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Magnetization, internal energy and specific heat in a three-sublattice ferrimagnet or ferromagnet with $|J_{ab}| = |J_{bc}| \neq J_{ca}$

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Abstract

The temperature dependences of the magnetization, internal energy and specific heat in a three-sublattice ferrimagnet or ferromagnet with $|J_{ab}| = |J_{bc}| \neq J_{ca}$ are calculated within the framework of the linear spin-wave approximation, by employing retarded Green's functions. For both the ferromagnet and the ferrimagnet, the internal energy and the specific heat decrease with increasing J'/J and/or the value of the spins. For fixed values of S_a, S_b, S_c and J'/J, the internal energy and the specific heat increase, whereas the sublattice magnetization decreases with increasing temperature θ. The three-sublattice ferrimagnet has some particular characteristics which are not shown by the systems with two sublattices. For ferrimagnets, the antiferromagnetism of the system becomes weaker with increasing J'/J. The sublattice magnetization at low temperatures (also the magnetization M_0 at 0 K) of a ferrimagnet increases with increasing J'/J for fixed values of S_a, S_b and S_c. The effects of the spins S_a (S_c) and S_b on the magnetizations of other sublattices differ. The characteristics of the a-sublattice are the same as those of the c-sublattice, due to their similarity as well as the symmetry of the system. The behaviours of the b-sublattice are different from those of the a- and c-sublattices, due to the asymmetry of the three-sublattice system. The spin-value dependences of the spin deviation Δm per spin (and also the energy for the zero-point quantum fluctuation) of the system are different for different sublattices. These differences are ascribed to the asymmetry of the three-sublattice systems, which leads to the new intrinsic properties of the systems.

1. Introduction

Magnetic superlattices have attracted much interest since the discovery of the interlayer exchange interaction between ferromagnetic layers separated by a nonmagnetic spacer [1, 2]

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and of the oscillations of the interlayer exchange coupling [3]. The magnetic properties of these composite materials are distinctly different from those of their bulk counterparts. On the other hand, one of the main directions which 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 [4].

Spin waves are elemental excitons in magnetic materials, from which one can derive the thermodynamic properties, such as magnetization and specific heat, and their dynamical behaviours. A multi-sublattice model, consisting of different sites in a unit cell, was proposed for describing the magnetic properties of rare-earth–transition-metal intermetallic compounds [5]. This model was extended so as to be suitable for interpreting the phenomena in magnetic superlattice systems [6]. The spin-wave spectra of three- and four-sublattice systems and the corresponding superlattices with elementary units of three or four different layers were studied analytically, by developing a complicated diagonalization procedure in terms of creation and annihilation operators [5, 6]. Recently, Pavkov *et al* found that the Bogoliubov and Tyablikov transformation could be used to diagonalize the Hamiltonian of a three- or four-sublattice system analytically in a simpler manner [7]. The elementary excitations and low-temperature behaviour of Heisenberg magnets with three or four sublattices were discussed.

On the other hand, various methods based on Green's function calculations, known as surface Green's function matching [8] and interface response theory [9], have been developed for layered structures. Theoretical Green's function studies of the bulk and surface magnons of a semi-infinite stack of two different ferromagnetic films [10] and two-sublattice ferrimagnets [11, 12] were performed. Hinchey and Mills studied the basic magnetic response characteristics of superlattice structures formed from alternating layers of ferromagnetic and antiferromagnetic materials [13]. The spin-wave spectrum of a system of localized spins interacting by periodically modulated exchange interactions was determined by solving numerically the equations of motion for the magnon Green function [14]. Chen *et al* used the tridiagonal matrix method and the random-phase approximation to obtain a closed form of the Green's functions for two different semi-infinite ferromagnets coupled across an interface [15]. Wei *et al* studied the spin waves of layered Heisenberg ferrimagnets [16] and the disordered ground-state properties of a double-layer Heisenberg antiferromagnet [17]. Wei *et al* also applied the Green's function technique after applying the linear spin-wave approximation to calculate the spin-wave spectra of layered Heisenberg ferrimagnets [16, 18]. Azaria and Diep investigated theoretically magnetic properties, such as spin-wave excitations and phase transitions, of antiferromagnetic superlattices at finite temperatures, using a multi-sublattice Green's function technique [19–21]. Mathon [22] developed a general recursion method for calculating the exact local spin-wave Green's function for arbitrary ferromagnetic interfaces, superlattices and disordered layers.

However, to our knowledge, none of the previous studies have dealt with multi-sublattice systems with both different exchange constants and different spins for different sublattices. It is also difficult to study such systems by using the analytical procedure developed previously [5–7], because of the complexity of the problem. Furthermore, the temperature dependences of the magnetization, internal energy and specific heat for the multi-sublattice or superlattice systems constitute one of the most interesting topics in this field.

In the present work, we shall investigate the temperature dependences of the magnetization, internal energy and specific heat of three-sublattice systems, by employing retarded Green's functions, within the framework of the linear spin-wave approximation. Emphasis will be put on the effect of the sublattice spins and the exchange constants on these physical properties at low temperatures. This work could be extended easily to the more complicated multi-sublattice systems and the corresponding superlattices. The paper is arranged as follows. The model and Hamiltonian are described in section 2. The calculation results and discussion are given in section 3. Section 4 gives a summary. The retarded Green's function matrix elements are given in the appendix.

2. Model and Hamiltonian

The model of a three-sublattice Heisenberg ferrimagnet or ferromagnet with different exchange constants is established following references [5, 6]. A schematic diagram of the three-sublattice model is given as figure 1, where only the spins located on the three crystallographic axes are illustrated. The lattice of sites is a simple cubic one and the three interpenetrating a-, b- and c-sublattices are distributed among these sites. As shown in figure 1, each a-site is surrounded by three b- and c-sites along each direction. The neighbours always belong to different lattices. The a-, b- and c-spins separately form sublattices with the triclinic unit cell.

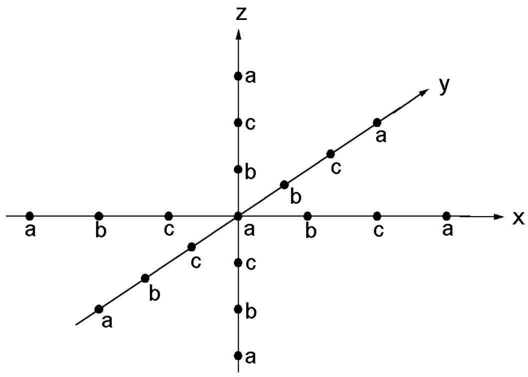

Figure 1. A schematic diagram of the three-sublattice model. Only the spins located on the three crystallographic axes are illustrated.

We take $i \in a$-sublattice, $j \in b$-sublattice and $m \in c$-sublattice, and the spins are $\vec{S}_a$, $\vec{S}_b$ and $\vec{S}_c$, respectively. There are N sites on each sublattice: a total of $3N$ sites for the system. The Hamiltonian is

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

where δ represents the fact that only the exchanges between nearest neighbours are taken into account. J_{ab}, J_{bc} and J_{ca} are the exchange constants for neighbouring sites.

In the case of a ferrimagnet, the direction of the spins of the initial states at a- and c-sites is along the positive z-axis, but that of spins at b-sites is along the negative z-axis. Therefore, the exchange constants J_{ab}, J_{bc} are negative, but J_{ca} is positive. To simplify, we assume $|J_{ab}| = |J_{bc}| \neq J_{ca}$, $|J_{ab}| = |J_{bc}| = J$ and $J_{ca} = J'$.

In the initial state of a ferromagnet, the directions of spins at the a-, b- and c-sites are all positive (along the z-axis), the values of J_{ab}, J_{bc} and J_{ca} also positive, and $J_{ab} = J_{bc} \neq J_{ca}$; we take $J_{ab} = J_{bc} = J$, $J_{ca} = J'$.

By use of the Holstein–Primakoff transform [23] and the linear spin-wave approximation [24, 25], introducing the spin-wave operators a_k (a_k^+), b_k (b_k^+) and c_k (c_k^+), we can rewrite equation (2.1) as follows:

$$\begin{aligned}H = {} & -NZ(S_aS_bJ + S_bS_cJ + S_cS_aJ') + Z(S_bJ + S_cJ')\sum_k a_k^+a_k \\ & + Z(S_a + S_c)J\sum_k b_k^+b_k + Z(S_bJ + S_aJ')\sum_k c_k^+c_k \\ & + \sqrt{S_aS_b}JZ\sum_k(\gamma_{-k}a_kb_k + \gamma_ka_k^+b_k^+) + \sqrt{S_bS_C}JZ\sum_k(\gamma_{-k}b_k^+c_k^+ + \gamma_kb_kc_k) \\ & - \sqrt{S_cS_a}J'Z\sum_k(\gamma_{-k}c_ka_k^+ + \gamma_kc_k^+a_k)\end{aligned} \tag{2.2a}$$

and

$$\begin{aligned}H = {} & -NZ(S_aS_bJ + S_bS_cJ + S_cS_aJ') + Z(S_bJ + S_cJ')\sum_k a_k^+a_k \\ & + Z(S_a + S_c)J\sum_k b_k^+b_k + Z(S_bJ + S_aJ')\sum_k c_k^+c_k \\ & - \sqrt{S_aS_b}JZ\sum_k(\gamma_{-k}a_kb_k^+ + \gamma_ka_k^+b_k) - \sqrt{S_bS_C}JZ\sum_k(\gamma_{-k}b_kc_k^+ + \gamma_kb_k^+c_k) \\ & - \sqrt{S_cS_a}J'Z\sum_k(\gamma_{-k}c_ka_k^+ + \gamma_kc_k^+a_k)\end{aligned} \tag{2.2b}$$

for the ferrimagnet and the ferromagnet, respectively.

Here $Z = 3$ represents the number of same-type nearest neighbours:

$$\gamma_{\pm k} = \frac{1}{Z}\sum_\delta \mathrm{e}^{\pm \mathrm{i}k\delta} \tag{2.3}$$

and $\gamma_k \neq \gamma_{-k}$, because the model has no inversion symmetry with respect to each site. γ_k and γ_{-k} are complex; the real coefficients in reference [6] were due to erroneous site summation. In fact, the Hamiltonians in references [5, 6] are valid explicitly only in the trivial limit of $k = 0$ (in this case, $\gamma_k = \gamma_{-k}$), and may be applicable at the limit of the long-wavelength approximation. It is hard to reach an analytical solution for the Hamiltonian (2.2*a*) or (2.2*b*). In the following, we try to employ the technique of retarded Green's functions to study the spin waves and the physical properties at zero and low temperatures. It is expected that the method used in this paper would be easily extended to other more complex multi-sublattice systems.

3. Calculation and discussion

We first define the third-order-matrix retarded Green's function:

$$G(k,\omega) = \begin{pmatrix} \langle\langle a_k, a_k^+\rangle\rangle_\omega & \langle\langle a_k, b_k\rangle\rangle_\omega & \langle\langle a_k, c_k^+\rangle\rangle_\omega \\ \langle\langle b_k^+, a_k^+\rangle\rangle_\omega & \langle\langle b_k^+, b_k\rangle\rangle_\omega & \langle\langle b_k^+, c_k^+\rangle\rangle_\omega \\ \langle\langle c_k, a_k^+\rangle\rangle_\omega & \langle\langle c_k, b_k\rangle\rangle_\omega & \langle\langle c_k, c_k^+\rangle\rangle_\omega \end{pmatrix} \tag{3.1a}$$

and

$$G(k,\omega) = \begin{pmatrix} \langle\langle a_k, a_k^+\rangle\rangle_\omega & \langle\langle a_k, b_k^+\rangle\rangle_\omega & \langle\langle a_k, c_k^+\rangle\rangle_\omega \\ \langle\langle b_k, a_k^+\rangle\rangle_\omega & \langle\langle b_k, b_k^+\rangle\rangle_\omega & \langle\langle b_k, c_k^+\rangle\rangle_\omega \\ \langle\langle c_k, a_k^+\rangle\rangle_\omega & \langle\langle c_k, b_k^+\rangle\rangle_\omega & \langle\langle c_k, c_k^+\rangle\rangle_\omega \end{pmatrix} \tag{3.1b}$$

for the ferrimagnet and the ferromagnet, respectively. By using the equation for the Green's function, we obtain the solution for the Green's function as follows:

$$G(k,\omega) = \frac{1}{D(\omega)}\begin{pmatrix} M_{11} & M_{21} & M_{31} \\ M_{12} & M_{22} & M_{32} \\ M_{13} & M_{23} & M_{33} \end{pmatrix}\begin{pmatrix} 1 & 0 & 0 \\ 0 & \mp 1 & 0 \\ 0 & 0 & 1 \end{pmatrix}. \tag{3.2}$$

The upper/lower sign above corresponds to the case of a ferrimagnet/ferromagnet. The retarded Green's function matrix elements in equations (3.2) are given in the appendix. In equation (3.2),

$$D(\omega) = \begin{vmatrix} \omega - H_{11} & \pm H_{12} & H_{13} \\ H_{21} & \omega \mp H_{22} & H_{23} \\ H_{31} & \pm H_{32} & \omega - H_{33} \end{vmatrix}. \tag{3.3}$$

Here ω represents the spectrum of systems. The upper/lower sign in equation (3.3) is for the ferrimagnet/ferromagnet, respectively. The parameters H_{ij} $(i, j = 1, 2, 3)$ are given in the appendix also.

3.1. Spin-wave spectrum

For vanishing of the determinant, i.e., $D(\omega) = 0$, we obtain the spectrum expressions as follows:

$$\begin{aligned} \omega_1 &= 2\sqrt{-\frac{p}{3}}\cos\frac{\psi}{3} \\ \omega_2 &= -\sqrt{-\frac{p}{3}}\cos\frac{\psi}{3} - \sqrt{3}\sqrt{-\frac{p}{3}}\sin\frac{\psi}{3} \\ \omega_3 &= -\sqrt{-\frac{p}{3}}\cos\frac{\psi}{3} + \sqrt{3}\sqrt{-\frac{p}{3}}\sin\frac{\psi}{3} \end{aligned} \tag{3.4}$$

where

$$\begin{aligned} p &= -\frac{b_1^2}{3} + b_2 \\ \psi &= \arctan\frac{\sqrt{-s}}{-q/2} \\ q &= \frac{2}{27}b_1^3 - \frac{1}{3}b_1 b_2 + b_3 \\ s &= \left(\frac{q}{2}\right)^2 + \left(\frac{p}{3}\right)^3 \end{aligned} \tag{3.5}$$

with

$$\begin{aligned} b_1 &= -(H_{11} \pm H_{22} + H_{33}) \\ b_2 &= \pm H_{11}H_{22} + H_{11}H_{33} \pm H_{22}H_{33} - H_{13}H_{31} \mp H_{32}H_{23} \mp H_{12}H_{21} \\ b_3 &= \pm H_{11}H_{23}H_{32} \pm H_{22}H_{13}H_{31} \pm H_{33}H_{12}H_{21} \pm H_{21}H_{32}H_{13} \pm H_{12}H_{23}H_{31} \mp H_{11}H_{22}H_{33}. \end{aligned} \tag{3.6}$$

The upper/lower sign in equation (3.6) is for the ferrimagnet/ferromagnet, respectively.

$s < 0$ is required because ω_l $(l = 1, 2, 3)$ are real numbers. The spin-wave spectra calculated numerically are shown in figures 2 and 3 for the ferrimagnet and the ferromagnet, respectively. From the two figures, the spin-wave spectra for the ferrimagnet and ferromagnet each have three branches, one for each sublattice. For the ferrimagnet, there is one branch with negative energy ω_2. As explained in references [11, 12], 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 the positive/negative energy [11, 12]. The negative eigenfrequency for the ferrimagnet might be related also to whether the spin-wave propagates clockwise or anticlockwise relative to the spins S_a, S_b and S_c. ω_3 represents the acoustic branch since $k_x \to 0$, $\omega_3 \to 0$, while ω_2 and ω_1 represent the optical branches. There are three positive

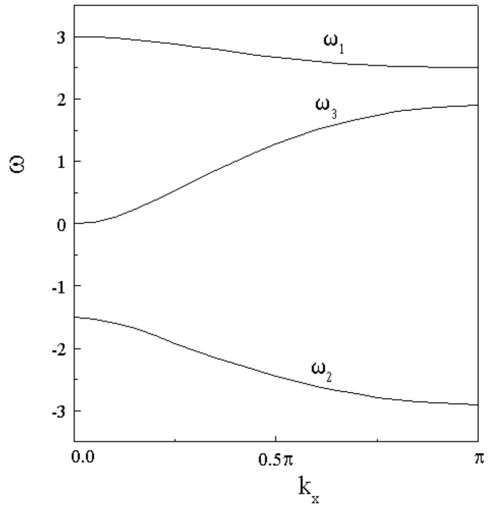

Figure 2. The spin-wave spectrum, ω versus k_x, with $k_y = k_z = 0$, $S_a = S_b = S_c = 0.5$ and $J'/J = 0.5$, for three-sublattice ferrimagnets.

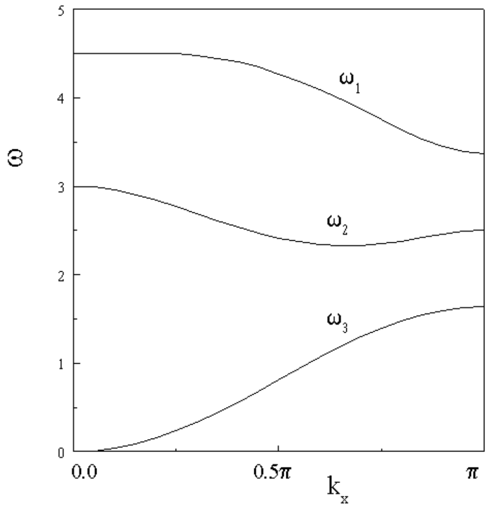

Figure 3. The spin-wave spectrum, ω versus k_x, with $k_y = k_z = 0$, $S_a = S_b = S_c = 0.5$ and $J'/J = 0.5$, for three-sublattice ferromagnets.

energies ω_l ($l = 1, 2, 3$) for spin-wave spectra of the ferromagnet, among which ω_3 is that for the acoustic branch and ω_1, ω_2 are those for the optical branches. When $S_a = S_b = S_c$ and $J = J'$, the spin-wave spectrum of the three-sublattice Heisenberg ferrimagnet does not reduce to that of the Heisenberg antiferromagnet.

3.2. Magnetization

After employing the spectral theorem, we finally derive the magnetization per site for each sublattice (the unit is taken to be $g\mu_B$) from

$$M_a = S_a - \frac{1}{N}\sum_k \sum_{l=1}^{3} \frac{M_{11}(\omega_l)}{(\mathrm{e}^{\beta\omega_l} - 1)\prod_{m\neq l}(\omega_l - \omega_m)} \tag{3.7}$$

$$M_b = -S_b - 1 + \frac{1}{N}\sum_k \sum_{l=1}^{3} \frac{-M_{22}(\omega_l)}{(\mathrm{e}^{\beta\omega_l} - 1)\prod_{m\neq l}(\omega_l - \omega_m)} \tag{3.8a}$$

$$M_b = S_b - \frac{1}{N}\sum_k \sum_{l=1}^{3} \frac{M_{22}(\omega_l)}{(\mathrm{e}^{\beta\omega_l} - 1)\prod_{m\neq l}(\omega_l - \omega_m)} \tag{3.8b}$$

$$M_c = S_c - \frac{1}{N}\sum_k \sum_{l=1}^{3} \frac{M_{33}(\omega_l)}{(\mathrm{e}^{\beta\omega_l} - 1)\prod_{m\neq l}(\omega_l - \omega_m)} \tag{3.9}$$

for the ferrimagnet and the ferromagnet, respectively; here $\beta = 1/k_BT$.

From equations (3.7)–(3.9), one can obtain the magnetization at zero temperature ($T = 0$ K) of the ferrimagnet as follows:

$$M_{a0} = S_a + \frac{1}{N}\sum_k \frac{M_{11}(\omega_2)}{(\omega_2 - \omega_1)(\omega_2 - \omega_3)} \tag{3.10}$$

$$M_{b0} = -S_b - 1 + \frac{1}{N}\sum_k \frac{M_{22}(\omega_2)}{(\omega_2 - \omega_1)(\omega_2 - \omega_3)} \tag{3.11}$$

$$M_{c0} = S_c + \frac{1}{N}\sum_k \frac{M_{33}(\omega_2)}{(\omega_2 - \omega_1)(\omega_2 - \omega_3)} \tag{3.12}$$

where M_{a0}, M_{b0} and M_{c0} are the zero-temperature magnetizations of the a-, b- and c-sublattices of the ferrimagnet, respectively.

By solving numerically equations (3.7)–(3.9) and (3.10)–(3.12) for different parameters J'/J, temperature ($\theta = k_B T$) dependences of the sublattice magnetization of the ferrimagnet are derived; these are shown in figure 4. The sublattice magnetizations at zero temperature are smaller than their classical values, owing to the zero-point quantum fluctuations of the spins (see equations (3.15a) and (3.16) below). For fixed values of S_a, S_b and S_c, the zero-point quantum fluctuations decrease and consequently the sublattice magnetizations at zero temperature increase with increasing J'/J. Figure 5 shows the dependence on J'/J of the sublattice magnetization at zero temperature for the ferrimagnet. It is evident that the sublattice magnetization increases with increase of J'/J, corresponding to the enhancement of the ferromagnetism. The values of the zero-point quantum fluctuations for the a- and c-sublattices are the same only if $S_a = S_c$. Even if $S_a = S_b$ (or $S_c = S_b$), the zero-point quantum fluctuation of the a-sublattice (or c-sublattice) differs from that of the b-sublattice. In any case, if $S_a = S_b$ (or $S_c = S_b$), the zero-point quantum fluctuation of the b-sublattice is always stronger than that of the a-sublattice (or c-sublattice) and, correspondingly, the magnetization at 0 K of the former is always weaker than that of the latter. For fixed values of J'/J and S_a, S_b, S_c, all of the sublattice magnetizations decrease with increasing temperature θ (figure 4).

Figures 6 and 7 represent the dependence of the magnetization at zero temperature for each sublattice on the spins S_b and S_a, respectively. It is seen from figure 6 that the magnetizations M_{a0} and M_{c0} first decrease with increasing spin S_b, reach a minimum at $S_b = S_a + S_c$ and then increase. This is attributed to the fact that the stronger the antiferromagnetism, the stronger the zero-point quantum fluctuation and thus the smaller the magnetization. The spins S_a and S_c, which are ferromagnetically coupled, may be treated as being coupled as a whole antiferromagnetically with the spin S_b. When the sum of the spins S_a and S_c is equal to the spin S_b, the effect of the b-sublattice on the a- and c-sublattices due to the antiferromagnetic coupling is strongest. It is also shown in figure 6 that in case of $S_a = S_c$, the magnetizations M_{a0} and M_{c0} are always the same. This means that the a- and c-sublattices are symmetric in the system and have the same behaviours, owing to the condition of $J_{ab} = J_{bc}$. From figure 7, the magnetization M_{c0} monotonically increases with increasing S_a, because the spins S_a and S_c couple with each other ferromagnetically. The correlation between the a- and c-sublattices behaves ferromagnetically. The magnetization M_{b0} varies with the change of the spin S_a, depending on the difference between S_b and the sum of S_a and S_c. The magnetization M_{b0} exhibits its minimum when the condition $S_b = S_a + S_c$ is satisfied. For $S_b = 1.5$ and $S_c = 0.5$, the minimum is at $S_a = 1.0$. For $S_b = 1.0$ and $S_c = 1.0$, the minimum should be at $S_a = 0$ and thus the magnetization M_{b0} increases monotonically with increasing S_a. Even if $S_c = S_b$, as shown in figure 7, the magnetization M_{b0} differs from the magnetization M_{c0}. These behaviours are attributed to the fact that the three-sublattice system is asymmetric. This is different to the situation in two-sublattice ferrimagnets [16]. This indicates that the symmetry of the multi-sublattice systems strongly affects the magnetic properties of the system.

From equations (3.7)–(3.9), the sublattice magnetizations at zero temperature of ferromagnets are same as the classical values, indicating that there is no zero-point quantum fluctuation in the three-sublattice ferromagnet. The sublattice magnetization decreases monotonically with increasing temperature as expected. The change of the exchange constants and/or of the spin strength weakly affects the low-temperature behaviours of the magnetization. This is ascribed to the fact that all of the spins in the system have ferromagnetic exchange couplings and there is no competition among these exchange couplings.

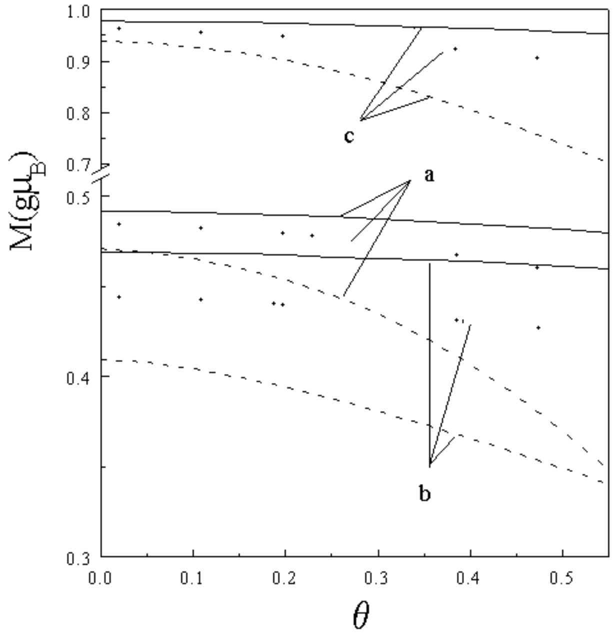

Figure 4. Temperature dependences of the sublattice magnetization M of the three-sublattice ferrimagnets with $S_a = S_b = 0.5$ and $S_c = 1.0$. The dashed, dotted and solid curves represent the values of $J'/J = 0.1$, 1.0 and 4.0, respectively. The labels a, b and c denote the magnetization of the a-sublattice, b-sublattice and c-sublattice, respectively.

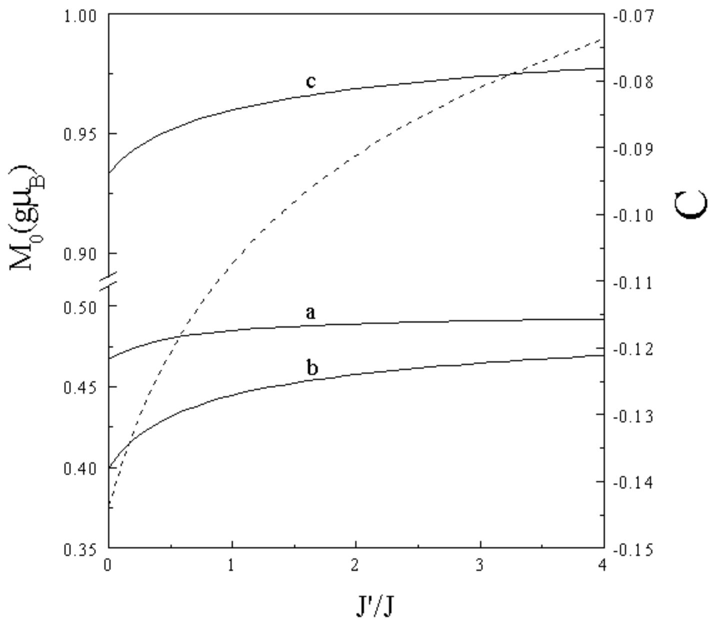

Figure 5. The dependences on J'/J of the magnetization M_0 (solid curves) at zero temperature and the energy C (dashed curve) of the zero-point quantum fluctuation for the three-sublattice ferrimagnets with $S_a = S_b = 0.5$ and $S_c = 1.0$. The curves a, b and c correspond to the a-sublattice, b-sublattice and c-sublattice, respectively.

3.3. Internal energy

From equations (2.2*a*), (2.2*b*), (3.1*a*), (3.1*b*), (3.2) and applying the spectral theorem, the internal energies per site are described as follows:

$$U = \frac{1}{3N}\langle H\rangle = -[(S_a + S_c)(S_b + 1)J + S_a S_c J'] + \frac{1}{N}\sum_k \sum_{l=1}^{3} \frac{A}{(\mathrm{e}^{\beta\omega_l} - 1)\prod_{m\neq l}(\omega_l - \omega_m)} \tag{3.13a}$$

and

$$U = \frac{1}{3N}\langle H\rangle = -[(S_a + S_c)S_b J + S_a S_c J'] + \frac{1}{N}\sum_k \sum_{l=1}^{3} \frac{A}{(\mathrm{e}^{\beta\omega_l} - 1)\prod_{m\neq l}(\omega_l - \omega_m)} \tag{3.13b}$$

for the ferrimagnet and the ferromagnet, respectively. Here

$$A = (S_b J + S_c J')M_{11}(\omega_l) \mp (S_a + S_c)J M_{22}(\omega_l) + (S_b J + S_a J')M_{33}(\omega_l)$$
$$\pm \sqrt{S_a S_b}\, J(\gamma_k M_{12}(\omega_l) \mp \gamma_{-k} M_{21}(\omega_l)) \pm \sqrt{S_b S_c}\, J(\gamma_{-k} M_{32}(\omega_l) \mp \gamma_k M_{23}(\omega_l))$$

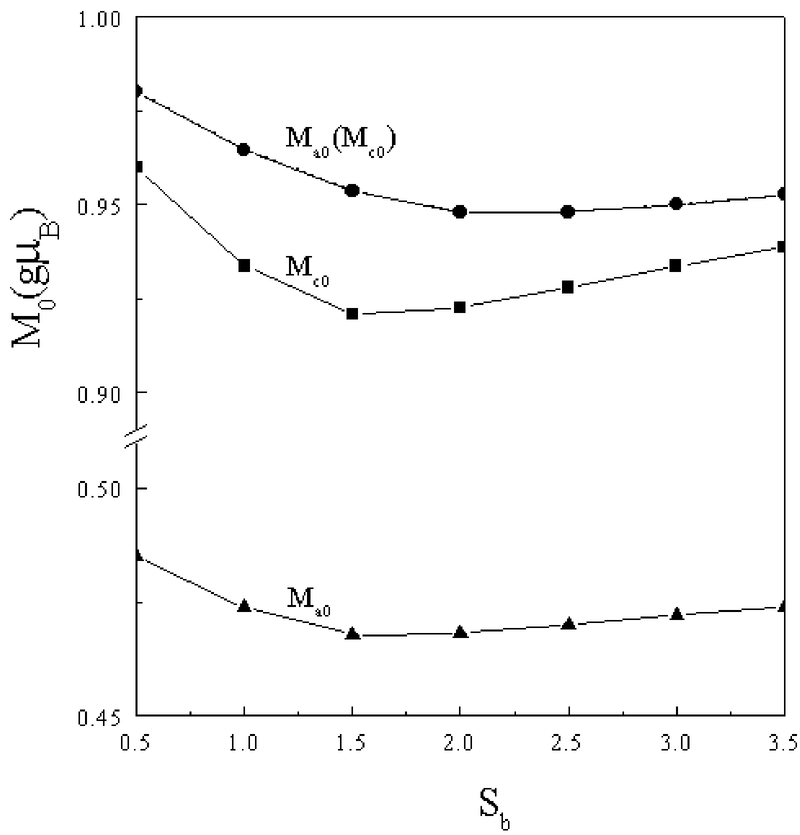

Figure 6. Dependences of the magnetizations M_{a0} and M_{c0} at zero temperature on the spin S_b for the three-sublattice ferrimagnets. The curves with the solid squares and the upward-pointing triangles correspond to $S_a = 0.5$, $S_c = 1.0$ and $J'/J = 1.0$. The curve with the circles is for $S_a = 1.0$, $S_c = 1.0$ and $J'/J = 1.0$.

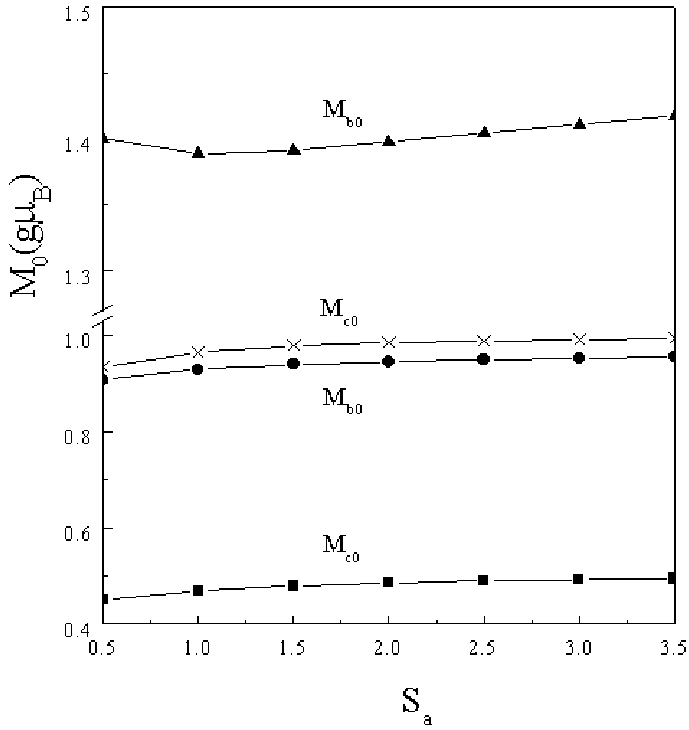

Figure 7. Dependences of magnetizations M_{b0} and M_{c0} at zero temperature on the spin S_a for the three-sublattice ferrimagnets. The curves with the solid squares and the upward-pointing triangles correspond to $S_b = 1.5$, $S_c = 0.5$ and $J'/J = 1.0$. The curves with the circles and the crosses are for $S_b = 1.0$, $S_c = 1.0$ and $J'/J = 1.0$.

$$- \sqrt{S_c S_a} J'(\gamma_{-k} M_{13}(\omega_l) + \gamma_k M_{31}(\omega_l)) \tag{3.14}$$

where the upper/lower sign above corresponds to the case of the ferrimagnet/ferromagnet. At $T = 0$ K, the ground-state energy per site of the ferrimagnet is obtained from equations (3.13*a*) and (3.14) as

$$U_0 = -[(S_a + S_c)S_b J + S_a S_c J'] + C \tag{3.15a}$$

$$C = -(S_a + S_c)J - \frac{1}{N}\sum_k \frac{A(\omega_2)}{(\omega_2 - \omega_1)(\omega_2 - \omega_3)}. \tag{3.16}$$

Here C is the energy for zero-point quantum fluctuation of the ferrimagnet.

For the ferromagnet, from equations (3.13*b*) and (3.14), one has

$$U_0 = -[(S_a + S_c)S_b J + S_a S_c J']. \tag{3.15b}$$

From equations (3.15) and (3.16), it is seen that the ferrimagnet with three sublattices has the energy of the zero-point quantum fluctuations, but the ferromagnet does not. From equation (3.15), increase of the spins S_a, S_b and S_c decreases the internal energy U_0 at zero temperature. The energy C for zero-point quantum fluctuations is illustrated as a function of J'/J (dashed line in figure 5). The absolute value of the energy C decreases with increasing J'/J, indicating that the effect of zero-point quantum fluctuations is weakened. This means that increasing J'/J decreases the antiferromagnetism of the system. This is consistent with the results in figure 5 for the magnetization at zero temperature.

In order to obtain a clearer understanding of the zero-point quantum fluctuations in the three-sublattice ferrimagnet, we calculate the spin deviation Δm per spin of the system as follows:

$$\Delta m = \frac{\Delta M_a}{S_a} + \frac{\Delta M_b}{S_b} + \frac{\Delta M_c}{S_c} \tag{3.17}$$

where ΔM_i $(i = a, b, c)$ is the averaged magnetization deviation at zero temperature for each sublattice; Δm represents the amplitude of the averaged deviation angle of the spins in the system. Figure 8 represents the dependence of the spin deviation Δm on the spin S_a (S_b). The $\Delta m \sim S_b$ curve exhibits a maximum, indicating that at that point, the deviation angle of the spin is largest and the antiferromagnetism of the whole system is strongest. The value of Δm decreases monotonically with increasing S_a, no matter whether $S_b < S_c$ or $S_b > S_c$. The larger the spin S_a, the weaker the antiferromagnetism of the system. For comparison, the dependence on the spin S_a (S_b) of the energy C for the zero-point quantum fluctuations is shown in figure 9. The energy C decreases monotonically with the increase of S_b. When $S_b < S_c$, the energy C hardly changes with change of the value of S_a, but still exhibits a maximum. When $S_b > S_c$, the energy C decreases monotonically with increasing S_a. The energy C for the zero-point quantum fluctuations depends not only on the averaged deviation angle of the spin, but also on the values of all of the spins S_a, S_b and S_c.

We numerically derive the temperature dependence of the internal energy from equations (3.13a), (3.13b) and (3.14). The results are illustrated in figure 10 as dashed and solid lines, respectively, for the ferrimagnet and the ferromagnet. For the same value of J'/J, the internal energy of the ferrimagnet is smaller than that of the ferromagnet at fixed temperature. For both the ferrimagnet and the ferromagnet, the internal energy U increases with increasing temperature θ for fixed values of J'/J, S_a, S_b and S_c. It decreases with increasing J'/J for fixed values of S_a, S_b, S_c and θ. From the discussion above, larger J'/J corresponds to stronger ferromagnetism. Why does the internal energy U of the ferrimagnet not increase with increasing J'/J? We try to explain this as follows. The absolute value of the initial energy is much larger than that of the energy C for the zero-point quantum fluctuations. Therefore, the modification to the internal energy U, owing to the energy C for the zero-point quantum fluctuations, is negligible, compared with that of the initial energy. Although the absolute value of the energy C for the zero-point quantum fluctuations decreases with increasing J'/J, the internal energy U increases with increasing J'/J because, from equation (3.15a), the absolute value of the initial energy is enhanced by increasing J'/J.

3.4. Specific heat

The specific heat at low temperatures for the ferrimagnet and the ferromagnet is derived as

$$C_v = \frac{\partial U}{\partial \theta} = \frac{1}{N}\sum_k \sum_{l=1}^{3} \frac{A\omega_l}{(\mathrm{e}^{\frac{1}{2}\beta\omega_l} - \mathrm{e}^{-\frac{1}{2}\beta\omega_l})^2\theta^2 \prod_{m\neq l}(\omega_l - \omega_m)}. \tag{3.18}$$

The parameter A was defined by equation (3.14). From equations (3.18), the temperature dependence of the specific heat was calculated numerically for various values of J'/J and spin. The results for $J'/J = 0.1$, 1.0, 2.0 are plotted in figures 11 and 12 for the ferrimagnet and the ferromagnet, respectively. For both the ferrimagnet and the ferromagnet, it is shown that the specific heat increases with increasing temperature θ for fixed values of J'/J and S_i $(i = a, b, c)$. The specific heat for both the ferrimagnet and the ferromagnet decreases with increasing J'/J and/or the spin S_i $(i = a, b, c)$.

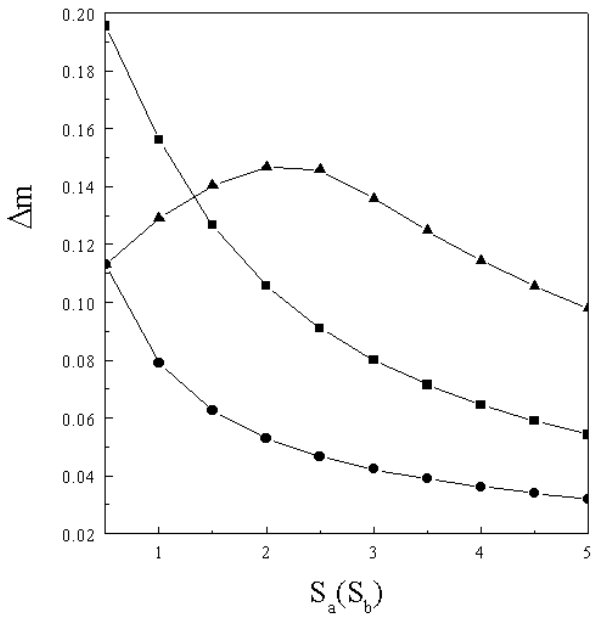

Figure 8. Δm versus S_a (S_b) for the three-sublattice ferrimagnets with $J'/J = 1.0$. The curve with the upward-pointing triangles represents $\Delta m \sim S_b$ with $S_a = 0.5$ and $S_c = 2.0$. The curve with the circles is for $\Delta m \sim S_a$ with $S_b = 0.5$ and $S_c = 2.0$, while the one with the squares corresponds to $\Delta m \sim S_a$ with $S_b = 2.0$ and $S_c = 1.0$.

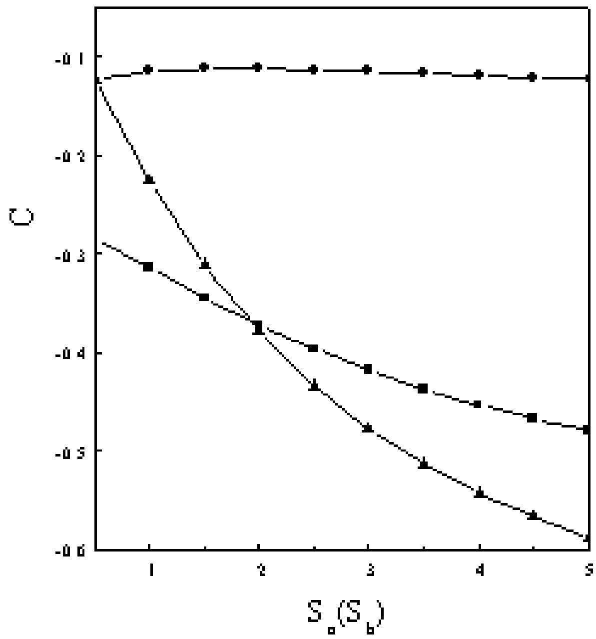

Figure 9. The dependence of the energy C of zero-point quantum fluctuation on the spin S_a (S_b) for the three-sublattice ferrimagnets with $J'/J = 1.0$. The curve with the upward-pointing triangles represents the condition with $S_a = 0.5$ and $S_c = 2.0$. The curve with the circles is for the case of $S_b = 0.5$ and $S_c = 2.0$, while the one with the squares corresponds to the case of $S_b = 2.0$ and $S_c = 1.0$.

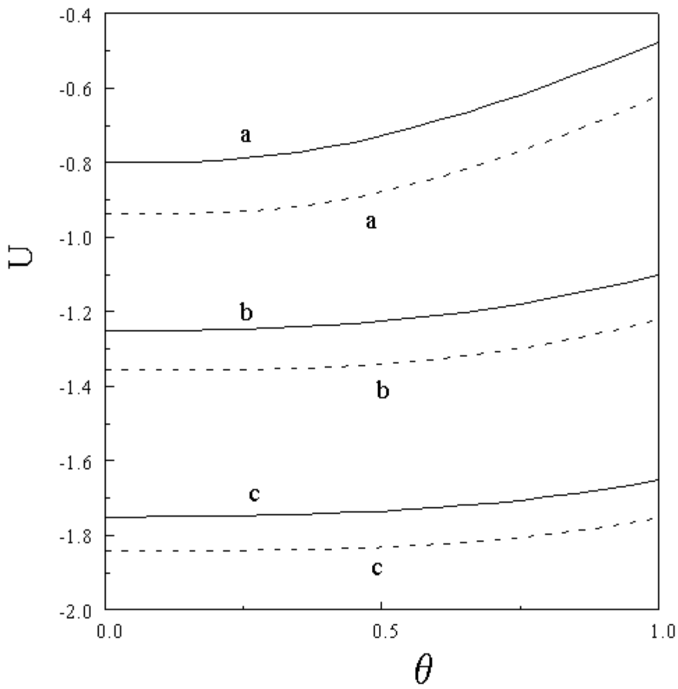

Figure 10. Temperature dependences of the internal energy U for the three-sublattice systems with $S_a = 1.0$ and $S_b = S_c = 0.5$. The curves a, b and c correspond to $J'/J = 0.1$, 1.0 and 2.0, respectively. The dashed curves are for ferrimagnets, while the solid curves are for ferromagnets.

4. Summary

In this paper, the linear spin-wave method has been applied to a three-sublattice Heisenberg ferrimagnetic or ferromagnetic system with different exchange constants ($|J_{ab}| = |J_{bc}| \neq J_{ca}$). By using the retarded Green's function technique, we have investigated the spin-wave spectrum, the sublattice magnetization, the internal energy and the specific heat for the ferrimagnet and the ferromagnet.

For the three-sublattice systems, there are some behaviours that are the same as those of the two-sublattice systems. For instance, there is zero-point quantum fluctuation at zero temperature in the ferrimagnet, but not in the ferromagnet. The sublattice magnetization decreases with increasing temperature θ for fixed values of S_a, S_b, S_c and J'/J for both the ferromagnet and the ferrimagnet. The internal energy and the specific heat increase with temperature, but decrease with increasing the values of the spins.

However, the three-sublattice ferrimagnet has some particular characteristics which are not shown by the two-sublattice systems. For example, the sublattices with different original alignments have different zero-point quantum fluctuations, even when the spins of the sublattices are same. The effects of the spins S_a (S_c) and S_b on the magnetizations of other sublattices differ. Since the a- and c-sublattices couple ferromagnetically, the larger the value of the spin S_a (S_c), the larger the interaction between these two sublattices. The characteristics of the a-sublattice are the same as those of the c-sublattice, due to their similarity as well as the symmetry of the system. On the other hand, the behaviours of the b-sublattice are different from those of the a- and c-sublattices, which is mainly attributed to the asymmetry of the three-sublattice system. Because the b-sublattice couples with the a- and c-sublattices

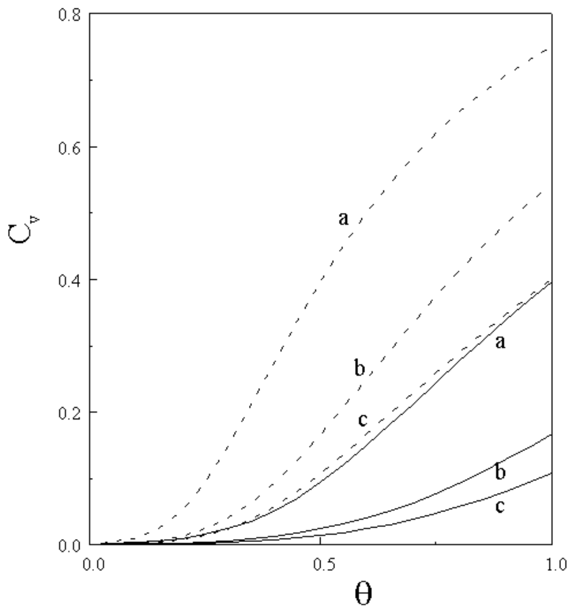

Figure 11. Temperature dependences of the specific heat C_v for the three-sublattice ferrimagnets. The curves a, b and c correspond to $J'/J = 0.1$, 1.0 and 2.0, respectively. The dashed curves correspond to $S_a = S_b = S_c = 0.5$, while the solid curves are for $S_a = S_b = S_c = 1.0$.

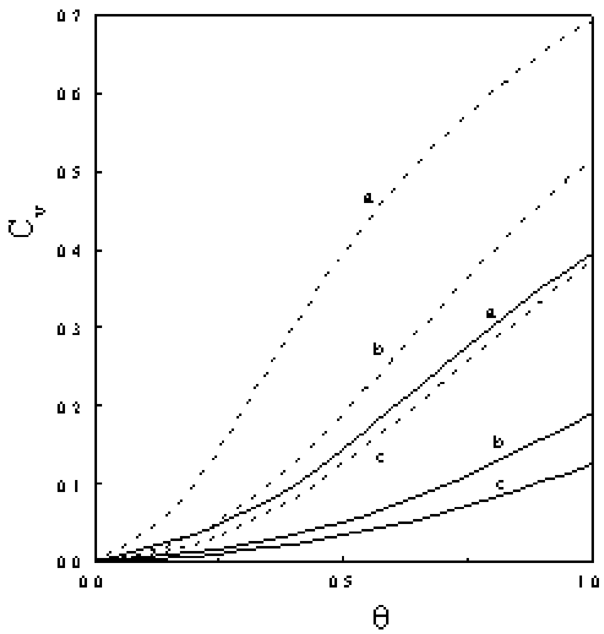

Figure 12. Temperature dependences of the specific heat C_v for the three-sublattice ferromagnets. The curves a, b and c correspond to $J'/J = 0.1$, 1.0 and 2.0, respectively. The dashed curves correspond to $S_a = S_b = S_c = 0.5$, while the solid curves are for $S_a = S_b = S_c = 1.0$.

antiferromagnetically, the effect of the b-sublattice on the a- and c-sublattices is strongest when $S_b = S_a + S_c$ and, at the same time, the strongest effect of the a-sublattice (c-sublattice) on the b-sublattice is found. Furthermore, the spin-value dependences of the spin deviation Δm per spin (and also the energy for the zero-point quantum fluctuation) of the system are different for different sublattices. These differences are ascribed to the asymmetry of the three-sublattice systems, which leads to the new intrinsic properties of the systems.

For a ferrimagnet, the antiferromagnetism of the system becomes weaker with increasing J'/J, and the coefficient J'/J represents the competition between ferromagnetism and antiferromagnetism of the system for the three-sublattice ferrimagnet. The sublattice magnetization at low temperatures (and also the magnetization M_0 at 0 K) increases with increasing J'/J for fixed S_a, S_b and S_c. That is, the magnetization deviation as well as the absolute value of the energy for the spin zero-point quantum fluctuation decrease with increasing J'/J for fixed S_a, S_b and S_c. For both the ferromagnet and the ferrimagnet, the internal energy and the specific heat decrease with increasing J'/J. For the ferromagnet, the coefficient J'/J represents only the ratio of the exchange coupling between the a-sublattices and c-sublattices, with respect to that between the b-sublattices and a-sublattices (c-sublattices), which can also evidently affect the properties of the systems. The sublattice magnetization at low temperatures of the ferromagnet increases with increasing J'/J for fixed values of θ, S_a, S_b and S_c, but the magnetization M_0 at 0 K is just equal to the value of the spin for each sublattice. Our results show that the coefficient J'/J plays an important role in the magnetic properties of the ferrimagnet and the ferromagnet.

The conclusions obtained in this work can be extended immediately to the corresponding superlattice system with the elementary unit of three different layers. This is because the superlattice is periodic not only in the y- and z-directions, but also in the x-direction with a larger periodicity [6]. Thus the spin-wave theory of the superlattice can be simplified so that it can be dealt with using 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 [6, 13, 26]. The method in this work can be applied also to other multi-sublattice systems and superlattice Heisenberg systems.

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Appendix

The retarded Green's function matrix elements in equations (3.2) are as follows:

$$
\begin{aligned}
M_{11} &= (\omega \mp H_{22})(\omega - H_{33}) \mp H_{23}H_{32} \\
M_{22} &= (\omega - H_{11})(\omega - H_{33}) - H_{13}H_{31} \\
M_{33} &= (\omega - H_{11})(\omega \mp H_{22}) \mp H_{12}H_{21} \\
M_{12} &= -H_{21}(\omega - H_{33}) + H_{31}H_{23} \\
M_{13} &= -H_{31}(\omega \mp H_{22}) \pm H_{32}H_{21} \\
M_{21} &= \mp H_{12}(\omega - H_{33}) \pm H_{32}H_{13} \\
M_{23} &= \mp H_{32}(\omega - H_{11}) \pm H_{12}H_{31}
\end{aligned}
$$

$$M_{31} = -H_{13}(\omega \mp H_{22}) \pm H_{12}H_{23}$$
$$M_{32} = -H_{23}(\omega - H_{11}) + H_{13}H_{21}.$$

The upper/lower sign above corresponds to the case of the ferrimagnet/ferromagnet. The parameters H_{ij} are

$$H_{11} = Z(S_b J + S_c J')$$
$$H_{12} = -\sqrt{S_a S_b} Z J \gamma_k$$
$$H_{13} = \sqrt{S_a S_c} Z J' \gamma_{-k}$$
$$H_{21} = \sqrt{S_a S_b} Z J \gamma_{-k}$$
$$H_{22} = -Z(S_c + S_a) J$$
$$H_{23} = \sqrt{S_b S_c} Z J \gamma_k$$
$$H_{31} = \sqrt{S_c S_a} Z J' \gamma_k$$
$$H_{32} = -\sqrt{S_b S_c} Z J \gamma_{-k}$$
$$H_{33} = Z(S_b J + S_a J').$$

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