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Magnetic properties of three-layer superlattices and three-layer systems

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Abstract
By employing retarded Green functions, the spin-wave spectrum and the layer-sublattice magnetization in Heisenberg ferrimagnetic three-layer superlattices and three-layer systems are calculated within the framework of the linear spin-wave approximation. The effects of the interlayer exchange constants and the intralayer exchange constants on the magnetic properties of the two systems are compared with those for the corresponding three-sublattice bulk ferrimagnets. It is found that all the differences between the magnetic properties of these systems originate from the differences between the exchange couplings in the three dimensions of the systems. The quantum correlations, such as the competition, cancellation, and transmission of the effects of the exchange couplings, are important for the magnetic properties of the systems. The asymmetry of the systems plays an important role in the zero-point quantum fluctuations and, correspondingly, in the layer-sublattice magnetizations of the layers.

1. Introduction

An exciting development in condensed matter physics and materials science is the appearance of an artificial, periodic, layered material structure whose separation between magnetic layers may be precisely controlled to be of the order of the intra-atomic distances. Layered composite materials have become of great interest, since the magnetic properties of these composite materials may be distinctly different from those of their bulk counterparts [1, 2]. In particular, research has been focused on systems such as magnetic superlattices [3–10] and multilayers [11–14]. Furthermore, one of the main

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directions that efforts to achieve an understanding of the mechanism of high-temperature superconductivity have taken is that of investigating two-dimensional magnetic systems and magnetic superlattices [15].

The so-called magnetic superlattices are defined as periodic layered structures with alternating layers having different magnetic and/or electrical properties. Spin waves in the superlattices and the multilayers have their own behaviours, which are different from those in the bulk materials. They have been investigated by use of various quantum microscopic theories [3–10]. By using a Green function method, the spin-wave excitation spectrum and the sublattice magnetizations of a system consisting of ferromagnetic and antiferromagnetic layers are calculated [3]. Herman *et al* [4] investigated the electronic and magnetic structure of ultrathin cobalt–chromium superlattices by the Green function method. Lattice-matched Co/Cr superlattice models were constructed for studying the exchange coupling and spin distributions at atomically abrupt ferromagnetic/antiferromagnetic interfaces. A superlattice consisting of alternating layers of two simple cubic Heisenberg ferromagnets was considered by Albuquerque *et al* [5] who showed that the transfer matrix method leads to a compact expression for the spin-wave dispersion relation of the magnetic superlattice. Most of the systems investigated previously [3–9] are superlattices made up of two kinds of material or two-layer systems. In our previous work [10], the spin-wave spectra of three- and four-layer superlattices were studied analytically by developing a complicated diagonalization procedure in terms of creation and annihilation operators. In our recent work [16], it was found that the results in [10] are valid explicitly only in the trivial limit of $k = 0$ and approximately at the limit of long wavelength. To our knowledge, previous studies have not dealt with the effects of different layer spins on physical properties of the three-layer superlattices with different interlayer and intralayer exchange constants. It is also difficult to study such systems by using the analytical procedure developed previously [10], because of the complexity of the problem.

In the recent work [16], the temperature dependences of the magnetization, internal energy, and specific heat of the three-sublattice systems were studied, by employing retarded Green functions, within the framework of the linear spin-wave approximation. In the present paper, on the basis of a superlattice model developed in [10], we investigate the magnetic properties of superlattices consisting of three different ferromagnetic materials, with layers coupled either ferromagnetically or antiferromagnetically, by using the linear spin-wave theory and the retarded Green function technique. The work is extended also to study the corresponding three-layer system. Our intention is to study the effects of the interlayer and intralayer exchange couplings on the magnetization, to explain the role of competition among these coupling constants, and to compare the magnetic properties of the three-layer superlattices, the corresponding three-layer systems, and the three-sublattice bulk materials.

The structure of the paper is as follows: the model and Hamiltonian are described in section 2. The results of numerical calculations and a discussion are given in section 3. Section 4 gives a summary.

2. Model and Hamiltonian

We consider a Heisenberg model for a three-layer ferrimagnetic superlattice on a simple cubic lattice. A schematic diagram of the model of the three-layer superlattices is given in figure 1(*a*). A unit cell of the superlattice consists of three layers, 1, 2, and 3, where the spins are denoted by S_i ($i = 1, 2, 3$) for each layer. The nearest-neighbouring spins within each layer are coupled ferromagnetically by the intralayer exchange couplings J_i ($i = 1, 2, 3$), respectively. The

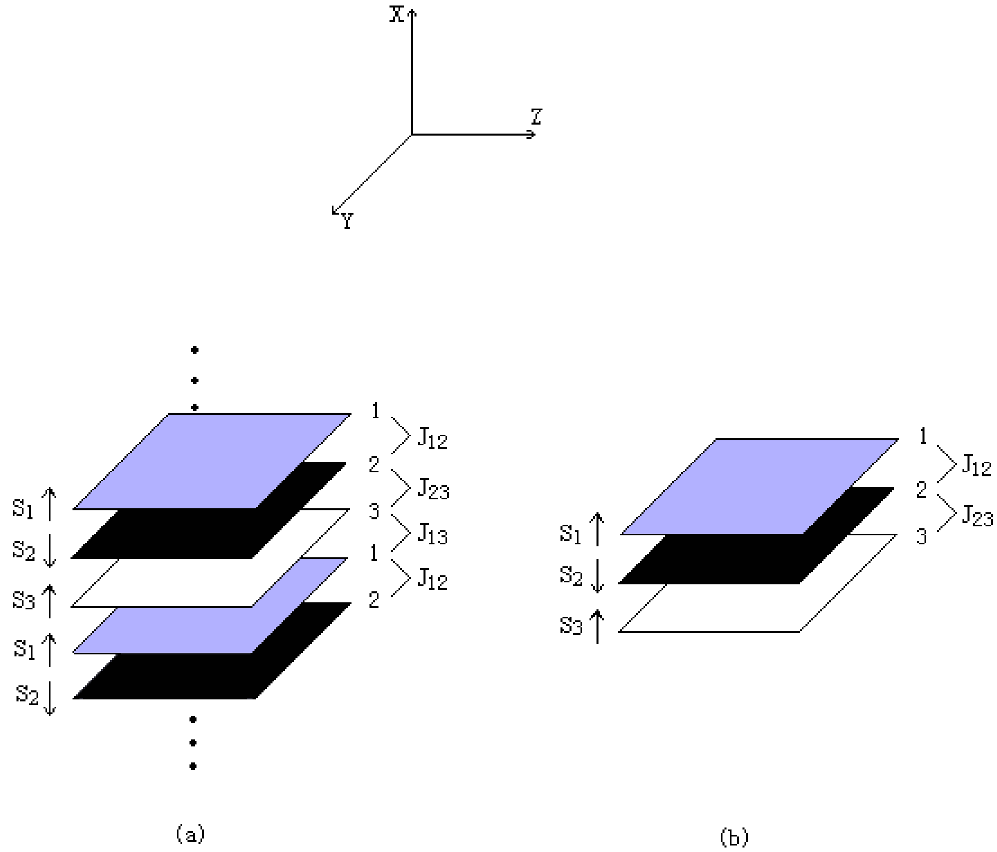

Figure 1. A schematic diagram of the models for (*a*) the three-layer superlattices and (*b*) the quasi-two-dimensional three-layer systems. Only the spin configurations and the interlayer exchange couplings are illustrated.

interlayer exchange couplings J_{12}, J_{23}, and J_{13} between the spins at the nearest-neighbouring layers can be ferromagnetic or antiferromagnetic (see figure 1(*a*)). The superlattice structure is stacked periodically along the x-direction, which is perpendicular to the layers (yz-planes), following [10]. The Hamiltonian is

$$H = -\tfrac{1}{2}\sum_{l=1}^{3}\sum_{\rho,\delta_{\parallel}} J_l S_{l,\rho} S_{l,\rho+\delta_{\parallel}} - \sum_{l=1}^{3}\sum_{\rho} J_{l,l+1} S_{l,\rho} S_{l+1,\rho} \qquad (2.1)$$

where l is the number of layers and δ (or $\delta_{\parallel}$) represents that only the exchanges between the nearest neighbours (or the nearest neighbours parallel to the yz-planes) are taken into account. In the special case of $J_{13} = 0$, we can obtain a three-layer system, as shown in figure 1(*b*). For the three-layer system, there is no periodic stacking along the x-direction; only separate three-layer systems exist. For comparison, the reader could refer to our previous work for a description of a three-sublattice bulk ferrimagnet where the lattice of sites is a simple cubic one and the three interpenetrating sublattices, a, b, and c, are distributed among these sites [10, 16].

By use of the Holstein–Primakoff transform [17] and the linear spin-wave approximation [18, 19], introducing the spin-wave operators b_{lk} (b_{lk}^{+}) ($l = 1, 2, 3$), we rewrite equation (2.1) as follows:

$$\begin{aligned} H = -N\left(\frac{Z_{yz}}{2}\sum_{l=1}^{3} J_l S_l^2 - J_{12}S_1S_2 - J_{23}S_2S_3 + J_{13}S_3S_1\right) \\ + (J_1S_1Z_{yz} - J_{12}S_2 + J_{13}S_3)\sum_k b_{1k}^{+}b_{1k} \\ + (J_2S_2Z_{yz} - J_{12}S_1 - J_{23}S_3)\sum_k b_{2k}^{+}b_{2k} \\ + (J_3S_3Z_{yz} - J_{23}S_2 + J_{13}S_1)\sum_k b_{3k}^{+}b_{3k} \\ - \sqrt{S_1S_2}J_{12}\sum_k (\gamma_{-kx}b_{1k}b_{2k} + \gamma_{kx}b_{1k}^{+}b_{2k}^{+}) \\ - \sqrt{S_2S_3}J_{23}\sum_k (\gamma_{-kx}b_{2k}^{+}b_{3k}^{+} + \gamma_{kx}b_{2k}b_{3k}) \\ - \sqrt{S_3S_1}J_{13}\sum_k (\gamma_{-kx}b_{3k}b_{1k}^{+} + \gamma_{kx}b_{3k}^{+}b_{1k}) \\ - \tfrac{1}{2}Z_{yz}\sum_{l=1}^{3} J_lS_l\sum_k \gamma_{k_\parallel}(2b_{lk}b_{lk}^{+} - 1). \end{aligned} \tag{2.2}$$

Here $Z_{yz} = 4$ represents the number of nearest neighbours in the yz-planes that are the same; the corresponding parameter for along the x-direction is $Z_x = 1$ for the present model. Also,

$$\gamma_{k_\parallel} = \frac{1}{Z_{yz}}\sum_{\delta_\parallel} e^{ik\delta_\parallel} \tag{2.3a}$$

$$\gamma_{\pm k_x} = \sum_{\delta_x} e^{\pm ik\delta_x} \tag{2.3b}$$

and $\gamma_{kx} \neq \gamma_{-kx}$, because the model has no inversion symmetry with respect to each site in the x-direction [16]. γ_{kx} and γ_{-kx} are complex because δ_x has only one value ($\delta_x = 1$), but $\gamma_{k_\parallel}$ is real because $\delta_\parallel$ can take four different values within the layer.

The direction of the spins of the initial state at the 1-layers and 3-layers is along to the positive x-direction, but that of the spins in the 2-layers is the negative x-direction. Therefore, the interlayer exchange constants J_{12}, J_{23} are negative, but J_{13} is positive. All the intralayer exchange constants J_1, J_2, and J_3 are positive. There are N sites on each layer sublattice, making a total of $3N$ sites for the system.

We first define the order-three-matrix retarded Green function:

$$G(k,\omega) = \begin{pmatrix} \langle\langle b_{1k}, b_{1k}^{+}\rangle\rangle_\omega & \langle\langle b_{1k}, b_{2k}\rangle\rangle_\omega & \langle\langle b_{1k}, b_{3k}^{+}\rangle\rangle_\omega \\ \langle\langle b_{2k}^{+}, b_{1k}^{+}\rangle\rangle_\omega & \langle\langle b_{2k}^{+}, b_{2k}\rangle\rangle_\omega & \langle\langle b_{2k}^{+}, b_{3k}^{+}\rangle\rangle_\omega \\ \langle\langle b_{3k}, b_{1k}^{+}\rangle\rangle_\omega & \langle\langle b_{3k}, b_{2k}\rangle\rangle_\omega & \langle\langle b_{3k}, b_{3k}^{+}\rangle\rangle_\omega \end{pmatrix}. \tag{2.4}$$

Using the equation for the Green function:

$$\begin{pmatrix} \omega - H_{11} & H_{12} & H_{13} \\ H_{21} & \omega - H_{22} & H_{23} \\ H_{31} & H_{32} & \omega - H_{33} \end{pmatrix} G(k,\omega) = \begin{pmatrix} 1 & 0 & 0 \\ 0 & -1 & 0 \\ 0 & 0 & 1 \end{pmatrix} \tag{2.5}$$

we obtain the solution of the Green function:

$$G(k,\omega) = \frac{1}{D(\omega)} \begin{pmatrix} M_{11} & -M_{21} & M_{31} \\ M_{12} & -M_{22} & M_{32} \\ M_{13} & -M_{23} & M_{33} \end{pmatrix} \tag{2.6}$$

$$D(\omega) = \begin{vmatrix} \omega - H_{11} & H_{12} & H_{13} \\ H_{21} & \omega - H_{22} & H_{23} \\ H_{31} & H_{32} & \omega - H_{33} \end{vmatrix}. \tag{2.7}$$

Here ω represents the spectrum of the systems. The expressions for the retarded Green function matrix elements M_{ij} $(i, j = 1, 2, 3)$ in equation (2.6) are same as those of the three-sublattice ferrimagnet in [16] and the parameters H_{ij} $(i, j = 1, 2, 3)$ in equations (2.5) and (2.7) are given as follows:

$$H_{11} = J_1 S_1 Z_{yz}(1 - \gamma_{k_\parallel}) - S_2 J_{12} + S_3 J_{13} \tag{2.8a}$$

$$H_{12} = \sqrt{S_1 S_2} J_{12} \gamma_{kx} \tag{2.8b}$$

$$H_{13} = \sqrt{S_3 S_1} J_{13} \gamma_{-kx} \tag{2.8c}$$

$$H_{21} = -\sqrt{S_1 S_2} J_{12} \gamma_{-kx} \tag{2.8d}$$

$$H_{22} = -(S_2 J_2 Z_{yz}(1 - \gamma_{k_\parallel}) - S_1 J_{12} - S_3 J_{23}) \tag{2.8e}$$

$$H_{23} = -\sqrt{S_2 S_3} J_{23} \gamma_{kx} \tag{2.8f}$$

$$H_{31} = \sqrt{S_3 S_1} J_{13} \gamma_{kx} \tag{2.8g}$$

$$H_{32} = \sqrt{S_3 S_2} J_{23} \gamma_{-kx} \tag{2.8h}$$

$$H_{33} = S_3 J_3 Z_{yz}(1 - \gamma_{k_\parallel}) - S_2 J_{23} + S_1 J_{13}. \tag{2.8i}$$

In the following, we shall try to compare the magnetic properties of the three-layer superlattices, the three-layer systems, and also the three-sublattice bulk ferrimagnets.

3. Calculation and discussion

3.1. Spin-wave spectrum

Setting the determinant to zero, i.e., $D(\omega) = 0$, we obtain numerical solutions for the spin-wave spectra of the magnetic three-layer superlattices and three-layer systems. The results are plotted in figures 2(*a*) and (*b*), for $\omega \sim K_x$ and $\omega \sim K_y$, respectively, where $K_x = 3ak_x$, $K_y = ak_y$, and a is the lattice constant. From the two figures, it can be seen that the spin-wave spectra for the three-layer superlattices or the three-layer systems have three branches—equal in number to the layers in a unit cell. One spectrum ω_3 (or ω_3') represents the acoustic branch since $k \to 0$, $\omega \to 0$; the other two branches ω_1 and ω_2 (or ω_1' and ω_2') are optical. The three energy spectra $\omega \sim K_x$ of the superlattices are lower than the corresponding ones in the ferrimagnetic three-sublattice bulk system [16]. This is ascribed to the antiferromagnetic orderings existing along three directions of the three-sublattice system, but only along one direction in the present three-layer superlattices. For the three-layer systems, there is no energy spectrum in the K_x-direction, because of the finite structure along the x-direction of the three-layer systems.

The energy spectra $\omega \sim K_y$ of the three-layer superlattices and the three-layer systems are shown in figure 2(*b*). The solid curves in figure 2(*b*) represent the energy spectra of the superlattices, and the dashed curves are for those of the three-layer systems. The energy ω_1 of

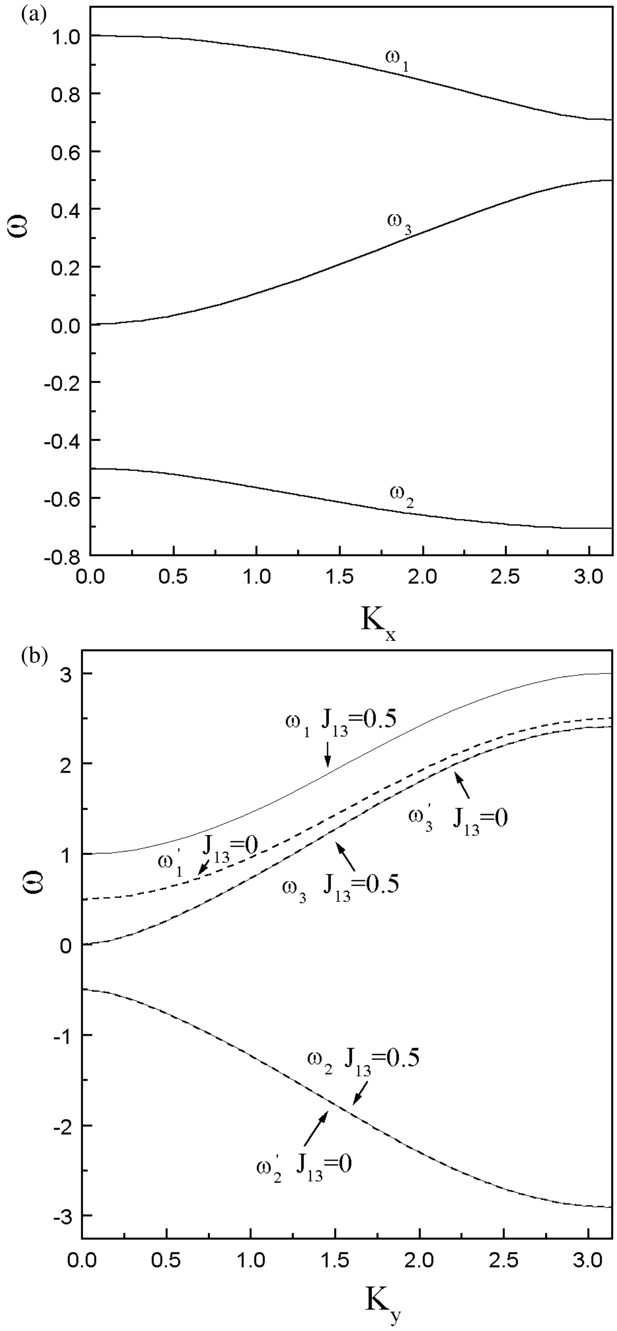

Figure 2. The spin-wave spectrum, ω versus (*a*) K_x and (*b*) K_y, of the three-layer superlattices. Here $K_x = 3ak_x$, $K_y = ak_y$, and a is the lattice constant. The parameters used during the calculation are: $k_z = 0$, $S_1 = S_2 = S_3 = 0.5$, $J_1 = J_2 = J_3 = 1.0$, and $J_{12} = J_{23} = -1.0$. For the calculation of K_x, $k_y = 0$ and $J_{13} = 0.5$. For the calculation of K_y, $k_x = 0$ and $J_{13} = 0.5$ for the superlattices, while $J_{13} = 0.0$ for the three-layer systems. In (*b*), the two spin-wave spectra ω_2 and ω_3 of the three-layer superlattices coincide with the two spin-wave spectra ω_2' and ω_3' of the three-layer systems.

the one optical branch with positive energy for the superlattices is higher than that, ω'_1, of the corresponding one of the three-layer system. The acoustic branch ω_3 of the former coincides with that, ω'_3, of the latter, while another optical branch ω_2 (ω'_2) with negative energy is also the same for the two systems. As explained in [16,20,21], one may consider the magnon vacuum as the ground state. As elementary excitations, the magnons excited out of the filled sea constitute the branches with positive/negative energy [16, 20, 21]. The negative eigenfrequency for the ferrimagnet might be related also to whether the spin wave propagates through clockwise or anticlockwise relative to the spins [16]. The parameters used during the calculation are the same for the two systems, except that J_{13} ($J_{13} \equiv 0$ in the case of the three-layer systems). This implies that the difference in that optical branch is caused by the antiferromagnetism in the two systems, because the increase of J_{13} actually increases the ferromagnetism in the systems. The coincidence of the two branches of the two systems indicates that although J_{13} appears in the solutions for these two branches, its effects may cancel each other out.

It is interesting to compare the energy spectra of the three-layer superlattices and the three-layer systems with those of the corresponding three-sublattice bulk ferrimagnets. The energy spectra in the k_x-, k_y-, k_z-directions of the three-sublattice bulk ferrimagnets are same, because the three crystallographic directions are equivalent. For the three-layer superlattices, the energy spectrum in the k_x-direction is lower than the corresponding one in the ferrimagnetic three-sublattice system. As shown above, it differs from the energy spectra in the k_y- and k_z-directions, due to the different crystallographic properties of the three-layer superlattices in the three directions. In the case of the superlattices, the cyclic condition as well as the translational invariance in the normal direction still hold, so the spin-wave spectra in this direction can be described in the form of the plane waves. The superlattices have a larger periodicity in the x-direction perpendicular to the y–z plane and therefore many magnon branches exist in the folded Brillouin zone. Another reason for the difference of the spin-wave spectra is the different spin alignments along the x-direction, where the spins couple antiferromagnetically/ferromagnetically as in the bulk three-sublattice model, and in the y–z plane, where all spins couple ferromagnetically. Because of the breakdown of the cyclic condition (and the translational invariance) in the limited three-layer systems, no energy spectrum can be calculated for the K_x-direction. Several characteristics of the spin-wave spectra for limited systems, such as thin films and multilayers, were discussed in a recent review article [22]. The discreteness of the spin-wave spectra originates from the finite number of spins along the direction normal to the plane [22]. The gap between the sequence points of the spin-wave spectra can be estimated by π/na (a is the lattice constant and n is the number of spins along the direction perpendicular to the plane). The breaking down of the translation symmetry makes the wavevector not a 'good' quantum number along the normal direction [22]. In the limit case of many magnetic layers, one could still treat the wavevector as a 'pseudo-good' quantum number and deal with the problem approximately using the bulk theory. In the limit case of few layers, like the present three-layer systems, such a concept and the corresponding approximation are no longer valid. The damping effects of spin waves become more pronounced when the number of spins along the normal direction decreases and therefore, according to the quantum uncertainty principle, it is difficult to determine precisely the energies of high-lying (or even low-lying) spin-wave modes [22].

3.2. Low-temperature magnetization

Employing equations (2.6), (2.7) and the spectral theorem, one derives the magnetization per site of each sublattice (the unit is taken to be $g\mu_B$) as represented in equations (3.7), (3.8a), and (3.9) of [16] for the three-sublattice bulk ferrimagnet (S_i and M_i (i = a, b, c) in [16]

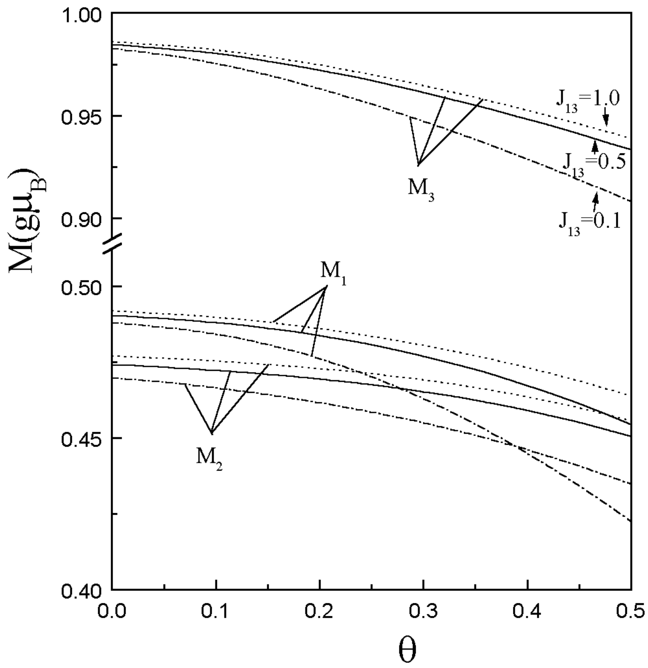

Figure 3. Temperature dependence of the layer-sublattice magnetizations of the three-layer superlattices with $S_1 = S_2 = 0.5$ and $S_3 = 1.0$. The parameters used during the calculation are: $J_1 = J_2 = J_3 = 1.0$, $J_{12} = J_{23} = -1.0$, and $J_{13} = 1.0$ (the dotted curves), 0.5 (the solid curves), 0.1 (the dash–dotted curves). The labels M_1, M_2, and M_3 denote the magnetizations of the 1-, 2-, and 3-layers, respectively.

are replaced by S_i and M_i ($i = 1, 2, 3$) for the superlattices). Temperature ($\theta = k_B T/J_1$) dependences at low temperatures of the layer-sublattice magnetization of the superlattices for different parameters J_{13} are shown in figure 3. For fixed values of the exchange coupling J_{13} and the spins S_i ($i = 1$, 2, and 3), all the magnetizations M_1, M_2, and M_3 for each layer decrease with increasing temperature θ, owing to thermal motion. For fixed values of S_1, S_2, and S_3, the layer-sublattice magnetizations decrease and, correspondingly, the zero-point quantum fluctuations increase with decreasing J_{13}. Although the spin-wave theory is not accurately valid in the high-temperature regime, one can estimate from the curves in figure 3 that the transition temperature will decrease with decreasing J_{13}. These curves confirm that the smaller the exchange coupling J_{13}, the stronger the antiferromagnetism of the system. These results coincide with ones for the three-sublattice bulk ferrimagnets studied previously [16]. However, the zero-point quantum fluctuation of the present superlattices is weaker than that of the three-sublattice bulk ferrimagnets. This is due to the antiferromagnetic coupling existing only along the x-direction in the former case, while the antiferromagnetic ordering exists in three directions in the latter case. On the other hand, the conclusion that there is no magnetic order in the three-layer system at finite temperatures ($T \neq 0$) can be drawn, in good agreement with the well-known results in the literature [23,24]. It is true that the thermal activity destroys the ordering in the two-dimensional isotropic Heisenberg systems.

3.3. Zero-temperature magnetization

The expressions for the layer-sublattice magnetization at zero temperature ($T = 0$ K) of the three-layer superlattices and the three-layer systems are the same as those of equations (3.10)–(3.12) in [16], except for the replacement of S_i and M_{i0} ($i =$ a, b, c) by S_i and M_{i0} ($i = 1, 2, 3$). M_{10}, M_{20} and M_{30} are the zero-temperature layer-sublattice magnetization of the sublattices of 1-layers, 2-layers, and 3-layers of the superlattices or the three-layer systems. In the following, we shall study the effects of the interlayer exchange constants J_{12} (J_{23}), J_{13} and the intralayer exchange constants J_3 (or J_1), J_2 on the properties of the three-layer superlattices and the three-layer systems. For convenience of discussion, the spin configurations and the interlayer exchange constants of the 1-, 2-, and 3-layers of the superlattices and the three-layer systems are illustrated in figures 1(*a*) and (*b*), respectively. Although only the exchange constants between the nearest neighbours have been included in Hamiltonian (2.1), the indirect exchange couplings may contribute to the zero-point quantum fluctuations and also other physical properties of the systems, due to the quantum correlations (see below for details). For simplicity, we consider the contribution of the indirect exchange couplings only up to the next-nearest neighbours in the following discussion.

3.3.1. Effects of the interlayer exchange constants. Figures 4(*a*) and (*b*) respectively show the layer-sublattice magnetization at zero temperature as a function of absolute value of the interlayer exchange constants J_{12} and J_{13} for the three-layer superlattices and the three-layer systems. For the three-layer systems, M_{10} (denoted by 1 in figure 4) has a tendency to decrease with increasing absolute value of the antiferromagnetic coupling J_{12}, corresponding to the enhancement of the zero-point quantum fluctuations in the 1-layer. There is a (very weakly pronounced) maximum value of M_{10} at about $J_{12} = 0.1$ for the three-layer superlattices. Such a maximum becomes obvious for M_{20} (denoted by 2 in figure 4) as the change of J_{12} for the two systems, which corresponds to the minimum of the zero-point quantum fluctuations in the 2-layer. As shown in figure 1, the antiferromagnetic couplings J_{12} and J_{23} act on the spin in the 2-layer for the two systems. The only difference is that the ferromagnetic coupling J_{13} acts indirectly on the 2-layer of the superlattice. Since the crystal structure has no inversion symmetry with respect to the sites of the 2-layer in the x-direction, the ratio between the antiferromagnetic couplings J_{12} and J_{23} can vary over a certain range. The zero-point quantum fluctuations of the 2-layer could be least when the ratio equals a critical value at which the competition among the three exchange couplings is balanced. The zero-point quantum fluctuations of the 2-layer increase gradually when the ratio departs from this value. If one set $|J_{12}| = |J_{23}|$, the zero-point quantum fluctuations of the 2-layer would increase monotonically with increasing absolute value of J_{12} (or J_{23}), in agreement with figure 5 for the three-sublattice bulk ferrimagnets. This indicates clearly that the maximum value of M_{20} as $|J_{12}|$ varies is related to the ratio of the antiferromagnetic couplings J_{12} and J_{23}. The effects of the antiferromagnetic couplings J_{12} and J_{23} of the 2-layer may partially cancel each other. It is also seen from figure 4(*a*) that the increase of $|J_{12}|$ enhances the value of M_{30} (denoted by 3 in figure 4), corresponding to the weak zero-point quantum fluctuations in the 3-layer. This is attributed to the effect of J_{12} on the 3-layer being indirect, through the 1- and 2-layers. It is understood that the antiferromagnetic exchange coupling J_{12} actually cancels out the partial effect of the antiferromagnetic coupling J_{23} on the zero-point quantum fluctuations in the 3-layer. This supports the notion that the antiferromagnetic exchange couplings J_{12} and J_{23} compete with each other when they act on the layers. A similar discussion of the effect of J_{23} could be given, in view of the symmetry of the system. It can be concluded that the zero-point quantum fluctuations and the spin magnetizations in each layer of both the three-layer systems and the three-layer superlattices depend sensitively on the competition among the exchange couplings J_{12}, J_{23}, and J_{13}.

It is interesting to compare the differences between the three-layer systems and the three-layer superlattices. As shown in figure 4(a), the layer-sublattice magnetizations M_{20} and M_{30} at zero temperature of the three-layer systems are smaller than those of the superlattices, regardless of the change of J_{12}. This means that the zero-point quantum fluctuations in the 2- and 3-layers of the three-layer systems are always stronger than the corresponding ones in the three-layer superlattices. As shown in figures 1(a) and (b), the only difference for the 2- or 3-layer is that the ferromagnetic coupling J_{13} acts indirectly on the 2-layer of the superlattices. Thus this suggests that the ferromagnetic coupling J_{13} weakens the zero-point quantum fluctuations indirectly, owing to the quantum correlations. However, the layer-sublattice magnetization M_{10} at zero temperature of the former is less than that of the latter only when the absolute value of J_{12} is bigger than a critical point. That is, only when the effect of J_{12} is large enough can the zero-point quantum fluctuations in the 1-layers of the three-layer systems be stronger than those in the 1-layers of the superlattices. Why does the 1-layer have such special character? Why does such abnormal behaviour occur? This can be understood as follows. The zero-point quantum fluctuations as well as the spin magnetization M_{10} in the 1-layer, for both the three-layer systems and the three-layer superlattices, depend sensitively on the competition among the exchange couplings J_{12}, J_{23}, and J_{13}. It can be seen from figure 1(a) that the antiferromagnetic coupling J_{12} and the ferromagnetic coupling J_{13} act directly on the spin in the 1-layer of the superlattice. Because of the quantum correlations, the antiferromagnetic coupling J_{23} could act indirectly via the ferromagnetic coupling J_{13} on the spin in the 1-layer of the superlattice, especially in the case of weak coupling J_{12}. If the exchange coupling J_{12} vanishes, the 1- and 3-layers are ferromagnetically coupled by J_{13}, which may be treated as a whole as being coupled antiferromagnetically with the 2-layer by J_{23}. Thus the zero-point quantum fluctuations exist in the 1-layer of the superlattices even when $J_{12} = 0$, owing to the correlation between the 1- and 2-layers, via the 3-layer. For the three-layer systems, as shown in figure 1(b), there is only the antiferromagnetic coupling J_{12} acting directly on the spin in the 1-layer, while the effect of the indirect antiferromagnetic coupling J_{23} can be neglected. In the limit case of $J_{12} = 0$, such indirect action of J_{23} on the 1-layer ceases. When the absolute value of J_{12} is very small, the 1-layer in the case of the three-layer systems is equivalent to an isolated ferromagnetic layer and, consequently, its zero-point quantum fluctuation almost vanishes. In fact, when $J_{12} = 0$, there is no zero-point quantum fluctuation in the 1-layer of the three-layer systems, because of its ferromagnetism. When the absolute value of J_{12} is above the critical point, the effect of the antiferromagnetic coupling J_{12} upon the 1-layer is dominant, so the zero-point quantum fluctuations in the 1-layer for the three-layer systems become large—which originates mainly from the direct correlation between the 1- and 2-layers. For the three-layer superlattices, when the absolute value of J_{12} is large, the correlation between the 1- and 3-layers becomes weaker, compared with the correlation between the 1- and 2-layers. It is known from figure 4(b) that the existence of J_{13} decreases the zero-point quantum fluctuations of the three-layer superlattices and the increase of J_{13} enhances all the layer-sublattice magnetizations. These effects result in stronger zero-point quantum fluctuations of the 1-layer of the three-layer systems when the effect of the exchange coupling J_{12} becomes strong. Therefore, the layer-sublattice magnetization M_{10} at zero temperature of the three-layer systems is less than that of the three-layer superlattices when the absolute value of J_{12} is above a critical point.

From the discussion above, one can see that each exchange coupling affects all magnetic properties of every sublattice in the system. With the quantum correlations (such as the

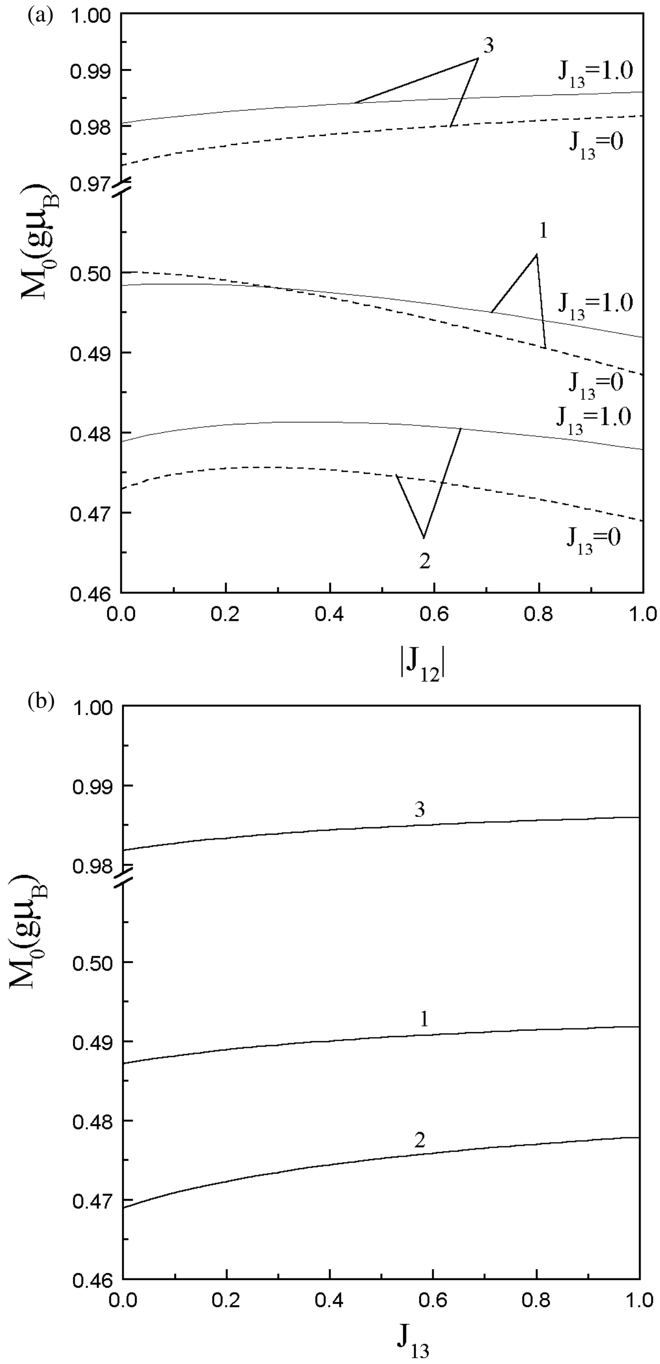

Figure 4. The dependences on (*a*) $|J_{12}|$ and (*b*) J_{13} of the layer-sublattice magnetization M_0 at zero temperature for the three-layer superlattices. The parameters used during the calculation are: $S_1 = S_2 = 0.5$, $S_3 = 1.0$, $J_1 = J_2 = J_3 = 1.0$, and $J_{23} = -1.0$. The labels 1, 2, and 3 correspond to the 1-, 2-, and 3-layers. In (*a*), the solid curves are for an example ($J_{13} = 1.0$) of a three-layer superlattice, while the dashed curves are for the three-layer systems ($J_{13} = 0.0$). For (*b*), $J_{12} = -1.0$. The three points on the ordinate (i.e., $J_{13} = 0$) of (*b*) correspond to the layer-sublattice magnetizations at zero temperature for the three-layer systems.

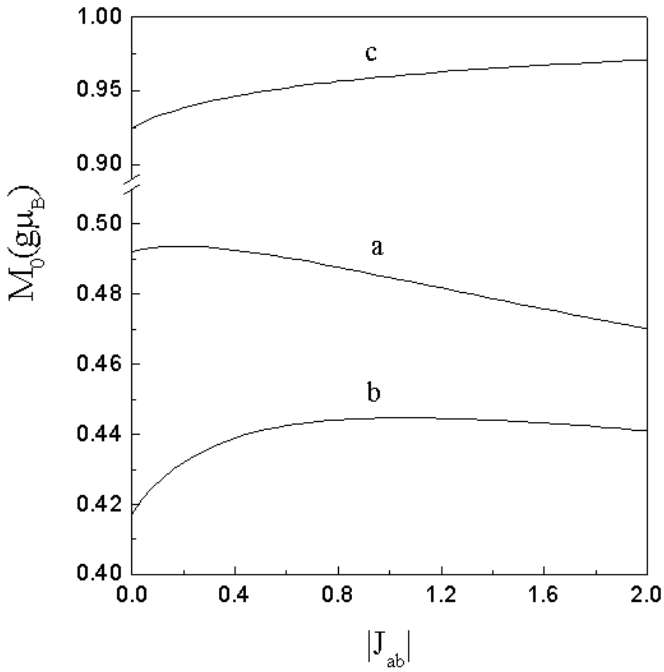

Figure 5. The dependence of the exchange constant $|J_{ab}|$ on the sublattice magnetization at zero temperature in the three-sublattice bulk ferrimagnets as described in [16]. The parameters used during the calculation are: $S_a = S_b = 0.5$, $S_c = 1.0$, $J_{bc} = -1.0$, $J_{ca} = 1.0$. The labels a, b, and c are for sublattices a, b, and c, respectively.

competition, cancellation, and transmission) of the exchange couplings, the system could finally achieve a state with a balance of exchange couplings. Generally, the increase of the ferromagnetic exchange coupling enhances the magnetization of each sublattice in the system, but the role of the antiferromagnetic exchange coupling is very rich. In our previous work [16], we did not study the effects of the quantum correlations in detail for the three-sublattice bulk ferrimagnets. For a better comparison between the superlattices and the bulk materials, we add here the results (in figure 5) for the dependence of the exchange constant $|J_{ab}|$ on the sublattice magnetization at zero temperature in the three-sublattice bulk ferrimagnets (as described in [16]). The trends of the variations in the two systems are similar. The sublattice magnetization of the bulk ferrimagnets changes with the exchange constant more pronouncedly than that in the superlattices. The maxima of magnetization, corresponding to the minima of the zero-point quantum fluctuations, are more pronounced and appear for both sublattices a and b in the bulk ferrimagnets. The values of the exchange constants corresponding to the maxima of magnetization in the bulk ferrimagnets are larger than those in the superlattices. This shows that the quantum competition in the bulk ferrimagnets is stronger than that in the superlattices, because there are more ferromagnetic couplings in the latter. There is no quantum fluctuation in the three-sublattice bulk ferromagnetic systems [16].

Figure 4(*b*) illustrates that, in the case of $J_{12} = J_{23}$, the zero-point quantum fluctuations in the present three-layer superlattices are always weaker than those in the three-layer systems, owing to the introduction of the ferromagnetic coupling J_{13}. If S_3 were set to be same as S_1, the values of the layer-sublattice magnetizations M_{10} and M_{30} would also be the same, due to the symmetry of the system. The difference between the spin values S_1 and S_3 introduces some asymmetry into the system and thus changes the zero-point quantum fluctuations in the two layers. It is noticed, from figure 4(*b*), that the ratio between the layer-sublattice magnetizations M_{10} and M_{30} is not same as that between the spin values S_1 and S_3. As shown in figure 4, the layer-sublattice magnetization M_{20} is always lower than the layer-sublattice magnetization M_{10} (M_{30}) when $S_2 = S_1$ (S_3). This is mainly due to both the interlayer exchange couplings J_{12} and J_{23}, which act directly on the 2-layer, being antiferromagnetic, resulting in the strongest zero-point quantum fluctuations in that layer. Although the effects of the two antiferromagnetic couplings on the 2-layer may cancel partially as indicated above, the zero-point quantum fluctuations in the 2-layer are the strongest. This implies that the negative effects of the ferromagnetic coupling J_{13} on the zero-point quantum fluctuations of the 1- or 3-layer are stronger than those due to the balance between the two antiferromagnetic couplings on the 2-layer. This confirms that the asymmetry of the systems plays an important role in the zero-point quantum fluctuations and correspondingly the layer-sublattice magnetizations of the layers, and that the competition among the three exchange couplings controls the physical properties of the systems. Comparing figure 4(*b*) with figure 5 of [16], one finds that the zero-point quantum fluctuations in the three-layer superlattices and the three-layer systems are weaker than those in the corresponding three-sublattice ferrimagnets, mainly because there is more antiferromagnetic coupling in the bulk case. The change of the zero-point quantum fluctuations with the ferromagnetic coupling J_{13} in the three-layer superlattices is less pronounced than in the bulk, because the ferromagnetic coupling J_{13} exists only in the x-direction of the three-layer superlattices. This means that the effects of the ferromagnetic coupling J_{13} on the zero-point quantum fluctuations in the bulk are stronger than those in the superlattices.

3.3.2. Effects of the intralayer exchange constants. It is evident from figures 6(*a*) and (*b*) that the zero-temperature layer-sublattice magnetization of all the layers increases with the increase of J_2 and J_3, indicating the enhancement of the ferromagnetism of the whole system. Because the 1- and 3-layers are symmetric, the curves in figure 5(*a*) for the two layers coincide with each other. The zero-temperature layer-sublattice magnetization of the 2-layer is lower than that of the 1- and 3-layers, which is the same situation as for the corresponding three-sublattice ferrimagnets [16]. In figure 6(*b*), compared to the changes in the 2- and 3-layers, the variation of the zero-temperature layer-sublattice magnetization in the 1-layer is not very evident, because the increase of the ferromagnetic intralayer coupling J_3 seriously decreases the antiferromagnetism within the 3-layer itself, and also in the 2-layer via the antiferromagnetic interlayer coupling J_{23}. The weakening of the effect of the intralayer coupling J_3 on the 1-layer is mainly due to the containing effects of the ferromagnetic interlayer coupling J_{13} between the 1- and 3-layers. It is noted that the variations in figure 6(*b*) for the 1-layer for the superlattices and the three-layer systems are mostly the same. From figure 6, the magnetization of the layers in the three-layer superlattices is always higher than that of the corresponding layers in the three-layer systems. This is attributed mainly to there being no ferromagnetic interlayer exchange coupling in the latter, but there is one ferromagnetic interlayer exchange coupling in the former. For the bulk three-sublattice ferrimagnet [16], there is no intralayer coupling and the three crystallographic directions are equivalent. As regards symmetry, there occurs symmetric breakdown when one goes from the bulk three-sublattice materials to the three-lattice superlattices and the three-layer systems. Of course, the magnetic properties of the systems depend not only on the symmetry but also on the distributions of the exchange couplings of different types.

It has been concluded that all the differences between the magnetic properties of the three-layer superlattices, the three-layer systems, and the three-sublattice bulk materials originate from the differences in the exchange couplings in the three dimensions of the systems. The

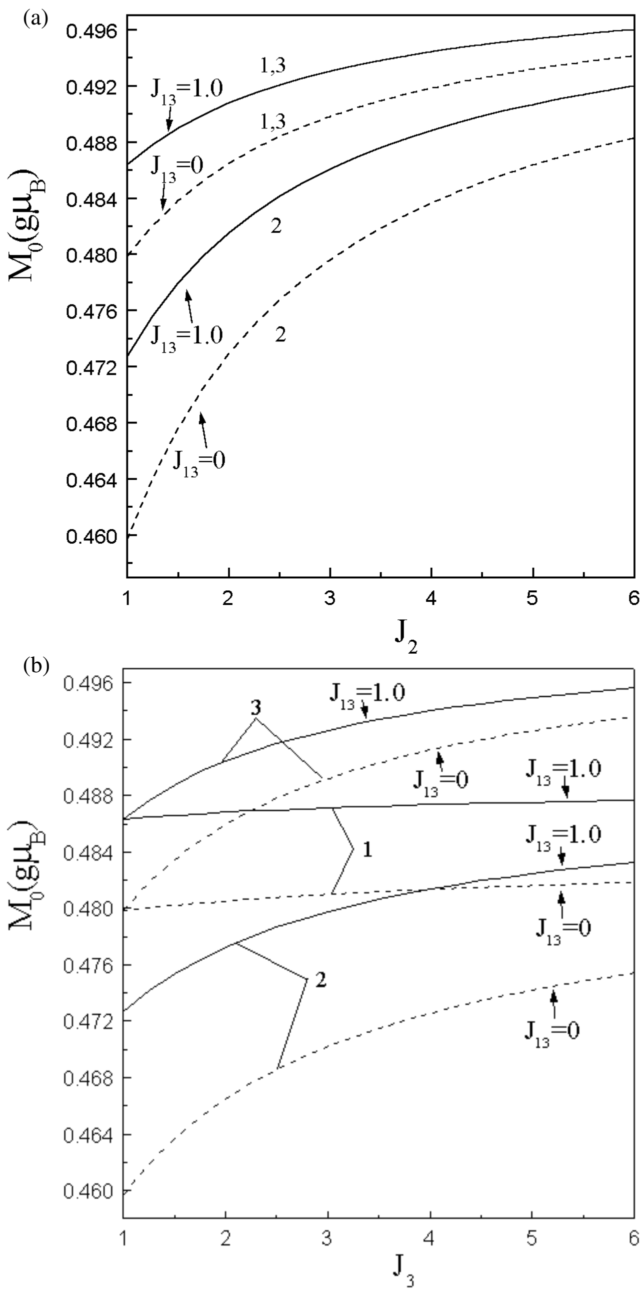

Figure 6. The dependences on (a) J_2 and (b) J_3 of the layer-sublattice magnetization M_0 at zero temperature for the three-layer superlattices (the solid curves, $J_{13} = 1.0$) and the three-layer systems (the dashed curves, $J_{13} = 0.0$). The parameters used during the calculation are: $S_1 = S_2 = S_3 = 0.5$, $J_1 = 1.0$, and $J_{12} = J_{23} = -1.0$. For (a), $J_3 = 1.0$, while for (b), $J_2 = 1.0$. The labels 1, 2, and 3 correspond to the 1-, 2-, and 3-layers.

differences between the three-layer superlattices and the three-sublattice bulk ferrimagnets are as follows. First, as a whole, the superlattice is a periodic layered structure, where only the same ferromagnetic exchange couplings exist within each layer in our model. Therefore the magnetic properties of the sites within one layer in the superlattice are same. But the corresponding three-sublattice bulk ferrimagnets do not possess this type of layered character, and thus the magnetic properties of sites within one two-dimensional plane are not same. This also results in the difference in spin-wave spectrum between the two systems. If there was also antiferromagnetic exchange coupling in each layer in a superlattice, more extensive differences would be found between the superlattices and the bulk. Second, from the magnetic properties of the sublattices in one unit cell, because only ferromagnetic exchange couplings exist within each layer in the three-layer superlattices, each layer may be treated as a whole with a large spin number. Therefore the effects of the change of the exchange coupling parameters on the quantum fluctuations in the superlattices are less pronounced than those in the bulk ferrimagnets.

4. Summary

By using the linear spin-wave method, we have investigated the spin-wave spectrum and the layer-sublattice magnetization for Heisenberg ferrimagnetic three-layer superlattices and three-layer systems. We have discussed the effects of the interlayer exchange constants and the intralayer exchange constants on the magnetic properties of the two systems, in comparison with those of the corresponding three-sublattice ferrimagnet previously studied in [16].

The spin-wave spectra for the three-layer superlattices and the three-layer systems have three branches—equal in number to the layers in a unit cell. One spectrum represents the acoustic branch, the other two the optical branches. Three energy spectra along the K_x-direction for the superlattices are lower than the corresponding ones of the ferrimagnetic three-sublattice system [16], owing to the difference as regards the existence of antiferromagnetic orderings in the various dimensions. The only difference between the spin-wave spectra along the K_y-direction for the superlattice and those for the three-layer systems is for one optical branch with positive energy, which is caused by the difference in the exchange coupling J_{13} between the two systems. The energy spectrum cannot be calculated for the K_x-direction in the limited three-layer systems, because of the breakdown of the cyclic condition (and the translation invariance) along the normal direction. Several characteristics, such as the discreteness, the 'bad' quantum number, and the damping effects of the spin-wave spectra, for the limited systems, originate from the finite spins along the normal direction, as discussed in a recent review article [22].

For fixed values of the exchange coupling J_{13} and the spins S_i ($i = 1, 2,$ and 3), all the layer-sublattice magnetizations of the superlattices decrease with increasing temperature θ, owing to thermal motion. For fixed values of S_1, S_2, and S_3, the layer-sublattice magnetizations decrease and, correspondingly, the zero-point quantum fluctuations increase with decreasing J_{13}. There is no magnetic ordering at finite temperatures for the three-layer isotropic Heisenberg systems.

The zero-point quantum fluctuation for the present three-layer superlattices and three-layer systems is weaker than that for the three-sublattice bulk ferrimagnet studied previously [16], due to the difference in antiferromagnetic couplings in the three directions between these systems. It has been found that the zero-temperature layer-sublattice magnetization for all the layers increases with increasing intralayer exchange coupling J_2 or J_3 (J_1), due to the enhancement of the ferromagnetism of the whole system. The increase of the ferromagnetic interlayer coupling J_{13} enhances (decreases) the zero-temperature layer-sublattice magnetizations (the zero-point quantum fluctuations) of all of the layers of the superlattices. However, the zero-point quantum fluctuations and the layer-sublattice magnetizations in each layer, for both the three-layer systems and the three-layer superlattices, depend sensitively on the competition among the interlayer exchange couplings J_{12}, J_{23}, and J_{13}. In general, the zero-point quantum fluctuations of the three-layer systems are stronger than those of the three-layer superlattices—with the exception of a special case (for M_{10} of the 1-layer). It is found that the quantum correlations, such as the competition, cancellation, and transmission of the effects of the exchange couplings, are important for the magnetic properties of the systems. The indirect effects of the exchange couplings evidently affect the magnetic properties of the non-nearest-neighbouring spins, owing to the quantum correlations. Furthermore, the asymmetry of the systems plays an important role in the zero-point quantum fluctuations and correspondingly the layer-sublattice magnetizations of the layers.

All the differences between the magnetic properties of the three-layer superlattices, the three-layer systems, and the three-sublattice bulk materials originate mainly from the difference in exchange couplings in the three dimensions between the systems.

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