Effect of a three-site four-spin interaction on the $S=1$ axial third-nearest-neighbor Ising model: Application to the magnetic phase diagrams of UNi_2Si_2

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(Received 10 May 2001; published 12 September 2001)

We consider the extended axial third-nearest-neighbor Ising model, which consists of $S=1$ Ising spins and includes not only the ordinary bilinear exchange interaction but also the three-site four-spin interaction as the next-nearest-neighbor interaction. The finite-temperature magnetic-phase diagram of this model is investigated by means of the site-dependent molecular-field approximation. The phase diagram shows the direct phase transition from the ground-state $\langle 12\rangle(q=1/3)$ phase to the antiferromagnetic (AF) phase, which has been observed in UNi_2Si_2. This phase transition is caused by a drastic increase of the entropy in the AF state. The magnetic-field-temperature phase diagram is also in agreement with that in UNi_2Si_2 qualitatively.

DOI: 10.1103/PhysRevB.64.134416 PACS number(s): 05.50.+q, 75.10.Hk, 75.30.Kz

I. INTRODUCTION

The axial next-nearest-neighbor Ising (ANNNI) model has attracted many investigators on account of the fact that it is a particularly simple model exhibiting spatially modulated phases that can be either commensurate or incommensurate with the underlying lattice.[1,2] The main feature of the results obtained by theoretical studies is the large numbers of modulated phases that can arise with variation of the interaction strengths and temperature,[2–7] and has been described by the ''devil's staircase'' and ''devil's flower.'' The ANNNI model also reproduced crucial qualitative features observed experimentally in adsorbate systems, ferroelectrics, magnetic systems, alloy, liquid crystal, and so on that exhibit modulated structures.[1,8]

Extended ANNNI models have been proposed concerning the spin quantum number[9,10] and/or the range of competing interactions.[11–13] The axial third-nearest-neighbor Ising (A3NNI) model described by the following Hamiltonian,

$$\mathcal{H}=-\sum_i\sum_j\left[\frac{J_0}{2}\sum_{k=1}^{z}S_{i,j}S_{i,k}+J_1S_{i,j}S_{i+1,j}+J_2S_{i,j}S_{i+2,j}+J_3S_{i,j}S_{i+3,j}+hS_{i,j}\right], \tag{1}$$

is one of such systems and has been studied as extensively as the ANNNI model. This model is composed of chains of spins parallel to the c axis (say), and nearest-neighbor chains interact through ferromagnetic interchain interactions, $J_0>0$. The spin on site j in the ith c plane is denoted by $S_{i,j}(=\pm 1)$, and the summations on i and j are taken over all the spins along the chain and in the c plane, respectively. The summation on k is over the z nearest neighbors of site j in the c plane. J_1, J_2, and J_3 are the nearest neighbor, the next-nearest-neighbor, and the third-nearest-neighbor intrachain interactions, respectively (Fig. 1), and h represents external magnetic field. Most of the observed features of incommensurate-commensurate phase transitions in A_2BX_4-type dielectrics and the ferrielectric phases appearing in the sequences of phase transitions of certain ferroelectric smectic liquid crystals have been explained by the phase diagram of the A3NNI model.[11,14]

In the case of the extended model concerning the spin quantum number, we can consider not only the ordinary bilinear exchange interaction—JS_iS_j, but also the higher-order spin interaction such as the biquadratic exchange interaction—$JS_i^2S_j^2$, the three-site four-spin interaction—$JS_iS_j^2S_k$, and so on. These higher-order spin interactions originate from the magnetostriction or can be derived from the higher-order perturbation procedure.[15] In previous works,[16,17] we have studied the $S\geqslant 1$ ANNNI model ($J_1>0$) with the three-site four-spin interaction. The molecular-field approximation and Monte Carlo calculation have revealed that there exists the sequence of phases written as $\langle 2\rangle\rightarrow$Ferromagnetic($F$) phase $\rightarrow$Modulated(M) phase $\rightarrow$Paramagnetic(P) phase with increasing temperature, and the parameter range corresponding to this successive transition is enlarged with increasing the spin quantum number, S.

UNi_2Si_2 belongs to compounds with the body-centered-tetoragonal structure of the generic formula AT_2X_2 and has long-range Ruderman-Kittel-Kasuya-Yosida interaction and strong c-axial anisotropy. Three-ordered phases were observed by the neutron diffraction, where the moment lie

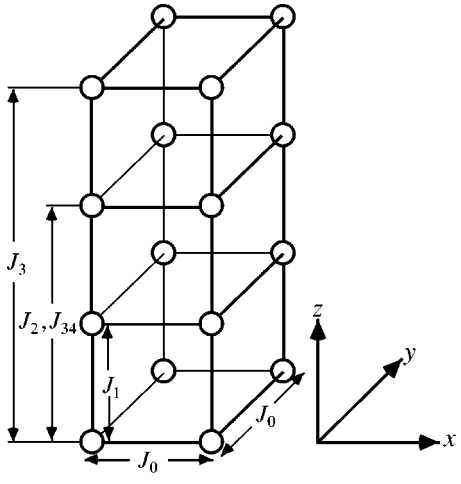

FIG. 1. The three-dimensional A3NNI model.

along the *c* axis and the ordering in the planes perpendicular to the *c* axis for three phases is ferromagnetic.[18] The low-temperature phase ($T<53$ K) is a $\langle 12\rangle(q=1/3)$ modulated phase, where *q* represents the wave number along *c* axis. The intermediate temperature phase ($53<T<103$ K) is an antiferromagnetic (AF) phase along the *c* axis. The high-temperature phase ($103<T<124$ K) is an incommensurate (IC) phase. The paramagnetic phase is stabilized for $T>124$ K. The sequence of phases observed with increasing temperature can be written as $\langle 12\rangle \rightarrow$ AF $\rightarrow$ IC $\rightarrow P$.

Although the $\langle 12\rangle$ phase corresponds to the low-temperature phase in the ordinary A3NNI model ($J_1<0$), there does not exist the sequence $\langle 12\rangle \rightarrow$ AF $\rightarrow$ IC $\rightarrow P$ in the phase diagram of this model.[19] The classical Heisenberg version of A3NNI model with both the easy-axis anisotropy—DS_i^2, and the nearest-neighbor biquadratic exchange interaction—$JS_i^2S_j^2$, along the *c* axis is proposed as a model for UNi_2Si_2 by Mailhot *et al.*[20] Although the low-temperature sequence founded in UNi_2Si_2 is not accountable within the mean-field Landau-type calculation for this model, the low-temperature spin-wave expansion reveals that thermal fluctuations select the AF phase only for sufficiently large biquadratic exchange interaction, that is, the phase transition from the $\langle 12\rangle$ phase to the AF phase takes place.

In this paper, we propose another model for UNi_2Si_2 in which the low-temperature sequence is accountable within the molecular-field approximation. We consider the $S=1$ A3NNI model ($J_1<0$) with the three-site four-spin interaction along the *c* axis described by the following Hamiltonian,

$$\mathcal{H}=-\sum_i \sum_j \left[\frac{J_0}{2} \sum_{k=1}^{z} S_{i,j}S_{i,k}+J_1S_{i,j}S_{i+1,j}+J_2S_{i,j}S_{i+2,j} + J_3S_{i,j}S_{i+3,j}+J_{34}S_{i,j}S_{i+1,j}^2S_{i+2,j}+hS_{i,j} \right], \tag{2}$$

where $S_{i,j}=0,\pm 1$, and J_{34} is the three-site four-spin interaction along the *c* axis (Fig. 1). The site-dependent molecular-field approximation reveals that the three-site four-spin interaction provides the $\langle 12\rangle \rightarrow$ AF phase transition at low temperature and this phase transition is attributed to the increase of the entropy in AF state.

The arrangement of the paper is as follows. In Sec. II, based on the site-dependent molecular-field approximation, the thermal averages, free energy, and entropy are derived. In Sec. III, by means of numerical calculation, magnetic phase diagrams under the zero and finite external field are obtained. The low-temperature sequence in UNi_2Si_2 is discussed. Finally, in Sec. IV the results are summarized and concluding remarks given.

II. THE SITE-DEPENDENT MOLECULAR-FIELD APPROXIMATION

For the ferromagnetic interchain interaction, J_0, it is believed that all spins within the same *c* plane take the same thermal average. Therefore, using a site-dependent molecular-field approximation, Eq. (2) can be reduced to the following chain Hamiltonian,

$$\mathcal{H}_{\mathrm{MF}}=-\sum_i [S_i(H_i+h)+S_i^2H_i^*], \tag{3}$$

where

$$H_i=4J_0\langle S_i\rangle+J_1(\langle S_{i-1}\rangle+\langle S_{i+1}\rangle)+J_2(\langle S_{i-2}\rangle+\langle S_{i+2}\rangle) + J_3(\langle S_{i-3}\rangle+\langle S_{i+3}\rangle)+J_{34}(\langle S_{i-2}\rangle\langle S_{i-1}^2\rangle + \langle S_{i+1}^2\rangle\langle S_{i+2}\rangle), \tag{4}$$

$$H_i^*=J_{34}\langle S_{i-1}\rangle\langle S_{i+1}\rangle, \tag{5}$$

S_i, $\langle S_i\rangle$, and $\langle S_i^2\rangle$ denote the spin on the *i*th *c* plane and its thermal averages, respectively. From Eqs. (3), (4), and (5), the thermal averages of S_i and S_i^2

$$\langle S_i\rangle=\frac{2\exp(\beta H_i^*)\sinh[\beta(H_i+h)]}{1+2\exp(\beta H_i^*)\cosh[\beta(H_i+h)]}, \tag{6}$$

$$\langle S_i^2\rangle=\frac{2\exp(\beta H_i^*)\cosh[\beta(H_i+h)]}{1+2\exp(\beta H_i^*)\cosh[\beta(H_i+h)]}, \tag{7}$$

and the free energy per spin and the entropy per spin

$$F=\frac{1}{N}\sum_{i=1}^{N}\left(-k_{\mathrm{B}}T\log\{1+2\exp(\beta H_i^*)\cosh[\beta(H_i+h)]\} + \frac{1}{2}\langle S_i\rangle H_i+\langle S_i^2\rangle H_i^* \right), \tag{8}$$

$$S=\frac{1}{N}\sum_{i=1}^{N}\left(k_{\mathrm{B}}\log\{1+2\exp(\beta H_i^*)\cosh[\beta(H_i+h)]\} - \langle S_i\rangle\frac{H_i+h}{T}-\langle S_i^2\rangle\frac{H_i^*}{T} \right), \tag{9}$$

are obtained, where $\beta=1/k_{\mathrm{B}}T$ and k_{B} is the Boltzmann constant.

III. MAGNETIC PHASE DIAGRAM

In this section, to investigate the ordered phase under the zero and finite external field, the coupled equations [Eqs. (6) and (7)] for spins up to $N=17$ are self-consistently solved by means of the iteration. The stable spin structure is determined as the solution that minimizes the free energy given by Eq. (8).

A. Magnetic Phase Diagram

The finite-temperature magnetic phase diagram for $\kappa_2=J_2/|J_1|=0.5, \kappa_3=J_3/|J_1|=0.1$, and $h/|J_1|=0$ is shown in Fig. 2, where $\kappa_{34}=J_{34}/|J_1|$. This phase diagram shows that the AF and $\langle 12\rangle(q=1/3)$ phases extend into the $\langle 12\rangle$ and $\langle 2\rangle(q=1/4)$ phases, respectively. Accordingly, the direct-phase transition from the ground-state $\langle 12\rangle(q=1/3)$ phase to the AF phase, which has been observed in UNi_2Si_2, takes place for $0.85<-\kappa_{34}<0.97$. The parameter range, where this phase transition occurs, enlarges with increasing the

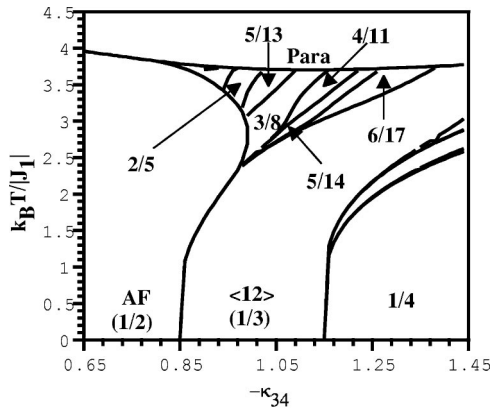

FIG. 2. The magnetic phase diagram of the $S=1$ A3NNI model with the three-site four-spin interaction for $\kappa_2=J_2/|J_1|=0.5$, $\kappa_3=J_3/|J_1|=0.1$, and $h/|J_1|=0$. Fraction in phase diagram denotes a wave number along the c axis.

next-nearest-neighbor bilinear exchange interaction, κ_2. Figure 3 shows the magnetic-field-temperature phase diagram for $\kappa_2=0.5, \kappa_3=0.1$, and $\kappa_{34}=-0.93$, where M denotes the modulated phases except for the $\langle 12\rangle$ phase. The region of the AF phase becomes narrow with increasing the magnetic field and there does not exist the AF phase under the high magnetic field. This phase diagram is qualitatively in agreement with that of UNi_2Si_2.[21]

In our calculation of $N\leqslant 17$, the spin structures corresponding to $q=0$ and $\frac{1}{17}\leqslant q\leqslant\frac{1}{2}$ can be taken into account. For higher values of N, complex structures with longer periodicities will appear in the phase diagram. These structures, however, are not stable except within an extremely narrow temperature region in the phase diagram, and so no additional significant insight can be achieved by extending the numerical calculation to such higher values of N.[3] In the present study, due to the small lattices ($N\leqslant 17$) and the periodic boundary condition, the systematics of the high-order commensurate phases are missed. To make up for these defects, it is necessary to do more sophisticated analysis of the mean-field equations, in paticular following Selke and Duxbury.[22] However these defects are not significant as far as the low-temperature sequence in UNi_2Si_2 is concerned.

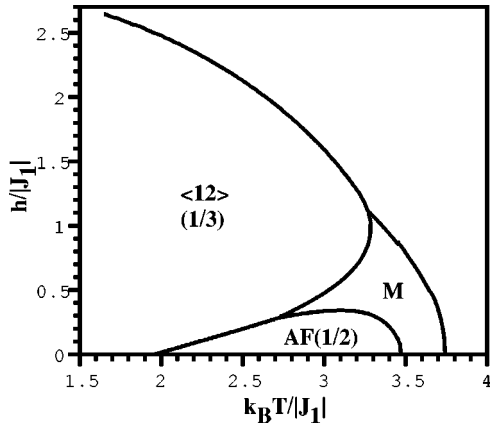

FIG. 3. The magnetic-field-temperature magnetic phase diagram of the A3NNI model with the three-site four-spin interaction for $\kappa_2=0.5, \kappa_3=0.1$, and $\kappa_{34}=-0.93$, where M denotes the modulated phases except for the $\langle 12\rangle$ phase.

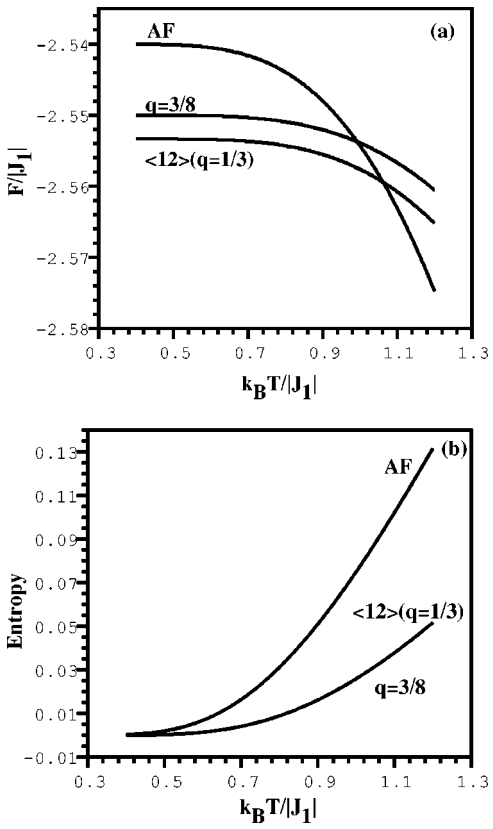

FIG. 4. Temperature dependences of (a) the free energy per spin and (b) the entropy per spin in the AF, $\langle 12\rangle(q=1/3)$ and $q=3/8$ phases. Although the entropy in $\langle 12\rangle$ phase is slightly larger than that in $q=3/8$ phase, the temperature dependences in both phases are described by the same curve in Fig. 4(b).

B. The $\langle 12\rangle\rightarrow$AF Phase Transition

Figure 4 shows the temperature dependence of the free energy per spin and the entropy per spin in the vicinity of the multiphase point, $(-\kappa_{34}^*, k_BT/|J_1|)=(1/2+\kappa_2-3\kappa_3/2, 0)$, between the $\langle 12\rangle(q=1/3)$ phase and the AF phase, where $\Delta=-\kappa_{34}+\kappa_{34}^*=0.01$. Although the entropy in $\langle 12\rangle$ phase is slightly larger than that in $q=3/8$ phase, the temperature dependences in both phases are described by the same curve in Fig. 4(b). It is found that with increasing temperature, the free energy in the AF phase decreases and is less than that in the $\langle 12\rangle$ phase in $k_BT/|J_1|\geqslant 1.06$. With increasing temperature, the entropy in the AF phase drastically increases comparing with those in $\langle 12\rangle(q=1/3)$ and $q=3/8$ phases. On the other hand, the internal energy in the AF phase increases with increasing temperature and is always larger than those in other phases in $k_BT/|J_1|\leqslant 1.4$. As a consequence, the free energy in the AF phase drastically decreases due to the increase of the entropy and the direct phase transition from the ground-state $\langle 12\rangle(q=1/3)$ phase to the AF phase, which has been observed in UNi_2Si_2, takes place.

This entropy effect stabilizing the AF phase is attributed to the existence of the three-site four-spin interaction. The

Hamiltonian in the molecular-field approximation [Eq. (3)] suggests that H_i^* can be regarded as the effective field applied to S_i^2. Since $H_i^*<0$ in the AF phase for $J_{34}<0$, H_i^* stabilizes the AF phase with small S_i^2, that is, the antiferromagnetic phase including many spins with $S_i=0$ and having small $\langle S_i\rangle$. Then, $\delta=S_i-\langle S_i\rangle$ becomes large enough not to be neglected and results in the increase of the entropy. Consequently, the three-site four-spin interaction has the same effect as the single-ion anisotropy, $DS_i^2(D>0)$, and the increase of the entropy in the AF phase by this effect results in the direct phase transition from the ground-state $\langle 12\rangle(q=1/3)$ phase to the AF phase.

The magnetoelastic effect is one of mechanisms to verify the existence of the three-site four-spin interaction.[15] The three-site four-spin interaction is derived from the change of both the coupling constant of the exchange interaction and the elastic energy due to the change of the distance between magnetic ions. In UNi_2Si_2, the temperature dependence of the relative length change along the *a* and *c* axes has been observed for the temperature range $4.2\leqslant T\leqslant 300$ K.[21] It is clearly seen that the thermal expansion in UNi_2Si_2 is very large. Our result, therefore, suggests that the direct phase transition from the $\langle 12\rangle$ phase to the AF phase is attributed to the large thermal expansion in UNi_2Si_2.

IV. CONCLUSION

We proposed $S=1$ A3NNI model ($J_1<0$) with the three-site four-spin interaction along the *c* axis as a model for UNi_2Si_2. By means of the site-dependent molecular-field approximation, we obtained the magnetic phase diagrams. The temperature-κ_{34} phase diagram (Fig. 2) has the parameter range where the direct phase transition from the $\langle 12\rangle(q=1/3)$ phase to the AF phase takes place, and our model has qualitatively reproduced the phase transition in UNi_2Si_2. The magnetic-field-temperature phase diagram (Fig. 3) is also in agreement with that in UNi_2Si_2 qualitatively. Consequently, in our model the low-temperature sequence observed in UNi_2Si_2 is accountable within the molecular-field approximation for sufficiently large three-site four-spin interaction.

The free energy per spin and the entropy per spin in the vicinity of the multiphase point were also calculated within the molecular-field approximation. In this calculation, we have found that the free energy in the antiferromagnetic phase drastically decreases due to the increase of the entropy and the direct phase transition from the $\langle 12\rangle(q=1/3)$ phase to the AF phase takes place. This entropy effect stabilizing the AF phase can be understood by regarding the three-site four-spin interaction in the molecular-field Hamiltonian [Eq. (3)] as the single-ion anisotropy, $DS_i^2(D>0)$.

The pressure-temperature phase diagram for UNi_2Si_2 has been obtained.[21,23] It is similar to the magnetic-field-temperature phase diagram and it seems that the effect of the pressure is the same as that of the magnetic field in UNi_2Si_2. Quirion *et al.* determined the pressure-temperature phase diagrams for UNi_2Si_2 and UPd_2Si_2 by measuring the resistivity and compared these phase diagrams to predictions made using a mean-filed Landau-type analysis.[23] For UPd_2Si_2 this analysis has reproduced qualitative features found in the experimental results. For UNi_2Si_2, however, this approach has been incompatible with the observed magnetic ground state. This problem is interesting and important. The molecular-field approximation for our model under the pressure is now in progress.

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