}-Z\gamma_{k_\parallel})J-2Z_xJ']B_{2k}B_{4k}\\&-Z\gamma_{k_x}\{J'[2B_{2k}^2+B_{4k}(B_{1k}+B_{3k})]\\&+JB_{2k}(B_{1k}+B_{3k})\}=0,\end{aligned} \tag{3.12b}$$

$$\begin{aligned}&2[ZJ(1-\gamma_{k_\parallel})-Z_xJ']B_{1k}B_{3k}+[(Z_{yz}-Z\gamma_{k_\parallel})J-2Z_xJ']B_{2k}^2\\&-Z\gamma_{k_x}[2J'B_{2k}(B_{1k}+B_{3k})+J(B_{1k}^2+B_{3k}^2)]=0.\end{aligned} \tag{3.13b}$$

The relations between the parameters b_{ik} and B_{jk} ($i=1,2,3,4$; $j=1,2,3,4$) are represented as

$$B_{1k}=\frac{1}{Y}(b_{1k}b_{4k}-b_{2k}^2), \tag{3.14b}$$

$$B_{2k}=\frac{1}{Y}b_{2k}(b_{3k}-b_{1k}), \tag{3.15}$$

$$B_{3k}=\frac{1}{Y}(b_{2k}^2-b_{3k}b_{4k}), \tag{3.16}$$

$$B_{4k}=\frac{1}{Y}(b_{1k}^2-b_{3k}^2). \tag{3.17}$$

Here

$$Y=(b_{1k}-b_{3k})[b_{4k}(b_{1k}+b_{3k})-2b_{2k}^2]. \tag{3.18}$$

Then the problem becomes solving an equation group (denoted as E_2), consisting of Eqs. (3.10b)–(3.18) above, in which eight unknowns are involved. The problem can be solved by the procedures shown in Appendixes D and E.

C. Spin wave spectra

After performing the transformation, one obtains the final form of the Hamiltonian for the present system:

$$\mathbf{H}=\mathbf{H}_0+\mathbf{H}_0'+\mathbf{H}_1, \tag{3.19}$$

where for the four-layer superlattice, one has

$$\mathbf{H}_0=-2NS^2(ZJ+Z_x|J'|), \tag{3.20a}$$

$$\begin{aligned}\mathbf{H}_0'=4S\sum_k\{&[ZJ(1-\gamma_{k_\parallel})+Z_x|J'|](A_{3k}^2+A_{4k}^2)\\&-Z[J(A_{1k}A_{2k}+A_{3k}A_{4k})+J'(A_{1k}A_{4k}+A_{2k}A_{3k})]\gamma_{k_x}\}\end{aligned} \tag{3.21a}$$

and

$$\begin{aligned}H_1=S\sum_k\{&[ZJ(1-\gamma_{k_\parallel})+Z_x|J'|](A_{1k}^2+A_{2k}^2+A_{3k}^2+A_{4k}^2)\\&-2Z[J(A_{1k}A_{2k}+A_{3k}A_{4k})+J'(A_{2k}A_{3k}+A_{4k}A_{1k})]\gamma_{k_x}\}\\&\times[\xi_{1,k}^+\xi_{1,k}+\xi_{2,k}^+\xi_{2,k}+\xi_{3,k}^+\xi_{3,k}+\xi_{4,k}^+\xi_{4,k}]\end{aligned} \tag{3.22a}$$

and for the three-layer superlattice, one has

$$\mathbf{H}_0^0=-NS^2[(Z+\tfrac{1}{2}Z_{yz})J+2Z_x|J'|], \tag{3.20b}$$

$$\begin{aligned}\mathbf{H}_0'=S\sum_k\{&3[(Z_{yz}-Z\gamma_{k_\parallel})J+2Z_x|J'|]B_{2k}^2+Z[2|J'|B_{2k}\\&\times(B_{1k}+B_{3k}+B_{4k})+J(2B_{1k}B_{3k}+B_{2k}^2)]\gamma_{k_x}\}\end{aligned} \tag{3.21b}$$

and

$$\begin{aligned}H_1=S\sum_k(&\{[ZJ(1-\gamma_{k_\parallel})+Z_x|J'|](B_{1k}^2+B_{3k}^2)\\&+[(Z_{yz}-Z\gamma_{k_\parallel})J+2Z_x|J'|]B_{2k}^2\\&+2Z[|J'|B_{2k}(B_{1k}+B_{3k})+JB_{1k}B_{3k}]\gamma_{k_x}\}\\&\times(\xi_{1,k}^+\xi_{1,k}+\xi_{3,k}^+\xi_{3,k})+\{2[ZJ(1-\gamma_{k_\parallel})+Z_x|J'|]B_{2k}^2\\&+[(Z_{yz}-Z\gamma_{k_\parallel})J+2Z_x|J'|]B_{4k}^2+2Z(2|J'|B_{2k}B_{4k}\\&+JB_{2k}^2)\gamma_{k_x}\}\xi_{2,k}^+\xi_{2,k}).\end{aligned} \tag{3.22b}$$

They are the energies for the initial state, the zero-point vibrating, and the spin waves, respectively.

The spin wave spectra of the superlattices, obtained above, depend on the strength of the exchange constants J and J'. For the four-layer superlattice, as shown in Eq. (3.22a), the degeneracy of the spin-wave spectra still remains and the number of the degeneracy of the spin-wave spectra is four. For the three-layer superlattice, however, as shown in Eq. (3.22b), the degeneracy of the spin wave spectra is partially removed and the number of the degeneracy of the spin-

wave spectra is two. This type of splitting of energy levels in the three-layer superlattice are ascribed to the different values of the exchange constants and to the asymmetry of the system.

IV. DISCUSSION AND CONCLUSION

In this work, we have treated the spin-wave theory of several complex Heisenberg Hamiltonians. One of them is for three-sublattice systems with exchange constants $J_{ab}=J_{bc}\neq J_{ca}$. The complication of the present three-sublattice system is ascribed to not only the correlation of the spins of the different sublattices, but also the asymmetry of the system. Without considering the anisotropy and/or the external magnetic field, as discussed in Sec. II B, the classical ground state can be either the collinear or the noncollinear spin structures, depending the strengths and the signs of the exchange constants. For all possible spin configurations, a rotation transformation was applied to deal with the different quantization axes of the three sublattices. In general, this rotation transformation is necessary in the three-sublattice system since the initial state in this system can be in various spin configurations. For eliminating the imaginary terms that appear in the Hamiltonian due to the rotation, it seems that only the noncollinear spin configurations within the basal plane and the collinear configurations are valid upon the quantum effect. Because of the asymmetry of the system, the quantization axes of the spins in different sublattices may vary on their angles with respect to the z axis. This results in the appearance of the linear terms in $\mathbf{a}_0^+$, $\mathbf{a}_0$; $\mathbf{b}_0^+$, $\mathbf{b}_0$; $\mathbf{c}_0^+$, $\mathbf{c}_0$ in the Hamiltonian (2.12) or (2.32), which represent the frozen-in uniform spin deviations. The linear terms can only be eliminated by using three coupled Cullen diagonalization transformations. From the derivatives of Hamiltonian (2.37) with respect to the angles θ_l and ψ_l $(l=a,b,c)$, one may derive the 0 K spin reorientation angles due to the quantum fluctuations. Such uniform $(k=0)$ quantum spin-canting fluctuations have been found also in the one-sublattice[7] and two-sublattice[39] systems with competing sublattice anisotropies. However, the 0 K spin reorientation angle due to the quantum fluctuations does not exist in the systems with high symmetry,[18] but may occur in other multisublattice systems with an odd number of sublattices, or with an even number, together with asymmetry, of different exchange constants.

Generally speaking, it is necessary to perform the three-step procedure developed in Sec. II C. 24 equations and 24 unknowns are involved in the second step of this procedure. However, this step will be simplified (see Appendix B) if the conditions of Eq. (2.22) and $\cos(\theta_a-\theta_b)=\cos(\theta_b-\theta_c)$ are satisfied. In special cases, i.e., the collinear spin configurations, the procedure can be reduced further. Only one step is needed to carry out an extended Bogoliubov transformation and the problem becomes solving an equation group consisting of eight equations and eight unknowns. This equation group can be treated by the procedures, similar to those represented in Appendixes D and E. The uniform $(k=0)$ quantum spin-canting fluctuations do not exist when the initial state is the collinear one.

It is worth noting that for performing the extended Bogoliubov transformation, it is important to select the suitable parameters a_{ik} and A_{jk} (or b_{ik} and B_{jk}) and to establish the equation group with the same values for the numbers of the equations and the unknowns. The form of the transformation matrixes should be chosen to be of physical significance so that the relations between the new and the old operators are meaningful in accordance with the signs of the exchange constants between the different sublattices. Although the new operators for different collinear spin configurations differ, the diagonalized Hamiltonians of these systems can be in the same form with the exception of the zero-point vibrating (i.e., the zero-point quantum) fluctuations. Some different signs appear in the Hamiltonians, which are originated from the differences of the commutation relations of the new operators as well as the signs of the exchange constants.

One of the real systems, corresponding to the present multisublattice system, is of the rare-earth—transition-metal (R-T) intermetallics.[40–43] In some R-T compounds,[40–43] the existence of different rare-earth and transition-metal sites results in the necessity of a multisublattice model for using the spin-wave theory to analyze the spin reorientations.[7] The method, as well as the transformation, developed in the present work could be applied to deal with the spin-wave spectrum of yttrium-iron-garnet (YIG) although the number of the sublattices in YIG may be much larger than three or four.[11,12] Most of the R-T compounds and YIG have large anisotropy for their application potential on permanent magnets. As shown in Sec. II B, the terms of the anisotropy and/or the external field need to be added into Hamiltonian (2.1) to remove the degeneracy of the classical ground state. However, these terms lead to a lot of difficulties, so the diagonalizing procedure cannot be easily performed. For simplicity, in Sec. III we neglected the terms of the anisotropy and the external field. The initial state was assumed to be of the completely ordered state, in which all spins couple parallel or antiparallel along the z axis. Actually, only a little of the anisotropy and/or external field is good enough to start from such an initial collinear state. The corresponding real systems may be R-T compounds with good magnetostrictive properties or CsCl-type ordered compounds or magnetic superlattices.

It is well known that the spin-wave theory of the superlattice can be simplified to be dealt with by a one-dimensional model by taking advantage of the periodic boundary condition on the basal y-z plane and by performing the two-dimensional in-plane Fourier transformation.[23,33] Up to now, the spin waves in the superlattices have been investigated by using a macroscopic Landau-Ginzburg theory,[28] multisublattice Green-function technique,[22,26,29] or a transfer matrix method,[24,25,33,34] which are usually the numerical methods. The present work shows the possibility of studying explicitly the spin-wave spectra of the magnetic superlattices in terms of creation and annihilation operators. The method used for the multisublattice systems (Sec. II in this work and Ref. 18), can be extended to be appropriate for deriving analytical solutions of the spin-wave spectra of the magnetic superlattices.

The spin-wave spectra obtained in the last section depend on the strength of the exchange constants J and J'. The dispersions of the spin waves in the magnetic superlattice are related not only to the two-dimensional wave vector $k_{\|}$ but also the one-dimensional wave vector k_x. Thus they depend

on the values of $\gamma_{k_\parallel}$ and γ_{k_x}. It can be seen from Eq. (3.21) that the quantum fluctuation, i.e., the zero-point vibrating, exists at 0 K in the magnetic superlattice. From the Hamiltonian (3.6) and the canonical transformation described by the matrix (3.9), the spin-wave modes in the superlattices depend on more than the plane wave vector k. An index τ, which is taken to label the perpendicular standing component of the spin waves, is needed for diagonalizing the spin-wave Hamiltonian. In the four-layer superlattice, as shown in Eq. (3.22a), the degeneracy of the spin-wave spectra exists and the number of the degeneracy of the spin-wave spectra is four. For the three-layer superlattice, as shown in Eq. (3.22b), the degeneracy of the spin-wave spectra is partially removed and the number of the degeneracy of the spin-wave spectra is two. This is ascribed to the different values of the exchange constants and the asymmetry of the system.

On the other hand, there occurs another type of splitting of the energy level for the four-layer superlattice. There are four different energy levels which are ascribed to that have positive and negative signs in the formula of K [see Eq. (C4) in Appendix C] and p_k [see Eq. (C9)], and thus a_{ik} [see Eqs. (C17), (C13), and (C9)] and A_{jk}. Therefore, at $T=0$ K, there are four energy levels in the case of the four-layer superlattice; one is the ground state and the others are exciton levels. Consequently, there are four different spin-wave dispersions in the superlattice with the elementary unit of the four different layers. Therefore, the number of the total modes for the spin waves of the four-layer superlattices are 16, i.e., the number of the energy levels times that of their degeneracy. This might be ascribed to the combinatorial number of the possible movements of the spins in the elementary unit.

The same is true for the superlattice with the elementary unit of the three different layers. The total number for the spin-wave modes should equal to 8, i.e., the combinatorial number of the different movements of the spins in the elementary unit of the three layers. The number of the spin-wave spectra of the three-layer superlattice is 3. The splittings of the energy levels should be different for those with or without the degeneracy. The possible situation of the spin-wave modes can be of $2\times1+6$, $2\times2+4$, $2\times3+2$, or $2\times4+0$. One of these possible states is reasonable, realized by evaluating the signs $\pm$ of the parameters b_{ik} in Eqs. (E5), (3.10b), (3.11b), B_{jk} in Eqs. (3.14b)–(3.17), and finally the spin-wave spectra in Eq. (3.22b). One may find, from the results obtained in Sec. III and Appendixes D and E, that the only reasonable state is $2\times2+4$. Namely, the number of the energy splitting is 2 (or 4) for the spin-wave modes with (or without) degeneracy.

In the case of the superlattices, the cyclic condition as well as the translation invariance in the x direction still remain so that the spin-wave spectra in this direction can be described in the form of the plane waves. The superlattice has a larger periodicity in the direction perpendicular to the slabs and therefore many magnon branches in the folded Brillouin zone. Dobrzynski *et al.* found that the surface-localized modes may appear within the extra gaps existing between these folded bulk bands.[22] We argue here that the surface-localized modes can exist only in the limit of the systems with few surfaces or interfaces. In this limit, the effect of the surfaces or interfaces is so weak that it can be treated as a perturbation on the energy of the bulk spin-waves. In the present system as well as those in which the fraction of the interfaces is comparable with that of the bulk, such surface-localized modes cannot be found. This means that the effects of the interfaces and the bulk on the spin-wave spectrum are *mixed* so that the dispersions of the interface and the bulk modes cannot be distinguished.

It was argued[24] that a spin which is not in an interface layer has the same nearest-neighbor environment and therefore the same equation of motion as a spin in the corresponding bulk medium. The spin-wave amplitudes were given within each component by a linear combination of the positive- and negative-going solutions for the bulk medium. For a travelling wave in the superlattice, the wave vectors in the y and z directions must be real whereas those in the x direction are only real when the frequency lies within a pass band for the corresponding bulk mode and can be either imaginary or complex outside the pass band. The results of our present work suggests that such an argument is appropriate only in the limit of few surfaces or interfaces. In the superlattice systems with large fraction of the interfaces, the wave vectors in the x direction can be either imaginary or complex *within* the pass band [see Eqs. (C4) and (C5)]. This is mainly due to the fact that a spin in a superlattice, which is not in an interface layer, *does not* have the same nearest-neighbor environment and the same equation of motion as a spin in the corresponding bulk medium. The quantum correlation between the spins in different layers of a magnetic superlattice results in the complication of the spin-wave spectra of the superlattice.

One of the most important results revealed in this work is that the degeneracy of the splitting of the energy levels of the spin waves can be different, due to the different symmetry of the systems. Since magnetic properties of a real material depend on the elementary excitons, the symmetry may affect the magnetic properties of the material in such a way that they can be quite different for the superlattices with the elementary unit of even and odd layers. It is predicted that even for superlattices having the same exchange constants (either J or J'), i.e., without the oscillation of the exchange coupling, the asymmetry can lead to the oscillation of magnetic properties, such as magnetization and giant magnetoresistence. This is a common fact of the multisublattice systems. Therefore, the same should be true for intermetallic compounds, such as, CsCl-type ordered compounds $Fe_{50}Rh_{50}$,[44,45] rare-earth-based systems $SmMn_2Ge_2$,[46] $NdCu_2$, and $HoCo_2$,[47] which have giant magnetoresistance behavior.

ACKNOWLEDGMENTS

This work has been supported by the National Natural Science Foundation of China, the President Foundation of Chinese Academy of Sciences, and the Science and Technology Commission of Liaoning and Shenyang.

APPENDIX A: PARAMETERS IN HAMILTONIAN (2.41)

The parameters in Hamiltonian (2.41) are

$$\mathbf{H}_0^2=\sum_k \{A_a(A_{2k}^2+A_{4k}^2+A_{6k}^2)+A_b(A_{4k}^2+A_{8k}^2+A_{10k}^2)+A_c(A_{6k}^2+A_{10k}^2+A_{12k}^2)+\gamma_k[C_{ab}(A_{1k}A_{4k}+A_{2k}A_{3k}+A_{3k}A_{8k}$$
$$+A_{4k}A_{7k}+A_{5k}A_{10k}+A_{6k}A_{9k})+D_{ab}(A_{1k}A_{3k}+A_{2k}A_{4k}+A_{3k}A_{7k}+A_{4k}A_{8k}+A_{5k}A_{9k}+A_{6k}A_{10k})+C_{bc}(A_{3k}A_{6k}+A_{4k}A_{5k}$$
$$+A_{7k}A_{10k}+A_{8k}A_{9k}+A_{9k}A_{12k}+A_{10k}A_{11k})+D_{bc}(A_{3k}A_{5k}+A_{4k}A_{6k}+A_{7k}A_{9k}+A_{8k}A_{10k}+A_{9k}A_{11k}+A_{10k}A_{12k})$$
$$+C_{ca}(A_{1k}A_{6k}+A_{2k}A_{5k}+A_{3k}A_{10k}+A_{4k}A_{9k}+A_{5k}A_{12k}+A_{6k}A_{11k})$$
$$+D_{ca}(A_{1k}A_{5k}+A_{2k}A_{6k}+A_{3k}A_{9k}+A_{4k}A_{10k}+A_{5k}A_{11k}+A_{6k}A_{12k})]\}, \tag{A1}$$

$$A_{k1}=A_a(A_{1k}^2+A_{2k}^2)+A_b(A_{3k}^2+A_{4k}^2)+A_c(A_{5k}^2+A_{6k}^2)+2\gamma_k[C_{ab}(A_{1k}A_{4k}+A_{2k}A_{3k})+D_{ab}(A_{1k}A_{3k}+A_{2k}A_{4k})+C_{bc}(A_{3k}A_{6k}$$
$$+A_{4k}A_{5k})+D_{bc}(A_{3k}A_{5k}+A_{4k}A_{6k})+C_{ca}(A_{1k}A_{6k}+A_{2k}A_{5k})+D_{ca}(A_{1k}A_{5k}+A_{2k}A_{6k})], \tag{A2}$$

$$B_{k1}=A_a(A_{3k}^2+A_{4k}^2)+A_b(A_{7k}^2+A_{8k}^2)+A_c(A_{9k}^2+A_{10k}^2)+2\gamma_k[C_{ab}(A_{3k}A_{8k}+A_{4k}A_{7k})+D_{ab}(A_{3k}A_{7k}+A_{4k}A_{8k})+C_{bc}(A_{7k}A_{10k}$$
$$+A_{8k}A_{9k})+D_{bc}(A_{7k}A_{9k}+A_{8k}A_{10k})+C_{ca}(A_{3k}A_{10k}+A_{4k}A_{9k})+D_{ca}(A_{3k}A_{9k}+A_{4k}A_{10k})], \tag{A3}$$

$$C_{k1}=A_a(A_{5k}^2+A_{6k}^2)+A_b(A_{9k}^2+A_{10k}^2)+A_c(A_{11k}^2+A_{12k}^2)+2\gamma_k[C_{ab}(A_{5k}A_{10k}+A_{6k}A_{9k})+D_{ab}(A_{5k}A_{9k}+A_{6k}A_{10k})$$
$$+C_{bc}(A_{9k}A_{12k}+A_{10k}A_{11k})+D_{bc}(A_{9k}A_{11k}+A_{10k}A_{12k})+C_{ca}(A_{5k}A_{12k}+A_{6k}A_{11k})+D_{ca}(A_{5k}A_{11k}+A_{6k}A_{12k})], \tag{A4}$$

$$A_{k2}=A_aA_{1k}A_{2k}+A_bA_{3k}A_{4k}+A_cA_{5k}A_{6k}+\gamma_k[C_{ab}(A_{1k}A_{3k}+A_{2k}A_{4k})+D_{ab}(A_{1k}A_{4k}+A_{2k}A_{3k})+C_{bc}(A_{3k}A_{5k}+A_{4k}A_{6k})$$
$$+D_{bc}(A_{3k}A_{6k}+A_{4k}A_{5k})+C_{ca}(A_{1k}A_{5k}+A_{2k}A_{6k})+D_{ca}(A_{1k}A_{6k}+A_{2k}A_{5k})], \tag{A5}$$

$$B_{k2}=A_aA_{3k}A_{4k}+A_bA_{7k}A_{8k}+A_cA_{9k}A_{10k}+\gamma_k[C_{ab}(A_{3k}A_{7k}+A_{4k}A_{8k})+D_{ab}(A_{3k}A_{8k}+A_{4k}A_{7k})+C_{bc}(A_{7k}A_{9k}+A_{8k}A_{10k})$$
$$+D_{bc}(A_{7k}A_{10k}+A_{8k}A_{9k})+C_{ca}(A_{3k}A_{9k}+A_{4k}A_{10k})+D_{ca}(A_{3k}A_{10k}+A_{4k}A_{9k})], \tag{A6}$$

$$C_{k2}=A_aA_{5k}A_{6k}+A_bA_{9k}A_{10k}+A_cA_{11k}A_{12k}+\gamma_k[C_{ab}(A_{5k}A_{9k}+A_{6k}A_{10k})+D_{ab}(A_{5k}A_{10k}+A_{6k}A_{9k})+C_{bc}(A_{9k}A_{11k}+A_{10k}A_{12k})$$
$$+D_{bc}(A_{9k}A_{12k}+A_{10k}A_{11k})+C_{ca}(A_{5k}A_{11k}+A_{6k}A_{12k})+D_{ca}(A_{5k}A_{12k}+A_{6k}A_{11k})]. \tag{A7}$$

APPENDIX B: THE SECOND STEP OF THE DIAGONALIZING PROCEDURE OF SEC. II C IN CASES OF EQ. (2.22) AND $\cos(\theta_a-\theta_b)=\cos(\theta_b-\theta_c)$

If the conditions of Eq. (2.22) and $\cos(\theta_a-\theta_b)=\cos(\theta_b-\theta_c)$ are satisfied, the second step of the diagonalizing procedure in Sec. II C can be simplified. The matrix (2.38) becomes

$$\begin{pmatrix}\alpha_k^+\\ \alpha_k\\ \beta_k^+\\ \beta_k\\ \xi_k^+\\ \xi_k\end{pmatrix}=\begin{pmatrix}a_{1k}&a_{2k}&a_{3k}&a_{4k}&a_{5k}&a_{6k}\\ a_{2k}&a_{1k}&a_{4k}&a_{3k}&a_{6k}&a_{5k}\\ a_{3k}&a_{4k}&a_{7k}&a_{8k}&a_{3k}&a_{4k}\\ a_{4k}&a_{3k}&a_{8k}&a_{7k}&a_{4k}&a_{3k}\\ a_{5k}&a_{6k}&a_{3k}&a_{4k}&a_{1k}&a_{2k}\\ a_{6k}&a_{5k}&a_{4k}&a_{3k}&a_{2k}&a_{1k}\end{pmatrix}\begin{pmatrix}\mathbf{d}_k^+\\ \mathbf{d}_k\\ \mathbf{f}_k^+\\ \mathbf{f}_k\\ \mathbf{g}_k^+\\ \mathbf{g}_k\end{pmatrix}. \tag{B1}$$

Comparing the matrix (B1) with the matrix (2.38), one has the relations between the parameters $a_{1k}=a_{11k}$, $a_{2k}=a_{12k}$, $a_{3k}=a_{9k}$, and $a_{4k}=a_{10k}$. The same is true for the parameters A_{jk}.

In this case, the relations between the parameters a_{ik} and A_{jk} ($i=1,2,\ldots,8$; $j=1,2,\ldots,8$) give eight equations and the commutation relations of the new operators become four equations. For eliminating the nondiagonal terms, one only needs to establish the other four equations. The problem becomes solving the equation group, consisting of 16 equations and 16 unknowns. Consequently, the parameters defined in Eqs. (A1)–(A7) are simplified as

$$\mathbf{H}_0^2=\sum_k (2A_a(A_{2k}^2+A_{4k}^2+A_{6k}^2)+A_b(2A_{4k}^2+A_{8k}^2)+\gamma_k\{2C_{ab}[A_{3k}(A_{2k}+A_{6k}+A_{8k})+A_{4k}(A_{1k}+A_{5k}+A_{7k})]+2D_{ab}[A_{3k}(A_{1k}$$
$$+A_{5k}+A_{7k})+A_{4k}(A_{2k}+A_{6k}+A_{8k})]+2C_{ca}(A_{1k}A_{6k}+A_{2k}A_{5k}+A_{3k}A_{4k})+D_{ca}(2A_{1k}A_{5k}+2A_{2k}A_{6k}+A_{3k}^2+A_{4k}^2)\}), \tag{B2}$$

$$A_{k1}=C_{k1}=A_a(A_{1k}^2+A_{2k}^2+A_{5k}^2+A_{6k}^2)+A_b(A_{3k}^2+A_{4k}^2)+2\gamma_k[C_{ab}(A_{1k}A_{4k}+A_{2k}A_{3k}+A_{3k}A_{6k}+A_{4k}A_{5k})+D_{bc}(A_{1k}A_{3k}+A_{2k}A_{4k}+A_{3k}A_{5k}+A_{4k}A_{6k})+C_{ca}(A_{1k}A_{6k}+A_{2k}A_{5k})+D_{ca}(A_{1k}A_{5k}+A_{2k}A_{6k})], \tag{B3}$$

$$B_{k1}=2A_a(A_{3k}^2+A_{4k}^2)+A_b(A_{7k}^2+A_{8k}^2)+2\gamma_k[2C_{ab}(A_{3k}A_{8k}+A_{4k}A_{7k})+2D_{ab}(A_{3k}A_{7k}+A_{4k}A_{8k})+2C_{ca}A_{3k}A_{4k}+D_{ca}(A_{3k}^2+A_{4k}^2)], \tag{B4}$$

$$A_{k2}=C_{k2}=A_a(A_{1k}A_{2k}+A_{5k}A_{6k})+A_bA_{3k}A_{4k}+\gamma_k[C_{ab}(A_{1k}A_{3k}+A_{2k}A_{4k}+A_{3k}A_{5k}+A_{4k}A_{6k})+D_{ab}(A_{1k}A_{4k}+A_{2k}A_{3k}+A_{3k}A_{6k}+A_{4k}A_{5k})+C_{ca}(A_{1k}A_{5k}+A_{2k}A_{6k})+D_{ca}(A_{1k}A_{6k}+A_{2k}A_{5k})], \tag{B5}$$

$$B_{k2}=2A_aA_{3k}A_{4k}+A_bA_{7k}A_{8k}+\gamma_k[2C_{ab}(A_{3k}A_{7k}+A_{4k}A_{8k})+2D_{ab}(A_{3k}A_{8k}+A_{4k}A_{7k})+C_{ca}(A_{3k}^2+A_{4k}^2)+2D_{ca}A_{3k}A_{4k}]. \tag{B6}$$

APPENDIX C: PROCEDURE FOR SOLVING THE EQUATION GROUP E_1

Using the relation of Eq. (3.11a), one may rewrite Eqs. (27)–(30) in Ref. 18 and then Eq. (3.12a) minus Eq. (3.14a) results in

$$2a_{1k}^2-2a_{4k}^2+2a_{1k}a_{3k}-2a_{2k}a_{4k}-1=0 \tag{C1}$$

or

$$Z(J-J')\gamma_{k_x}(a_{1k}-a_{3k})^2-2[ZJ(1-\gamma_{k_\parallel})-Z_xJ'](a_{1k}-a_{3k})\times(a_{2k}-a_{4k})+Z(J-J')\gamma_{k_x}(a_{2k}-a_{4k})^2=0. \tag{C2}$$

Equation (C1) is not meaningful since it is independent of the parameters J, J', and γ_k. Eq. (C2) can be reduced to

$$(a_{1k}-a_{3k})=K(a_{2k}-a_{4k}). \tag{C3}$$

Here

$$K=\frac{1\pm\sqrt{1-X^2}}{X}, \tag{C4}$$

with

$$X=\frac{Z(J-J')\gamma_{k_x}}{2[ZJ(1-\gamma_{k_\parallel})-Z_xJ']}. \tag{C5}$$

Equation (3.12a) plus Eq. (3.14a) leads to

$$p_{1k}-p_{2k}=0 \tag{C6}$$

or

$$p_k^2+\frac{2[ZJ(1-\gamma_{k_\parallel})-Z_xJ']+Z(J-J')\gamma_{k_x}^2}{2ZJ\gamma_{k_x}}p_k+\frac{1}{4}=0. \tag{C7}$$

Here one defines the parameter p_k as

$$p_k=a_{1k}a_{2k}-a_{3k}a_{4k}. \tag{C8}$$

Other parameters p_{1k}, p_{2k}, p_{3k}, and q_k, used in this work, are the same as those defined in Eqs. (36)–(38) and Eq. (40) of Ref. 18, respectively.

Equation (C6) should be omitted since it is not meaningful. From Eq. (C7), one has

$$p_k=-\frac{2[ZJ(1-\gamma_{k_\parallel})-Z_xJ']}{4ZJ\gamma_{k_x}}\times[1+X\gamma_{k_x}\pm\sqrt{(1-X^2)(1-\gamma_{k_x}^2)}]. \tag{C9}$$

Then one may derive from Eq. (3.13a) the relation

$$(J-J')p_{3k}Q_{1k}+(Jp_{2k}-J'p_{1k})Q_{2k}=0. \tag{C10}$$

with

$$Q_{1k}=4p_k^2-4Xp_k+1, \tag{C11}$$

$$Q_{2k}=4Xp_k^2-4p_k+X. \tag{C12}$$

Now one is only concerned with a smaller equation group, consisting of Eqs. (3.11a), (C3), (C7) [or (C8) or (C9)] and (C10), since it is not important to deal with the relations between the parameters a_{ik} and A_{jk} at this step.

From Eqs. (3.11a) and (C3), one has

$$a_{1k}=\frac{1}{2K}\left[\frac{1}{a_{2k}-a_{4k}}-(1-K^2)a_{2k}-(1+K^2)a_{4k}\right], \tag{C13}$$

$$a_{3k}=\frac{1}{2K}\left[\frac{1}{a_{2k}-a_{4k}}-(1+K^2)a_{2k}-(1-K^2)a_{4k}\right]. \tag{C14}$$

Inserting Eqs. (C13) and (C14) into Eqs. (C8) and (C10), one respectively obtains two equations in which only a_{2k} and a_{4k} are unknowns:

$$a_{2k}^2-a_{4k}^2=\frac{1-2Kp_k}{1-K^2}=R \tag{C15}$$

and

$$\{Z(J-J')K^4Q_{1k}+4[ZJ(1-\gamma_{k_\parallel})-Z_xJ']K^3Q_{2k}]$$
$$\times(a_{2k}-a_{4k})^4-4Z(J-J')K^2Q_{1k}a_{2k}a_{4k}(a_{2k}-a_{4k})^2$$
$$+Z(J-J')(R-1)[2KQ_{2k}R-Q_{1k}(R-1)\}. \quad \text{(C16)}$$

Setting $a_{2k}=E+F$ and $a_{4k}=-E+F$, one has two equations for the parameters E and F (omitted here for simplicity). After solving these equations, finally, one obtains the solutions of the equation group as

$$a_{1k}=\pm\frac{1+K^2}{2K}\sqrt{1-R}, \quad \text{(C17a)}$$

$$a_{2k}=\pm\frac{1}{2\sqrt{1-R}}, \quad \text{(C17b)}$$

$$a_{3k}=\pm\frac{1-K^2}{2K}\sqrt{1-R}, \quad \text{(C17c)}$$

$$a_{4k}=\pm\frac{2R-1}{2\sqrt{1-R}}. \quad \text{(C17d)}$$

Then A_{jk} $(j=1,2,3,4)$ are given by the relations between the parameters a_{ik} and A_{jk} and the transformation can be performed.

APPENDIX D: PROCEDURE FOR SOLVING THE EQUATION GROUP E_2

Using the relations of Eqs. (3.14b)–(3.17), one may rewrite Eqs. (3.12b) and (3.13b) as

$$[ZJ(1-\gamma_{k_\parallel})-Z_xJ']b_{2k}b_{4k}$$
$$+[(Z_{yz}-Z\gamma_{k_\parallel})J-2Z_xJ']b_{2k}(b_{1k}+b_{3k})$$
$$+Z\gamma_{k_x}\{J'[2b_{2k}^2+b_{4k}(b_{1k}+b_{3k})]-Jb_{2k}b_{4k}\}=0, \quad \text{(D1)}$$

$$2[ZJ(1-\gamma_{k_\parallel})-Z_xJ'](b_{1k}b_{4k}-b_{2k}^2)(b_{2k}^2-b_{3k}b_{4k})$$
$$+[(Z_{yz}-Z\gamma_{k_\parallel})J-2Z_xJ']b_{2k}^2(b_{1k}-b_{3k})^2$$
$$+Z\gamma_{k_x}\{2J'b_{2k}b_{4k}(b_{1k}-b_{3k})^2-J[(b_{1k}b_{4k}-b_{2k}^2)^2$$
$$+(b_{2k}^2-b_{3k}b_{4k})^2]\}=0. \quad \text{(D2)}$$

From Eqs. (3.10b) and (3.11b), one has

$$2b_{1k}^2+2b_{3k}^2-b_{4k}^2=1. \quad \text{(D3)}$$

From Eqs. (3.11b) and (D1), one has

$$b_{2k}=\frac{-ZJ'\gamma_{k_x}[b_{4k}^2-1+b_{4k}(b_{1k}+b_{3k})]}{[(Z_{yz}-Z\gamma_{k_\parallel})J-2Z_xJ'](b_{1k}+b_{3k})+[ZJ(1-\gamma_{k_\parallel}-\gamma_{k_x})-Z_xJ']b_{4k}}. \quad \text{(D4)}$$

Combining Eq. (D4) with Eq. (3.10b) leads to

$$b_{1k}^2+b_{3k}^2-1=\frac{ZJ'\gamma_{k_x}[b_{4k}^2-1+b_{4k}(b_{1k}+b_{3k})]}{[(Z_{yz}-Z\gamma_{k_\parallel})J-2Z_xJ'](b_{1k}+b_{3k})+[ZJ(1-\gamma_{k_\parallel}-\gamma_{k_x})-Z_xJ']b_{4k}} \quad \text{(D5)}$$

which can be reduced by using Eq. (D3) to

$$b_{4k}^2+pb_{4k}+q=0 \quad \text{(D6)}$$

with

$$p=\{4Z^2J'^2\gamma_{k_x}^2-2[(Z_{yz}-Z\gamma_{k_\parallel})J-2Z_xJ']$$
$$\times[ZJ(1-\gamma_{k_\parallel}-\gamma_{k_x})-Z_xJ']\}$$
$$\times\frac{(b_{1k}^2+b_{3k}^2-1)(b_{1k}+b_{3k})}{T}, \quad \text{(D7)}$$

$$q=\{4Z^2J'^2\gamma_{k_x}^2(b_{1k}^2+b_{3k}^2-1)-[(Z_{yz}-Z\gamma_{k_\parallel})J-2Z_xJ']^2$$
$$\times(b_{1k}+b_{3k})^2\}\frac{b_{1k}^2+b_{3k}^2-1}{T}. \quad \text{(D8)}$$

Here

$$T=Z^2J'^2\gamma_{k_\parallel}^2(b_{1k}+b_{3k})^2$$
$$-[ZJ(1-\gamma_{k_\parallel}-\gamma_{k_x})-Z_xJ']^2(b_{1k}^2+b_{3k}^2-1). \quad \text{(D9)}$$

Inserting Eq. (D4) into Eq. (D2) results in

$$b_{4k}^5+rb_{4k}^4+sb_{4k}^3+tb_{4k}^2+ub_{4k}+v=0 \quad \text{(D10)}$$

with

$$r=\frac{[Z_{yz}-Z(2-\gamma_{k_\parallel}-2\gamma_{k_x})]J(b_{1k}+b_{3k})}{ZJ(1-\gamma_{k_\parallel}-\gamma_{k_x})-Z_xJ'}. \quad \text{(D11)}$$

Here the descriptions of the parameters s, t, u, and v are omitted for simplicity.

Equation (D3) can be rewritten as

$$b_{4k}^2=w \quad \text{(D12)}$$

with

$$w=2b_{1k}^2+2b_{3k}^2-1. \quad \text{(D13)}$$

Now the problem can be simplified to solve an equation group (denoted as E_3), consisting of Eqs. (D6), (D10), and (D12) in which only three unknown parameters b_{1k}, b_{3k}, and b_{4k} are involved. The procedure for solving this small equation group E_3 is illustrated in Appendix E.

APPENDIX E: PROCEDURE FOR SOLVING THE EQUATION GROUP E_3

In this appendix, we shall represent the procedure for solving the equation group E_3 which consists of Eqs. (D6), (D10), and (D12).

From Eqs. (D6) and (D12), one derives

$$pb_{4k}+q+w=0. \tag{E1}$$

From Eqs. (D10) and (D12), one obtains

$$(w^2+sw+u)b_{4k}+rw^2+tw+v=0. \tag{E2}$$

Combining Eq. (D12) with Eq. (E1) leads to

$$wp^2-(w+q)^2=0. \tag{E3}$$

Combining Eqs. (E1) and (E2) results in

$$p(rw^2+tw+v)-(w+q)(w^2+sw+u)=0. \tag{E4}$$

The equation group, including Eqs. (E3) and (E4), are the eighth-order equations of two unknowns b_{1k} and b_{3k}, which can be reconstructed to be of the quartic equations of x and y by the definitions of $(b_{1k}+b_{3k})^2=x$ and $(b_{1k}-b_{3k})^2=y$. The quartic equations can be solved in a normal method,[18,48] or a numerical method with the aid of computer. Here we omit the detailed expressions of these lengthy algebraic calculations. Then one obtains

$$b_{1k}=\pm\frac{\sqrt{x}+\sqrt{y}}{2}, \tag{E5a}$$

$$b_{3k}=\pm\frac{\sqrt{x}-\sqrt{y}}{2}, \tag{E5b}$$

or

$$b_{1k}=\pm\frac{\sqrt{x}-\sqrt{y}}{2}, \tag{E5c}$$

$$b_{3k}=\pm\frac{\sqrt{x}+\sqrt{y}}{2}. \tag{E5d}$$

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