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Effects of biquadratic exchange in ferromagnets

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The effects of the biquadratic-exchange interaction on the temperature dependence of the magnetic specific heat and the magnetization have been calculated for ferromagnets with the use of spin-wave theory and compared with those for antiferromagnets. The ferromagnetic spin structure changes abruptly to a canted one at a certain ratio P_c of the biquadratic exchange to the Heisenberg interaction. This change of spin structure is shown to have significant effects on the magnetic properties. The results of calculations for one-, two-, and three-dimensional systems are compared with one another. It is also shown that the qualitative changes of the magnetic properties of ferromagnets due to the biquadratic-exchange interaction are different from those due to the Dzyaloshinski-Moriya interaction.

I. INTRODUCTION

Recently, the existence of higher-order spin couplings has been pointed out by many authors.[1-5] It has been made clear that these interactions have significant effects on the magnetic properties of compounds containing iron-group ions[6-11] or rare-earth metal ions.[12] The theoretical explanation for the appearance of the biquadratic-exchange interaction was given by Anderson[13,14] and Kittel.[15]

In the previous studies,[16,17] we have discussed the appearance of higher-order spin couplings and estimated the order of magnitude of these terms as one tenth or one hundredth of the Heisenberg-type exchange interaction in polynuclear complex compounds. Afterward, with the use of spin-wave theory, we have shown the importance of the biquadratic-exchange interaction to the magnetic properties of antiferromagnets.[18] In a spin system with biquadratic-exchange interaction with positive sign as well as with Heisenberg-type exchange interaction, the ferromagnetic or the antiferromagnetic spin structure changes abruptly and begins to make a cant at a certain ratio P_c of the biquadratic-exchange to the Heisenberg-type interaction. It has been shown that[18] this abrupt change of spin structure caused by the biquadratic-exchange term has significant effects on the temperature dependence of the specific heat and the magnetization of antiferromagnets.

In view of these facts, it seems necessary and worthwhile to investigate how the biquadratic-exchange interaction affects the magnetic properties of ferromagnets. In the present paper, the ordinary ferromagnetic spin-wave theory is extended to the case of the two-sublattice ferromagnet when the canted spin structure has been caused by the biquadratic-exchange interaction. The temperature dependences of the magnetic specific heat and the magnetization are evaluated by numerical calculations. These calculations are carried out not only for the three-dimensional ferromagnet but also for the one- and two-dimensional ones, and the results obtained are compared with one another.

The effects of the Dzyaloshinski-Moriya interaction on the specific heat and the magnetization of ferromagnets are also examined. It is clarified that the qualitative changes of the magnetic properties of ferromagnets due to the biquadratic-exchange interaction are different from those due to the Dzyaloshinski-Moriya interaction.

In Sec. II, the ordinary ferromagnetic spin-wave theory and the two-sublattice one are developed and their spin-wave dispersion relations are derived. In Sec. III, the magnetic specific heat and the magnetization are evaluated for various values of the biquadratic-exchange interaction and the results are discussed.

II. FERROMAGNETIC SPIN-WAVE THEORY

We consider the ferromagnetic spin system with three kinds of exchange interactions of the Heisenberg type between pairs of spins which are coupled by the constants J_1, J_2, and J_3 as shown in Fig. 1. We also assume the three kinds of biquadratic-exchange interactions denoted by the coupling constants J_1', J_2', and J_3' between pairs of spins coupled by J_1, J_2, and J_3, respectively. In order to maintain the stability of the spin waves the single ionic-type anisotropy terms represented by the constants D and E are introduced.

The Hamiltonian of the present spin system can be written as follows:

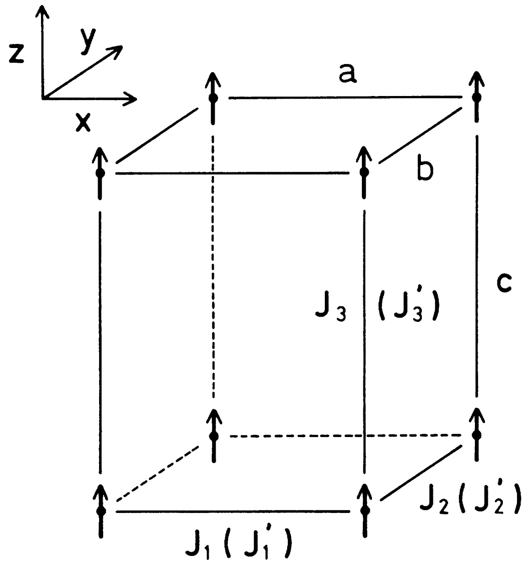

FIG. 1. Ferromagnetic spin structure and the assumed pairs of the Heisenberg-type exchange interactions and the biquadratic-exchange interactions.

$$H=J_1\sum_{\langle ij\rangle}\vec{S}_i\cdot\vec{S}_j+J_2\sum_{\langle ij'\rangle}\vec{S}_i\cdot\vec{S}_{j'}+J_3\sum_{\langle ij''\rangle}\vec{S}_i\cdot\vec{S}_{j''}$$
$$+J_1'\sum_{\langle ij\rangle}(\vec{S}_i\cdot\vec{S}_j)^2+J_2'\sum_{\langle ij'\rangle}(\vec{S}_i\cdot\vec{S}_{j'})^2$$
$$+J_3'\sum_{\langle ij''\rangle}(S_i\cdot S_{j''})^2+D\Big(\sum_i S_{iz}^2+\sum_j S_{jz}^2\Big)$$
$$+E\Big(\sum_i(S_{iy}^2-S_{ix}^2)+\sum_j(S_{jy}^2-S_{jx}^2)\Big),\qquad(1)$$

where $\vec{S}_i$ and $\vec{S}_j$ denote the spin operators belonging to the ith and kth sublattices, and the coordinate axes x, y, and z are defined as shown in Fig. 1.

A. Ordinary ferromagnetic spin wave

In the ferromagnetic spin arrangement, the anisotropy terms in the spin Hamiltonian (1) can be reduced to

$$H_A=D\sum_i S_{iz}^2+E\sum_i(S_{iy}^2-S_{ix}^2).\qquad(2)$$

This Hamiltonian can easily be written in the spin-wave representation. Omitting the higher-order-than-quadratic terms of the creation and annihilation operators, we obtain the spin-wave Hamiltonian. The Hamiltonian thus obtained is diagonalized by making use of the Bogoliubov transformation, and the spin-wave dispersion relation can be obtained as follows:

$$\hbar\Omega=(B_1^2-B_2^2)^{1/2},\qquad(3)$$

where the B_i's are given by

$$B_1=-4S[J_1+J_2+J_3+D+2S^2(J_1'+J_2'+J_3')$$
$$-(J_1\gamma_{1\lambda}+J_2\gamma_{2\lambda}+J_3\gamma_{3\lambda})$$
$$-2S^2(J_1'\gamma_{1\lambda}+J_2'\gamma_{2\lambda}+J_3'\gamma_{3\lambda})],\qquad(4)$$
$$B_2=-2SE.$$

In this expression, the γ_i's are defined as

$$\gamma_{1\lambda}=\cos(a\lambda_x),\quad\gamma_{2\lambda}=\cos(b\lambda_y),\quad\gamma_{3\lambda}=\cos(c\lambda_z).\qquad(5)$$

B. Two-sublattice ferromagnetic spin wave

When the biquadratic-exchange terms in Eq. (1) are effective, the ferromagnetic spin arrangement will be turned into a canted one and the spin system constitutes two sublattices. Let us take the preferred spin directions of the two sublattices as the quantization axes ζ_i and ζ_j, respectively. The relation between the spin components in coordinate systems (x,y,z) and (ξ_i,η_i,ζ_i) or (ξ_j,η_j,ζ_j) is given by the following coordinate transformation:

$$S_{ix}=S_{i\xi}\cos\theta+S_{i\zeta}\sin\theta,\quad S_{iy}=S_{i\eta},$$
$$S_{iz}=-S_{i\xi}\sin\theta+S_{i\zeta}\cos\theta,\qquad(6)$$
$$S_{jx}=S_{j\xi}\cos\theta-S_{j\zeta}\sin\theta,\quad S_{jy}=S_{j\eta},$$
$$S_{jz}=S_{j\xi}\sin\theta+S_{j\zeta}\cos\theta,$$

where θ is the angle between the z axis and the preferred spin axis as shown in Fig. 2. The spin

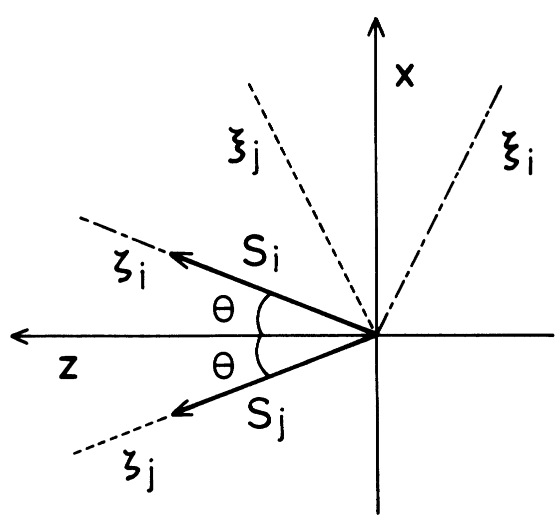

FIG. 2. Spin arrangement in ferromagnetic state with canted angle θ. The new coordinate axes of quantization are shown by ξ and ζ.

operators S_i and S_j are rewritten with the use of the following creation and annihilation operators:

$$S_{i\zeta}=S-a_i^*a_i,\quad S_{j\zeta}=S-A_j^*A_j,$$
$$S_i^+=(2S)^{1/2}f_i(S)a_i,\quad S_j^+=(2S)^{1/2}f_j(S)A_j, \tag{7}$$
$$S_i^-=(2S)^{1/2}a_i^*f_i(S),\quad S_j^-=(2S)^{1/2}A_j^*f_j(S),$$

where $f_i(S)$ and $f_j(S)$ are defined as

$$f_i(S)=(1-a_i^*a_i/2S)^{1/2},\quad f_j(S)=(1-A_j^*A_j/2S)^{1/2}. \tag{8}$$

The spin Hamiltonian expressed by the creation and annihilation operators is further rewritten with the use of the following Fourier transformation:

$$a_i=\left(\frac{2}{N}\right)^{1/2}\sum_\lambda e^{-i\lambda r_i}a_\lambda,\quad A_i=\left(\frac{2}{N}\right)^{1/2}\sum_\lambda e^{-i\lambda x_j}A_\lambda,$$
$$a_i^*=\left(\frac{2}{N}\right)^{1/2}\sum_\lambda e^{i\lambda r_i}a_\lambda^*,\quad A_i^*=\left(\frac{2}{N}\right)^{1/2}\sum_\lambda e^{i\lambda r_j}A_\lambda^*. \tag{9}$$

Omitting the higher-order-than-quadratic terms of these operators, we obtain the spin-wave Hamiltonian as follows:

$$\begin{aligned}H=\sum_{\lambda>0}\,[&C_1(a_\lambda^*a_\lambda+a_{-\lambda}^*a_{-\lambda}+A_\lambda^*A_\lambda+A_{-\lambda}^*A_{-\lambda})\\&+C_2(a_\lambda A_\lambda^*+a_\lambda^*A_\lambda+a_{-\lambda}A_{-\lambda}^*+a_{-\lambda}^*A_{-\lambda})\\&+C_3(a_\lambda A_{-\lambda}+a_{-\lambda}A_\lambda+a_\lambda^*A_{-\lambda}^*+a_{-\lambda}^*A_\lambda^*)\\&+C_4(a_\lambda a_{-\lambda}+a_\lambda^*a_{-\lambda}^*+A_\lambda A_{-\lambda}+A_\lambda^*A_{-\lambda}^*)],\end{aligned} \tag{10}$$

where the constant terms are neglected. In the above expression, the C_i's are given by

$$\begin{aligned}C_1&=-2F_1S(J_1+J_2+J_3)-2F_4S+S(F_3+E)\\&\quad-4S^3(F_1^2-2F_2^2)(J_1'+J_2'+J_3'),\\C_2&=S(F_1-1)(J_1\gamma_{1\lambda}+J_2\gamma_{2\lambda}+J_3\gamma_{3\lambda})\\&\quad+2S^3(F_1^2-F_1-4F_2^2)(J_1'\gamma_{1\lambda}+J_2'\gamma_{2\lambda}+J_3'\gamma_{3\lambda}),\\C_3&=S(F_1+1)(J_1\gamma_{1\lambda}+J_2\gamma_{2\lambda}+J_3\gamma_{3\lambda})\\&\quad+2S^3(F_1^2+F_1-4F_2^2)(J_1'\gamma_{1\lambda}+J_2'\gamma_{2\lambda}+J_3'\gamma_{3\lambda}),\\C_4&=S(F_3-E)+8F_2^2S^3(J_1'+J_2'+J_3'),\end{aligned} \tag{11}$$

where the F_i's are defined as follows:

$$F_1=\cos^2\theta-\sin^2\theta,\quad F_2=\sin\theta\cos\theta,$$
$$F_3=D\sin^2\theta-E\cos^2\theta,\quad F_4=D\cos^2\theta-E\sin^2\theta. \tag{12}$$

The spin-wave Hamiltonian (10) is easily diagonalized by making use of the Bogoliubov transformation. The diagonalized Hamiltonian is given by

$$H=\sum_{\lambda>0}\,[\hbar\Omega_1(Q_\lambda^*Q_\lambda+R_\lambda^*R_\lambda)+\hbar\Omega_2(Q_{-\lambda}^*Q_{-\lambda}+R_{-\lambda}^*R_{-\lambda})], \tag{13}$$

where the spin-wave dispersion relations are obtained as

$$\hbar\Omega_1=[(C_1+C_2)^2-(C_3+C_4)^2]^{1/2},$$
$$\hbar\Omega_2=[(C_1-C_2)^2-(C_3-C_4)^2]^{1/2}. \tag{14}$$

From the condition that the linear terms of operators in the spin Hamiltonian must vanish under the equilibrium condition,[19] we can determine the value of θ. If we define the parameter r as

$$r=-\frac{S^2(J_1'+J_2'+J_3')}{J_1+J_2+J_3+\frac{1}{2}(D+E)}, \tag{15}$$

the equilibrium spin direction is obtained as follows:

$$\sin\theta=0\quad(\theta=0)\quad\text{for } r\le\tfrac{1}{2},$$
$$\cos 2\theta=1/2r\quad\text{for } r>\tfrac{1}{2}. \tag{16}$$

Therefore, in the range of small values of the biquadratic-exchange interaction which satisfy the relation $r\le\frac{1}{2}$, the spin arrangement is ferromagnetic and the spin-wave dispersion relations, Eq. (3), obtained in subsection A are effective. On the other hand, in the range $r>\frac{1}{2}$, there appears the canted spin structure and the spin system should be described by the formulas developed in subsection B, and the spin-wave dispersion relations, Eq. (14), are used.

III. RESULTS AND DISCUSSION

With the use of the spin-wave dispersion relations derived in Sec. II, we have evaluated the temperature dependences of the magnetic specific heat C_M and the magnetization $\langle S_\zeta\rangle$ by making use of equations similar to those given in the previous paper.[18] As the integrals in those equations are very complicated, C_M and $\langle S_\zeta\rangle$ have been calculated numerically over the magnetic first Brillouin zone of the three-dimensional wave-vector space.

First, the wave-number dependence of the spin-wave dispersion relations has been calculated in order to facilitate conjecture on the qualitative behaviors of the temperature dependences of C_M and $\langle S_\zeta\rangle$. The results for several values of the biquadratic-exchange interaction are shown in Fig. 3. The parameter P in Fig. 3 represents a ratio of the biquadratic exchange to the Heisenberg interaction, which is defined as

$$P=\frac{J_1'+J_2'+J_3'}{J_1+J_2+J_3}=\frac{J_1'}{J_1}=\frac{J_2'}{J_2}=\frac{J_3'}{J_3}. \tag{17}$$

It turns out from Eq. (16) that the spin structure begins to make a cant at the critical ratio P_c, which is estimated as

$$P_c=|J'/J|=1/2S^2+(D+E)/4S^2J\simeq 0.0813, \tag{18}$$

for the spin system of $S=\frac{5}{2}$ with a set of values of

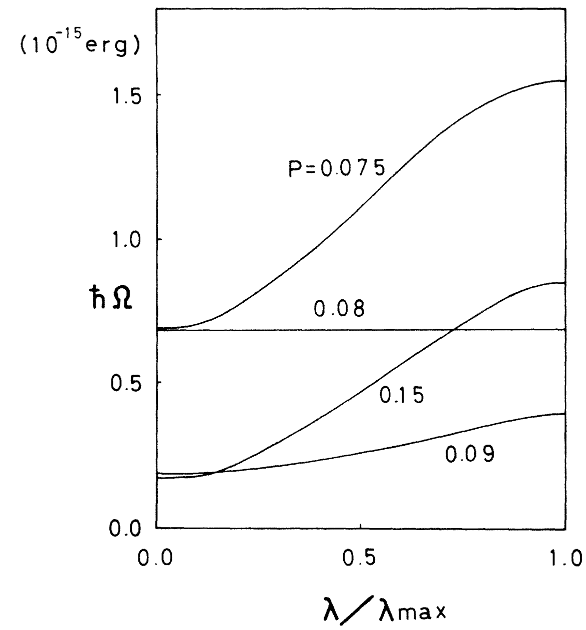

FIG. 3. Spin-wave dispersion curves along the x axis (J_1 direction). The curves have been calculated for $J_1 = -15.0k_B$, $J_2 = J_3 = -0.001k_B$, $D = -0.5k_B$, $E = 0.1k_B$, and $S = \frac{5}{2}$.

the interaction parameters $J_1 = J_2 = J_3 = -5.0k_B$, $D = -0.5k_B$, and $E = 0.1k_B$. As the parameter P increases and closes to P_c, the spin-wave dispersion curve becomes flat and almost independent of the wave number. As the parameter P increases over P_c, the dispersion curve begins to have more rapid wave-number dependence again. This fact suggests to us that the spin structure must be unstable near or at P_c. This is the same result as was obtained in the case of antiferromagnets of the previous paper.

At the beginning, the temperature dependences of C_M and $\langle S_\xi \rangle$ have been calculated for the three-dimensional ferromagnetic spin system of $S = \frac{5}{2}$ with the same set of values of the interaction parameters as in Eq. (18). The results of numerical calculation of C_M and $\langle S_\xi \rangle$ for various values of the biquadratic-exchange interaction are shown in Figs. 4(a) and 4(b) by the solid lines. The specific-heat curves show an abrupt increase and the magnetization curves a rapid decrease near or at P_c. These behaviors of C_M and $\langle S_\xi \rangle$ are thought to originate in an instability of the spin system. As has been shown by Thorpe,[20] the biquadratic-exchange term $(\vec{S}_i \cdot \vec{S}_j)^2$ can be written as $C^2(\vec{S}_i) \cdot C^2(\vec{S}_j) - \frac{1}{2}(\vec{S}_i \cdot \vec{S}_j) + \frac{1}{3}S^2(S+1)^2$ and has a seemingly antiferromagnetic character. Therefore the instability may be due to an effective cancellation of the ferromagnetic exchange by the biquadratic-

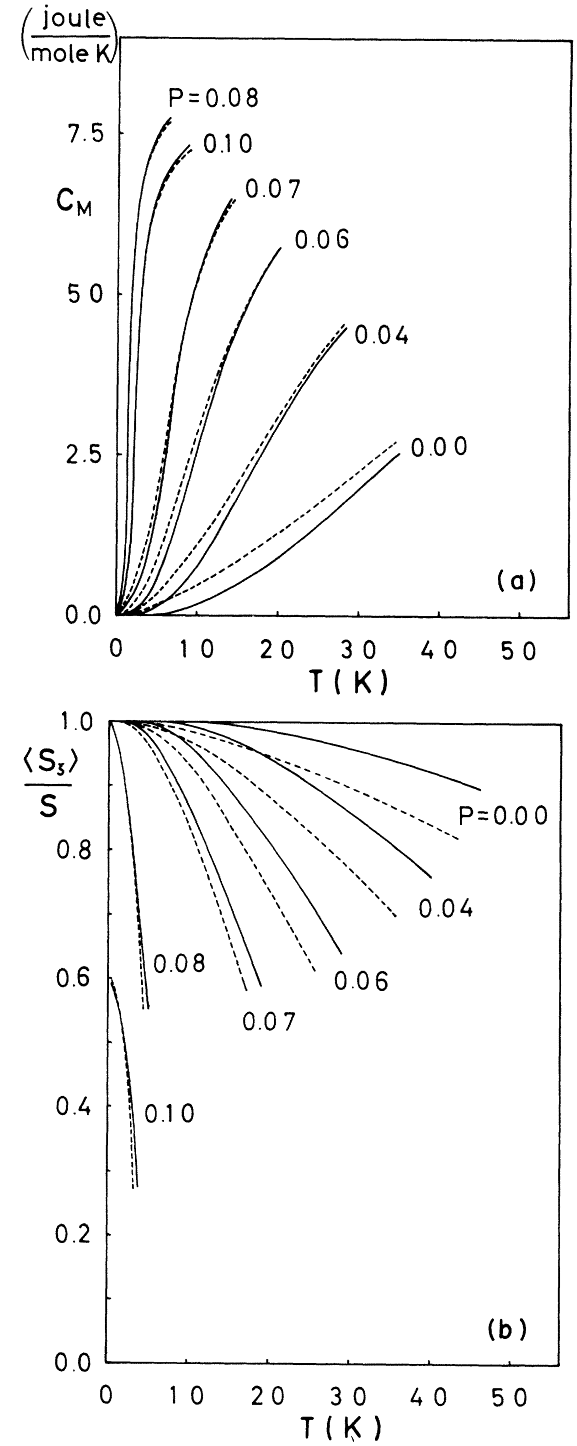

FIG. 4. Temperature dependences of the magnetic specific heat (a) and the thermal average of S_ξ (b) for several values of P $(=J_1'/J_1 = J_2'/J_2 = J_3'/J_3)$. The full lines and the broken ones are calculated for the three-dimensional ferromagnet with $J_1 = J_2 = J_3 = -5.0k_B$ and the two-dimensional one with $J_1 = J_2 = -7.5k_B$ and $J_3 = -0.001k_B$, respectively. In this case, other parameters are fixed as $D = -0.5k_B$, $E = 0.1k_B$, and $S = \frac{5}{2}$.

exchange interaction. These results may be reasonably understood from the conjecture made on the spin-wave dispersion relation. It is noticeable that the zero-point spin deviation in ferromagnets is much smaller than that in antiferromagnets in the range of parameter P below P_c. However, the qualitative behaviors of C_M and $\langle S_\xi \rangle$ curves are similar to those for antiferromagnets.

The temperature dependence of C_M and $\langle S_\xi \rangle$ for the ferromagnetic spin system of $S=2$ has also been calculated. The qualitative behaviors are almost similar to those for the spin system of $S=\frac{5}{2}$ and abrupt changes of the specific-heat and the magnetization curves also occur near or at P_c. In this case, the value of P_c is estimated from Eq. (18) as $P_c=0.1271$.

Next, the temperature dependences of C_M and $\langle S_\xi \rangle$ for a substantially two-dimensional ferromagnetic spin system has been calculated and compared with those for the three-dimensional one. The results are shown in Figs. 4(a) and 4(b) by the broken lines. In this calculation, the exchange and anisotropy parameters are fixed as $J_1=J_2=-7.5k_B$, $J_3=-0.001k_B$, $D=-0.5k_B$, and $E=0.1k_B$. The specific-heat curves rise more rapidly and the magnetization curves fall faster than those for the three-dimensional case. These qualitative behaviors may suggest to us that the two-dimensional spin system is more unstable than the three-dimensional one.

Furthermore, we have calculated the temperature dependence of C_M and $\langle S_\xi \rangle$ for a substantially one-dimensional ferromagnetic spin system. The results for various values of the biquadratic-exchange interaction are shown in Figs. 5(a) and 5(b) and are compared with those for the two-dimensional spin system. In this calculation, the exchange and anisotropy parameters are fixed as $J_1=-15.0k_B$, $J_2=J_3=-0.001k_B$, $D=-0.5k_B$, and $E=0.1k_B$. As seen from the figures, the specific-heat and the magnetization curves for the one-dimensional spin system have more rapid temperature dependences than those for the two-dimensional one. These behaviors of C_M and $\langle S_\xi \rangle$ may be explained by taking into consideration that the one-dimensional spin system is much more unstable than the two-dimensional one. An exact calculation by Thorpe and Blume[21] of a linear chain of classical spins with near-neighbor bilinear- and biquadratic-isotropic-exchange interactions may be compared with the present results.

As is well known, the canted spin structure is also established by the existence of the Dzyaloshinski-Moriya (DM) interaction. Although seemingly both the biquadratic and DM interactions are similarly effective for the canting mechanism,

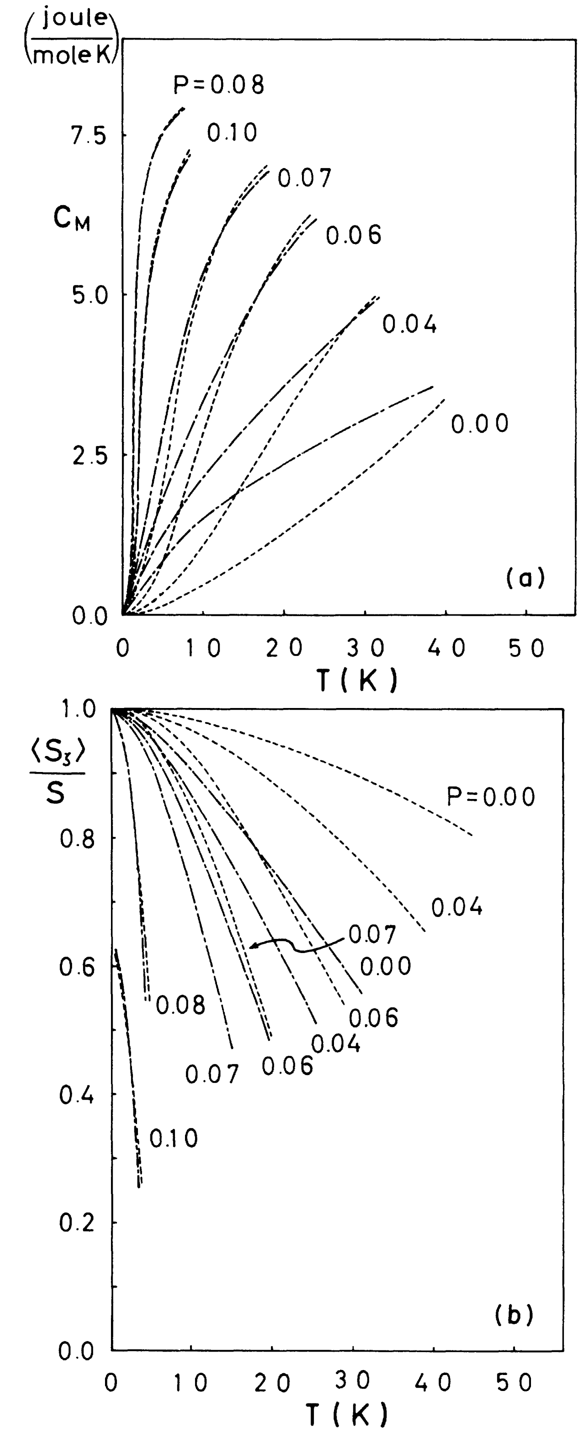

FIG. 5. Temperature dependences of the magnetic specific heat (a) and the thermal average of S_ξ (b) for several values of P $(=J_1'/J_1=J_2'/J_2=J_3'/J_3)$. The broken lines and the dash-dotted ones are calculated for the two-dimensional ferromagnet with $J_1=J_2=-7.5k_B$ and $J_3=-0.001k_B$, and the one-dimensional one with $J_1=-15.0k_B$ and $J_2=J_3=-0.001k_B$, respectively. In this case, other parameters are fixed as $D=-0.5k_B$, $E=0.1k_B$, and $S=\frac{5}{2}$.

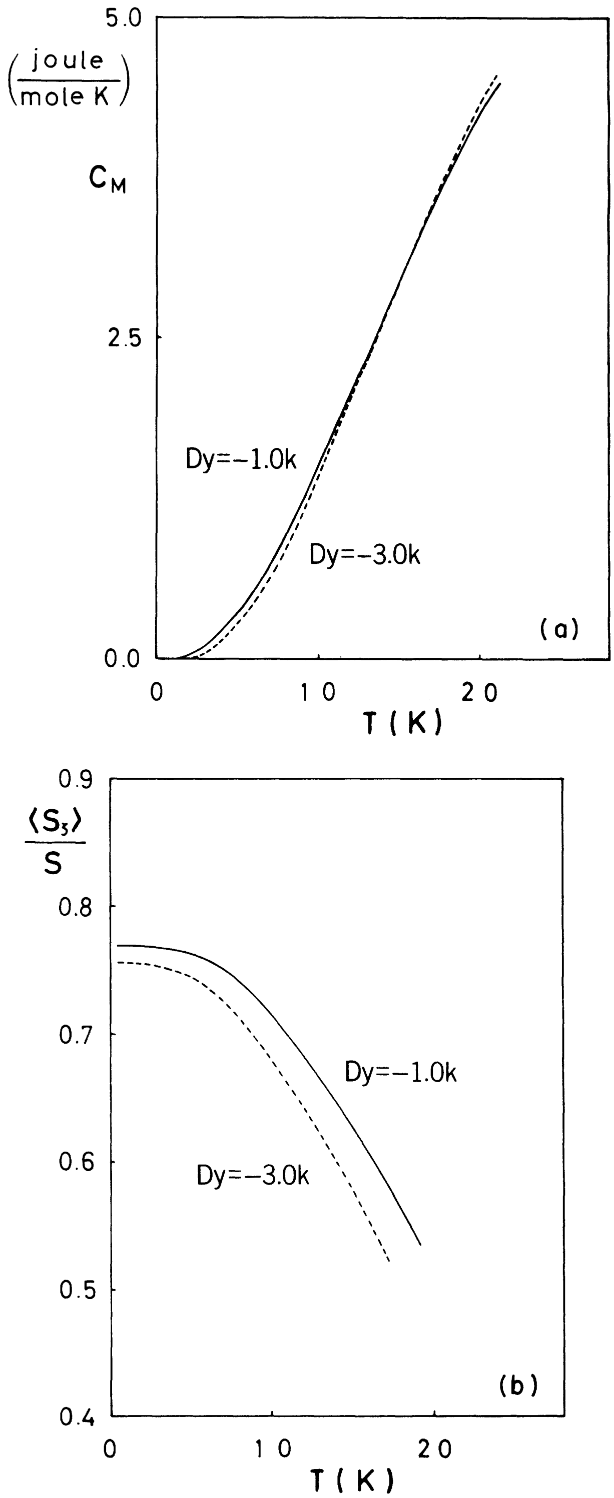

FIG. 6. Temperature dependences of the magnetic specific heat (a) and the thermal average of S_ζ (b) for a couple of values of the DM interaction. The theoretical curves have been calculated for $J_1=-15.0k_B$, $J_2=J_3=-0.001k_B$, $D=-0.5k_B$, $E=0.1k_B$, and $S=\frac{5}{2}$.

it is interesting to investigate the effects of the DM interaction on the temperature dependence of C_M and $\langle S_\zeta\rangle$, and compare the results with those of the biquadratic-exchange interaction. Instead of the biquadratic-exchange interaction, we introduce the following DM interaction term in the spin Hamiltonian, Eq. (1):

$$\vec{D}_y \cdot (\vec{S}_i \times \vec{S}_j) = D_y(S_{iz}S_{jx} - S_{ix}S_{jz}). \tag{19}$$

The spin-wave Hamiltonian related to the DM interaction is obtained as follows:

$$H_{DM} = 2SD_y \sum_\lambda F_2[-(a_\lambda^* a_\lambda + A_\lambda^* A_\lambda) + \gamma_{1\lambda}(a_\lambda A_{-\lambda} + a_\lambda^* A_{-\lambda}^*) + \gamma_{1\lambda}(a_\lambda A_\lambda^* + a_\lambda^* A_\lambda)]. \tag{20}$$

The temperature dependences of C_M and $\langle S_\zeta\rangle$ have been obtained by numerical calculations for a couple of values of the DM interaction. The results are shown in Figs. 6(a) and 6(b). In this calculation, other parameters are fixed as $J_1 = -15.0k_B$, $J_2=J_3=-0.001k_B$, $J_1'=J_2'=J_3'=0$, $D=-0.5k_B$, and $E=0.1k_B$. The DM interaction turns out to give no drastic change of the temperature dependences of C_M and $\langle S_\zeta\rangle$. This fact shows that the biquadratic-exchange interaction and the DM interaction are essentially different in the effects on the magnetic properties of the ferromagnetic spin system.

Summarizing the present results, we may conclude as follows:

(i) The magnetic properties of ferromagnets show drastic changes near or at P_c due to an instability of the spin system. The qualitative behaviors of the temperature dependences of C_M and $\langle S_\zeta\rangle$ for the ferromagnetic spin system are almost similar to those for the antiferromagnetic one, although the zero-point spin deviation in ferromagnets is much smaller than that in antiferromagnets in the range of parameter P below P_c.

(ii) The specific-heat and the magnetization curves for the low-dimensional spin systems show more rapid temperature dependence than those for the three-dimensional one. These facts may be understood by taking into consideration that the low-dimensional spin system is more unstable than the three-dimensional one.

(iii) Qualitatively the effects of the biquadratic-exchange interaction on the magnetic properties of the ferromagnetic spin system are different from those of the DM interaction, which also leads to a canted spin structure.

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