Spin melting and refreezing driven by uniaxial compression on a dipolar hexagonal plate

Spin melting and refreezing driven by uniaxial compression on a dipolar hexagonal plate

K Matsushita[1]**, R Sugano**[2]**, A Kuroda**[3]**, Y Tomita**[3] **and H Takayama**[3]

[1] National Institute for Materials Science, 1-2-1 Sengen, Tsukuba, Ibaraki 305-0047, Japan
[2] Advanced Research Laboratory, Hitachi, Ltd, 1-280 Higashi-Koigakubo, Kokubunji-shi, Tokyo 185-8601, Japan
[3] Institute for Solid State Physics, University of Tokyo, 5-1-5 Kashiwanoha, Kashiwa, Chiba 277-8581, Japan

E-mail: MATSUSHITA.Katsuyoshi@nims.go.jp

Abstract

We investigate the freezing characteristics of a finite dipolar hexagonal plate by Monte Carlo simulation. The hexagonal plate is cut out from a piled triangular lattice of three layers with FCC-like (ABCABC) stacking structure. In the present study an annealing simulation is performed for a dipolar plate uniaxially compressed in the direction of layer-piling. We find spin melting and refreezing driven by the uniaxial compression. Each of the melting and refreezing corresponds one-to-one with a change of the ground states induced by compression. The freezing temperatures of the ground-state orders differ significantly from each other, which gives rise to the spin melting and refreezing of the present interest. We argue that these phenomena are originated by a finite size effect combined with a peculiar anisotropic nature of the dipole–dipole interaction.

1. Introduction

In recent years, systems consisting of arrayed single-domain ferromagnetic nanoparticles have received much attention from many researchers because of the possibility of their use as a high storage density material [1]. In such array systems the dipole–dipole interaction is considered to play an important role on their magnetic properties. The interaction not only intrinsically involves frustration, i.e., the competition between ferromagnetic and antiferromagnetic interactions, but also it connects the anisotropy in the spin space with that in the real space. In finite systems the latter is expected to yield a vast variety of ground-state spin configurations depending on the shapes of the systems. Various ordered spin patterns have been actually observed, for example, in infinite [2–4] or finite [5, 6] systems only with the dipole–dipole interaction, which we call *dipolar systems*, on the triangular lattice. In dipolar systems such a peculiar phenomenon as the from-edge-to-interior freezing has also been found
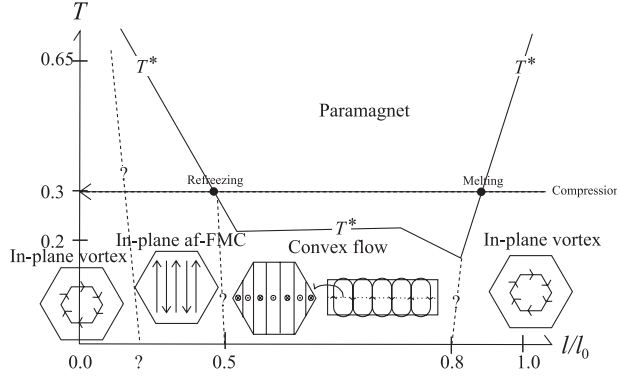
Figure 1. The schematic spin-pattern and T^* diagram of the dipolar hexagonal plate. Arrows in insets show directions of spins.

recently [7]. In the present study we investigate a dipolar hexagonal plate cut out from a piled triangular lattice of three layers with FCC-like structures and with uniaxial compression in the direction of layer-piling. Our aim here is to demonstrate that, in the dipolar system even with such a restricted shape, an interesting crossover is predicted to exist when the interlayer distance l is changed.

Figure 1 shows a schematic spin-pattern and T^* diagram obtained by the present study of the dipolar hexagonal plate, where T^* is the temperature at which spins start to be frozen. At low temperatures there appear three kinds of pattern, i.e., in-plane-vortex, convex-flow, in-plane-af-FMC (anti-ferromagnetic ferromagnetic-chain) (see figure 3(c)) patterns depending on the value of l. The freezing temperature, T^*, depends on the spin pattern of the corresponding ground state. In particular, T^* of the convex flow state is much lower than those of other states. Because of this l-dependence of T^*, we expect the peculiar spin melting and refreezing driven by uniaxial compression, say, at $T = 0.3$.

In the present study we perform simulated annealing of the dipolar hexagonal plate by the Monte Carlo (MC) method. The well-known Hamiltonian of a dipolar system,

$$H = \frac{D}{2a^3} \sum_{\langle i,j \rangle} \vec{S}_i \cdot \frac{1 - \vec{e}_{ij} \otimes \vec{e}_{ij}}{r_{ij}^3} \cdot \vec{S}_j, \tag{1}$$

is employed. Here the $\vec{S}_i$ denote classical Heisenberg spins which represent magnetic moments of nanoparticles, D is a coupling constant, and a a lattice constant of the triangular lattice. The number of piled layers in a plate examined, N_L, is 3. Each layer contains about 100 sites. The z-direction is set perpendicular to the layers. We perform typically eight cooling runs with a fixed cooling rate, in which we use different sets of random numbers generating their initial configurations as well as spin-renewing for each MC step. For each cooling run, the temperature T is initially set to about 1 in units of D/a^3 and is decreased by a step of $\Delta T = 0.0025$. At each temperature, states are sampled over $\tau = 5 \times 10^4$ MC steps. The results described below are insensitive to this cooling rate in the sense that values of the freezing temperature T^*, for example, do not change significantly even when the cooling rate is set to be several times faster.

In the present paper we examine a freezing parameter, $\mathcal{S}$, in order to study the freezing characteristics of the dipolar plate. It is defined as

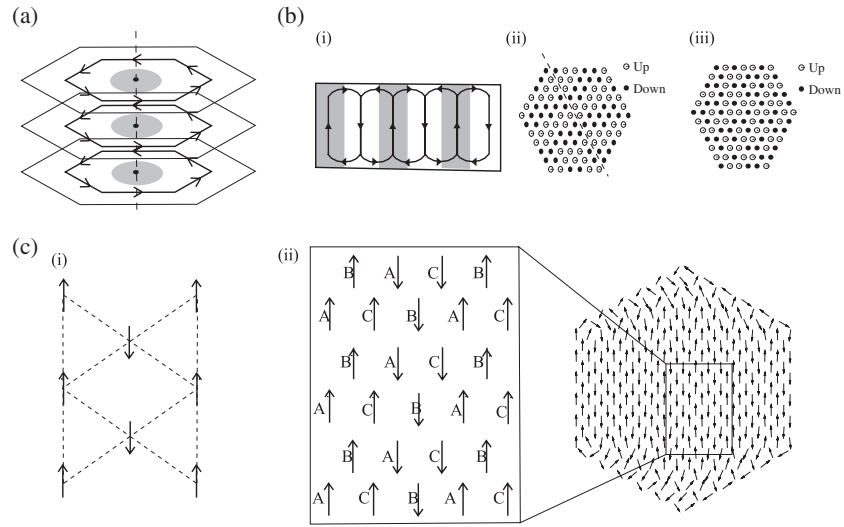$$\mathcal{S} = \frac{1}{N} \sum_i [|\langle \vec{S}_i \rangle|]_r, \tag{2}$$

Figure 2. (a) The temperature dependence of the freezing parameter for each *l*. (b) The freezing parameter as a function of *l* at $T = 0.3$.
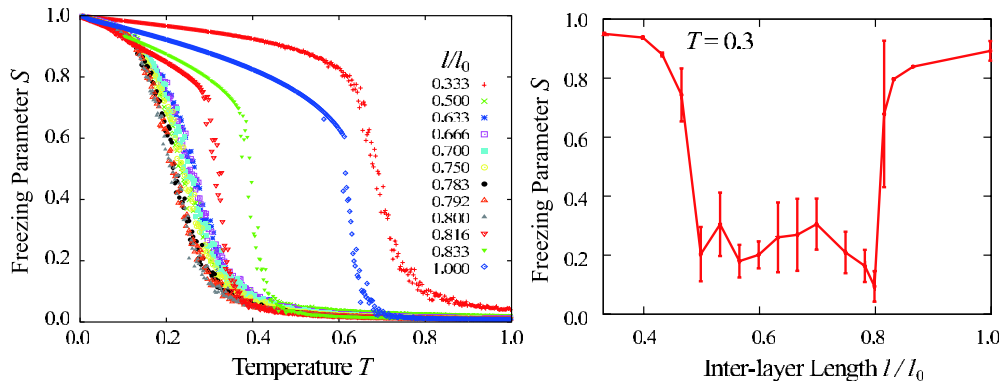

Figure 3. The ground state for (a) $0.8l_0 \lesssim l$ (in-plane vortex), (b) $0.5l_0 \lesssim l \lesssim 0.8l_0$ (convex flow), and (c) $l \lesssim 0.5l_0$ (in-plane-af-FMC). In each schematic picture, spin configurations are shown by arrows. The symbols A, B, and C in (c-ii) denote sites on the top, intermediate and bottom layers, respectively.

(This figure is in colour only in the electronic version)

where N denotes the number of sites, $\langle \cdots \rangle$ the thermal average over the MC steps τ, and $[\cdots]_r$ the average over cooling runs. This parameter represents the frozen degree of spins and is important as an order parameter of dipolar systems. In the present work, $\mathcal{S}$ is measured during cooling processes for a dipolar hexagonal plate whose interlayer distance l is set from $l_0/3$ to l_0, with l_0 being the interlayer distance of the FCC structure.

2. Freezing characteristics

In this section we discuss freezing characteristics based on the freezing parameter whose temperature dependence is shown in figure 2(a). Data points with each symbol represent the

values of $\mathcal{S}$ for each fixed interlayer distance, l. At $T \simeq T^*$, $\mathcal{S}$ for each l starts to increases rapidly and the specific heat (not shown) exhibits a peak. Let us examine the behaviour of T^* in order to clarify trends of the freezing characteristics (see also figure 1). At $l = l_0$, T^* is nearly equal to 0.65. It steadily decreases as l decreases from l_0. When l reaches nearly $0.8l_0$, T^* reaches its lowest value, which is nearly equal to 0.2. As l further deceases to around $0.7l_0$, T^* slowly increases. After l passes $0.7l_0$, T^* stays nearly constant up to l of about $0.5l_0$, where T^* starts to increase rapidly. When l is further decreased, T^* increases continuously.

From the above observation of figure 2(a), we can expect at least the following three ranges of l in which the freezing characteristics of the system differs significantly: $0.8l_0 \lesssim l$, $0.5l_0 \lesssim l \lesssim 0.8l_0$, and $l \lesssim 0.5l_0$. Actually, at the corresponding temperatures such as at $T = 0.3$, we observe the spin melting and refreezing respectively at $l \sim 0.8l_0$ and $l \sim 0.5l_0$ driven by uniaxial compression, as shown in figure 2(b). This spin melting and refreezing is related to changes of the corresponding ground states as shown in figure 1 and discussed in detail in the next section.

3. Ground state

Let us first discuss the spin patterns of the ground state (or the state reached by the present analysis at the lowest temperatures) of the plates with $l \gtrsim 0.8l_0$. That of $l = l_0$ is shown in figure 3(a). Spins are confined in the x–y plane and they form a single vortex. Such an in-plane vortex state is often observed in two-dimensional (2D) dipolar systems [5, 6]. In the present dipolar hexagonal plate the vortex state consists of six ferromagnetic domains extended from all the edges of the plate. In the cooling process to this ground state from-edge-to-interior freezing [7] is observed. Both the energetics of the ground state and the origin of these freezing characteristics have been discussed in our previous paper [8]. In a whole range of $0.8l_0 \lesssim l$ this vortex state is observed at lowest temperatures. When l becomes close to $0.8l_0$, however, the in-plane vortex state is gradually broken from around the centre of every hexagonal layer, which is indicated by the grey region in figure 3(a). In this region spins possess an out-of-plane component.

The ground state of the dipolar plate with $0.5l_0 \lesssim l \lesssim 0.8l_0$ is found to be a convex-flow state, shown schematically in figure 3(b-i). The z-component pattern of spins on the middle layer of the plate with $l = 0.7l_0$ is shown in figure 3(b-ii), in which a stripe pattern of ferromagnetic orders is clearly seen. In fact the absolute magnitudes of these z-components are close to unity. Spins on the first (top) and third (bottom) layers lie almost within the layers and align to connect smoothly with the neighbouring ferromagnetic orders, yielding a convex-flow pattern as a whole. At $l \simeq 0.5l_0$, the stripe pattern is destabilized to the one shown in figure 3(b-iii). We may call the latter a randomly distributed convex-flow state. Whether the range of l, where this state appears, is finite or not has not been confirmed, but so far it has not been observed at the other end of the convex-flow state, i.e., at $l \simeq 0.8l_0$.

For further compressed systems such as the one with $l = l_0/3$, an in-plane state again appears, as shown in figure 3(c). Each on-layer pattern of this state, however, has not the vortex structure but the anti-ferromagnetic ferromagnetic-chain (af–FMC) order shown in figure 3(c-i); spins on one lattice axis align ferromagnetically directed to the axis, and this ferromagnetic chain order alternates in the direction perpendicular to the axis. It is similar to one of the (continuous) ground states of the bulk dipolar system on the 2D square lattice [9]. In the present hexagonal plate, the on-layer af-FMC orders align ferromagnetically in the z-direction, as shown in figure 3(c-ii). Spins on two sides of the plate are directed within the side plane, keeping the af-FMC order with the interior spins, while those on the other four sides align to connect continuously between the af-FMC pattern on the top layer and that on the bottom

layer. The present in-plane-af-FMC ground state is confirmed in the range $0.33l_0 \leqslant l \leqslant 0.46l_0$ at least. We expect one more change of the ground state at a further shorter l than $0.33l_0$, as shown in figure 1, because at $l = 0$ the system is a 2D triangular lattice layer whose density of spins is tripled as compared with the original one, and so the vortex state is naturally expected.

4. Discussion

In our recent work [8] the same dipolar hexagonal systems but with the numbers of layers, N_L, up to 15 and with $l = l_0, l_0/\sqrt{2}$ were studied. We observed the in-plane-vortex ground state in systems with $l = l_0$ $(l_0/\sqrt{2})$ for all N_L $(N_L \leqslant 2)$, and the convex-flow ground state for $N_L \geqslant 3$ in systems with $l = l_0/\sqrt{2}$. In a bulk system with $l = l_0$, i.e., the FCC lattice, the ferromagnetic order has the global O(3) symmetry [10]. Therefore, when the hexagonal boundary is introduced, there appears the in-plane-vortex ground state which gains the boundary-induced anisotropic energy including that from all edges. When the FCC structure is compressed in the z-direction, the magnetization prefers to align to this direction, and competition with the boundary-induced anisotropic energy takes place. An important finding of the present work is that the critical distance, l_{cr}, at which the ground state changes from the in-plane-vortex to the convex-flow ones, is given as $l_{cr} \simeq 0.8l_0$ for $N_L = 3$. More interestingly, the spin melting phenomenon is observed at low temperatures, such as $T \sim 0.3$, as l decreases to around l_{cr}, corresponding to the ground state transition. Furthermore, when l is further decreased, the spin refreezing phenomenon is found. It corresponds to the ground state change to the in-plane af-FMC one. The occurrence of the latter is certainly related to the fact that the present system with $l = l_0/2$ is just a simple cubic lattice, one of whose bulk ground states is the af-FMC one [10], though details of the shape effect are not completely understood yet.

The results so far described are obtained for the hexagonal dipolar plate by varying l but with N_L $(= 3)$ and a size of the plate fixed. Although we have not completed MC simulations with a systematic change of the other parameters, nor have we analysed the effects of the boundary roughness, the present results are enough to demonstrate that the dipolar systems of a finite size exhibit a vast variety of magnetic phenomena, which are hardly thought of for ordinary spin systems with only simple exchange-type interactions.

Acknowledgments

References

[1]　Martin J J, Nogues J, Liu K, Vicent J L and Schuller I 2003 *J. Magn. Magn. Mater.* **256** 449
[2]　Garel T and Doniach S 1982 *Phys. Rev.* B **26** 325
[3]　Iglesias J R, Gomçalves S, Nagel A and Kiwi M 2002 *Phys. Rev.* B **65** 064447
[4]　Jagla E A 2004 *Phys. Rev.* E **70** 046204
[5]　Belobrov P I, Voevodin V A and Ignatchenko V A 1985 *Sov. Phys.—JETP* **61** 522
[6]　Vedmedenko E I, Ghazali A and Lèvy J-C S 1999 *Phys. Rev.* B **59** 3329
[7]　Matsushita K, Sugano R, Kuroda A, Tomita Y and Takayama H 2005 *J. Phys. Soc. Japan* **74** 2651
[8]　Matsushita K, Sugano R, Kuroda A, Tomita Y and Takayama H 2006 at press
[9]　De'Bell K, MacIsaac A B, Booth I N and Whitehead J P 1997 *Phys. Rev.* B **55** 15108
[10]　Luttinger J M and Tisza L 1946 *Phys. Rev.* **70** 954