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Randomly modulated phase in a hexagonal Ising antiferromagnet

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A Monte Carlo study of a hexagonal Ising antiferromagnet reveals the occurrence of a new type of modulated phase in which domainlike regions with a partially disordered antiferromagnetic or three-sublattice ferrimagnetic spin configuration are separated by flexible ferrimagnetic walls. As the temperature is decreased, the wall region extends all over the lattice and a ferrimagnetic phase occurs. It is suggested that this modulated phase occurs in $CsCoBr_3$ and $CsCoCl_3$.

The magnetic ordering in a hexagonal antiferromagnetic Ising model has been a topic in recent years. Interest has been focused on the occurrence of an unusual magnetic ordering process which has been given to explain unusual temperature dependences of the magnetic neutron-diffraction intensities in $CsCoBr_3$ (Ref. 1) and $CsCoCl_3$ (Refs. 2 and 3) in that initially only two-thirds of the spins on each basal plane order antiferromagnetically and the remaining one-third order at a lower temperature [we call these phases a partially disordered antiferromagnetic (PDAF) phase and a ferrimagnetic (FR) phase, respectively]. An Ising model with intraplane antiferromagnetic nearest-neighbor and weak ferromagnetic next-nearest-neighbor couplings has been studied and shown to exhibit this ordering process.[4-7] These theoretical results have predicted that the specific heat shows anomalies at both the higher and lower transition temperatures. The anomaly of the specific heat, however, has been experimentally observed only at the higher transition temperature.[1] Recently, a Monte Carlo (MC) method was applied to a two-dimensional model.[8] The result showed that the intermediate (PDAF) phase does not occur in the two-dimensional model.

In this Rapid Communication, we present preliminary results of a MC simulation on the three-dimensional model. Our study is focused on the possibility of the occurrence of a new phase in an intermediate temperature range and a mechanism for how the phase, if it occurs, changes into the FR phase when the temperature is decreased. Actually, we find a new type of modulated phase in which domainlike regions are separated by FR walls. This magnetic structure changes in time, which causes a large fluctuation of the sublattice magnetizations. As the temperature is decreased, the wall region extends all over the lattice and the FR phase occurs. We discuss these problems. Some comments on experimental results for $CsCoCl_3$ are also given.

We start with the model

$$\mathscr{H} = -\frac{J_0}{2}\sum_i\sum_\lambda \sigma_{i\lambda}\sigma_{i+1\lambda} + \frac{J_1}{2}\sum_i\sum_{\lambda\mu}^{nn}\sigma_{i\lambda}\sigma_{i\mu} - \frac{J_2}{2}\sum_i\sum_{\lambda\mu}^{nnn}\sigma_{i\lambda}\sigma_{i\mu} \ ,$$

where three terms represent the interplane coupling energy, and the intraplane nearest-neighbor and next-nearest-neighbor coupling energies, respectively. The coupling constants J_0, J_1, and J_2 are assumed to be all positive. We use a MC method[9] to study this model on $N_a \times N_a \times N_c$ ($\equiv N$) rhomboid-shaped finite hexagonal lattices with periodic boundary conditions and $15 < N_a < 30$, $10 < N_c < 30$. In Fig. 1, we show the sublattices and the magnetic phases which are supposed to occur in this model. We study the following three cases with $J_2/J_1 = 0.1$: (i) $J_0/J_1 < 1$, (ii) $J_0/J_1 = 1$, and (iii) $J_0/J_1 > 1$. After about 200 MC steps per spin (MCS) are discarded, data for L (3000 or sometimes 8000) MCS are kept for calculating averages. Average values of the energy and the sublattice magnetizations are determined directly, and those of the specific heat and the standard deviations are determined from the fluctuations. In all cases, we obtain qualitatively the same results. Our results do not depend so much on the size of the lattice except for those in case (iii) in which we need to treat the lattice with $N_c > N_a$. Here, we present only the results in case (ii).

In Fig. 2, we present the results of the specific heat. We see that the specific heat depends little on the size of the lattice and exhibits a divergent peak at T_N ($\simeq 1.8J_1/k$) and a hump around T_S ($\simeq 0.7J_1/k$). This shows that an ordered phase occurs below T_N. This result is different from that obtained in the two-dimensional model in which a hump occurs at a higher temperature and a divergent peak at a lower temperature.[8] Below T_N, the hexagonal lattice seems to be divided into three equivalent sublattices.

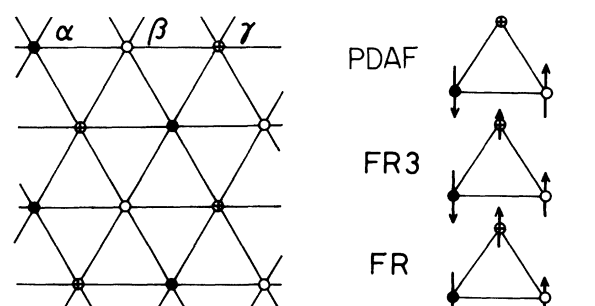

FIG. 1. Sublattices in the c plane and the magnetic phases. A three-sublattice ferrimagnetic (FR3) phase temporarily appears before the FR phase is realized.

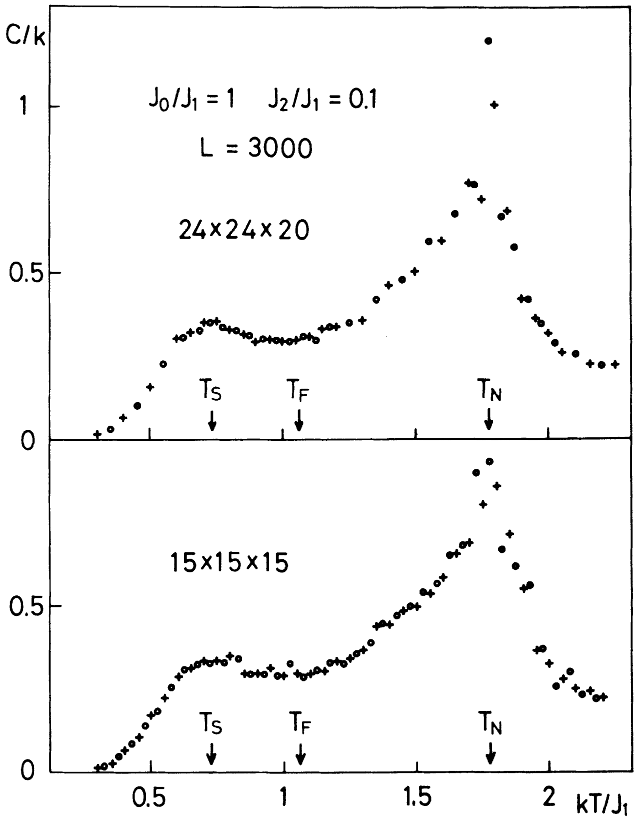

FIG. 2. Temperature dependences of the specific heat. The results determined from the fluctuations are indicated by + and those from the gradient of the energy by ○.

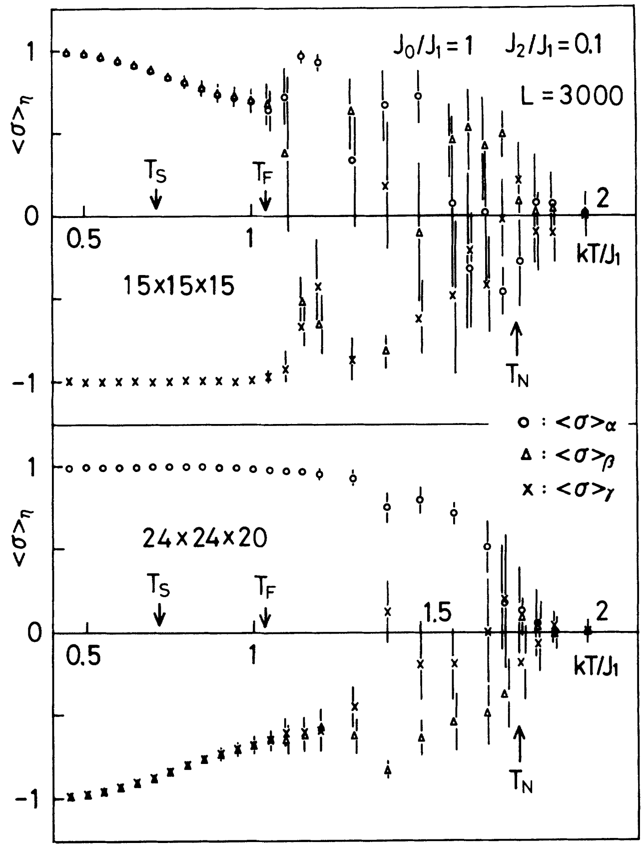

FIG. 3. Temperature dependences of the sublattice magnetizations and their standard deviations (vertical bars). Note that, for a fixed L, σ_η^t's reduce as N increases because they need a larger MCS to reverse all spins in a larger sublattice (see Fig. 4). Contrary to this, σ_η^s's increase with N. These also suggest the occurrence of the RMP discussed in the text.

Hence we define these as the sublattices of this model and refer them to α, β, and γ sublattices (see Fig. 1). In Fig. 3, we plot the averaged sublattice magnetizations per spin $\langle\sigma\rangle_\eta$ and their standard deviations σ_η^t as functions of T, where $\eta=\alpha$, β, γ. In a temperature range below T_N, $\langle\sigma\rangle_\eta$'s exhibit irregular temperature dependences and σ_η^t's are very large compared with those in low temperatures. These indicate that, in this temperature range, the sublattice magnetizations fluctuate in MCS and interchange their roles. In Fig. 4, we show typical results of the temporal variations of the sublattice magnetizations, in which the fluctuations are found. If we take the averages of the sublattice magnetizations over a sufficiently large MCS, they should vanish. This type of ordered phase is quite unusual. As the temperature is decreased, the fluctuations become smaller and the usual FR phase is realized. This gradual phase change occurs around T_F ($\simeq 1.1 J_1/k$). We note that T_F does not in general coincide with T_S, because the hump of the specific heat occurs in the temperature range in which all $\langle\sigma\rangle_\eta$'s tend to saturate.

We discuss the mechanism responsible for the occurrence of the fluctuations of the sublattice magnetizations. Mekata's calculations have indicated that in the temperature range between T_N and a lower temperature, free-energy minima occur at six PDAF spin configurations and free energy barriers separating these minima are very small.[4] In this situation, an initial PDAF state may easily give way to another PDAF state and so on. Although this seems to lead to the fluctuations of the sublattice magnetizations, this is unphysical because it needs a very long time to reverse all spins in a large sublattice. If, however, the lattice is divided into many domains, the change in the magnetic structure can easily occur. This multidomain structure may be possible only when the domain-wall free energy is very small. Calculations of free energies of several types of domains in this model have been done by one of the authors (F.M.) and the results have suggested that this multidomain structure is, in fact, possible.[10] If it is true, we may find this structure in our results. To see this, we study a temporal spin structure and the standard deviations of averaged values ($L=100$ MCS) of individual spins σ_η^s which are measures of spatial inhomogeneousness. The results are shown in Figs. 4 and 5. In fact, we find complex domain-like regions and very large values of σ_η^s (we call these regions domains). We note that the domain structure shown in Fig. 5 changes from plane to plane and a similar domain structure can be found in a plane perpendicular to the c plane.

We have seen that in this phase the lattice is divided into domains. This phase is analogous to modulated phases occuring in the axial next-nearest-neighbor Ising model[11-13] and in an Ising model on the hexagonal lattice with $J_0>0$, $J_1>0$, and $J_2<0$ in the presence of the magnetic field.[14] These modulated phases are characterized by periodically ar-

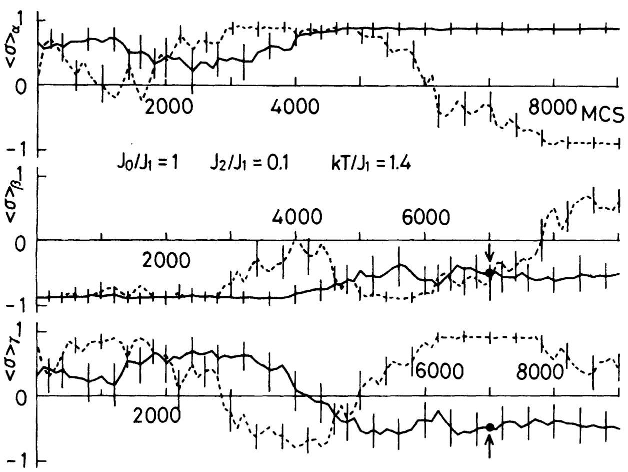

FIG. 4. Time variations of the sublattice magnetizations and the standard deviations σ_η^s (vertical bars). Solid and broken curves indicate those in the lattices with $30\times30\times20$ and $15\times15\times15$, respectively.

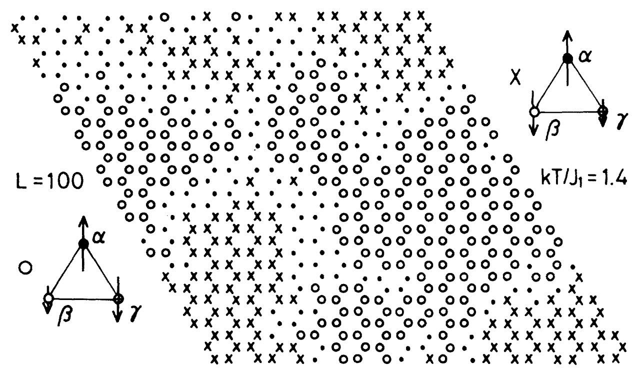

FIG. 5. Typical results of the temporal spin structure on a c plane. This result is one occurring in the case shown by arrows in Fig. 4 (spin configurations on β and γ sublattices in the lattice with $30\times30\times20$). The symbols ○ and × indicate the FR3 domain regions ($|\langle\sigma_\lambda\rangle_\eta - [\langle\sigma_\lambda\rangle_{\eta'}]_{av}| > 0.2$, where $\eta=\beta$ or γ, $\eta'=\gamma$ or β, and $[\langle\sigma_\lambda\rangle_{\eta'}]_{av} = \frac{1}{3}\sum_\Delta^{nn}\langle\sigma_{\lambda+\Delta}\rangle_{\eta'}$) and dots indicate the wall region.

ranged domain walls which separate commensurate regions. As seen in Fig. 5, however, in our model we cannot find any periodicity of the arrangement of the domains. From these facts, we suggest that in our model there occurs a new type of modulated phase in which the domains are separated by randomly arranged flexible domain walls.[15] We tentatively call this phase a randomly modulated phase (RMP). The occurrence of this RMP is not supposed to be due to the disappearance of the domain-wall free energy but to the thermal fluctuations of the domain walls (or wall wanderings).[10]

We note that the RMP is an ordered phase because the specific heat diverges at T_N. The occurrence of a long-range order in this phase may be recognized as follows. As seen in Fig. 5, two neighboring domains have restricted spin configurations. This means that, at least in one sublattice, the spins in the neighboring two domains are strongly correlated (the spins in the α sublattice in Fig. 5). The spins in each domain are, of course, correlated. Thus we may find paths of the correlations extending all over the lattice. We will discuss this problem in more detail in a future paper.

As the temperature is decreased, the FR wall region extends all over the lattice and the FR phase occurs. These phase changes are continuous, so that no anomaly of the specific heat occurs. This seems to explain the experimental result on $CsCoBr_3$.[1]

We give some comments on experiments on $CsCoCl_3$ which have been done to give experimental evidences of the occurrence of the PDAF phase. In the temperature range in which the PDAF phase is supposed to occur, neutron diffuse scattering has been observed.[3] This shows that the magnetic structure markedly fluctuates in time. Magnetic Raman scattering studies[16,17] and NMR studies[18] have revealed the occurrence of the PDAF phase. Contrary to this result, measurements of magnetoelectric effects have given a doubtful result about the occurrence of a single domain PDAF phase.[19] This difference may give experimental evidence of the occurrence of the RMP. In conclusion, we suggest that the intermediate phase occurring in $CsCoCl_3$ (and also $CsCoBr_3$) is the RMP.

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