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Magnetization behavior and magnetic entropy change of frustrated Ising antiferromagnets on two- and three-dimensional lattices

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Abstract
The magnetization behavior and the magnetic entropy change of frustrated Ising antiferromagnets with spin-1/2 on two-dimensional triangular and three-dimensional hexagonal closed-packed lattices are calculated by using Monte Carlo simulation. The results indicate that the normalized magnetization as a function of external field shows a 1/3 plateau on the two-dimensional system, while it shows 0 and 1/2 plateaus on the three-dimensional system at low temperature. Consequently, it causes an inverse magnetocaloric effect, namely, the value of magnetic entropy change may be positive in some ranges of magnetic fields. This phenomenon may be used to cool in a field by adiabatic demagnetization. Moreover, we analyze the spin configurations of systems on the magnetization plateaus, and discuss the impact of the frustration. We also find a mapping of the magnetization plateaus to the magnetic entropy changes at low temperature. The study of such systems showing field-induced metamagnetic transition in relation to frustrated antiferromagnets may open an important field in searching for good candidates for room-temperature magnetic refrigeration.

1. Introduction

Since the end of the last century, magnetic refrigeration with well refrigerated performance and special applications in some areas has received much attention [1, 2]. One of the key points is searching for magnetic materials with a large magnetocaloric effect (MCE) [3, 4]. In general, the investigation of MCE is focused on ferromagnets [5, 6], and the largest MCE can occur in the vicinity of the temperature of magnetic-phase transition from paramagnetism to ferromagnetism. On the other hand, many antiferromagnetic (AF) systems showing different temperature/field-induced metamagnetic transitions may exhibit a large MCE, and the sign of magnetic entropy change may be positive. This behavior is called inverse MCE [7]. Therefore, magnetic materials with multiple magnetic-phase transitions are of importance in the search for 'table-like' MCE [8]. In 2007, Du and her colleagues found a large inverse MCE at a field-induced metamagnetic transition from a collinear to a triangular AF configuration in the antiferromagnet ε-$(Mn_{0.83}Fe_{0.17})_{3.25}Ge$ [9] and a large room-temperature MCE at a field-induced metamagnetic transition from an AF to a ferrimagnetic configuration in $Fe_{0.8}Mn_{1.5}As$ compound [10], respectively.

In recent years, theoretical investigations have predicted an enhanced MCE in frustrated classical spin systems at low temperature [11, 12]. In the frustrated YbAs compound, an inverse MCE had been reported [13]. In 2005, Sosin *et al* [14] figured out that frustration can induce a large adiabatic temperature change by studying magnetic refrigerant material $Gd_2Ti_2O_7$ on a pyrochlore lattice, and the next year Singh *et al* [15] also found thermomagnetic irreversibility in the intermetallic compound TbNiAl, which was attributed to magnetic frustration.

The concept of geometric frustration dates back to 1950. It is a common feature of condensed matter systems. Geometric frustration arises when a system cannot, because of local geometric constraints, minimize all the pairwise interactions simultaneously. The Ising model with AF nearest-neighbor interactions on the triangular lattice is the simplest spin system with total frustration; it shows that the entropy at absolute zero temperature is finite, that is, the system is disordered, violating the third principle of thermodynamics [16]. The three-dimensional (3D) hexagonal closed-packed (hcp) lattice itself also presents frustration due to geometrical topology [17]. Frustration is able to restrain the formation of long-range AF order, whereas it is in favor of the formation of a non-Néel state for the spin-$1/2$ system. In 1988, Selke analyzed the phase diagrams and experimental applications of the one-, two- and three-dimensional axial next-nearest-neighbor Ising (or ANNNI) models in detail [18]. He came to the conclusion that configurational entropy was shown to play the decisive role in forming spatially modulated spin patterns in this prototype model with discrete symmetry and short-range competing interactions. Moreover, several phases appeared versus different ranges of temperatures and fields, and they were stable against different kinds of perturbations at low temperature. Therefore we conjecture that systems with an ANNNI model structure are also able to enhance the MCE. Commonly, the two- and three-dimensional frustration systems can be obtained by packing side-sharing or corner-sharing structural units. Recently, scientists have paid much attention to the two-dimensional triangular corner-sharing Kagomé lattice and side-sharing hexagonal lattice, and the three-dimensional tetrahedral side-sharing face-centered cubic (fcc) lattice and corner-sharing spinel, laves planes and pyrochlore lattices. Moessner and his colleague studied systematically the ground-state and low-temperature properties of the magnets on corner-sharing (Kagomé, pyrochlore, SCGO, and GGG) lattices with strong geometric frustration [19, 20]. They found that the universal features of these magnets were traced back to a large ground-state degeneracy in model systems, which rendered them highly unstable towards perturbations.

In some cases, the frustration can be so intense that it induces novel and complex phenomena. Zhitomirsky *et al* [21] by theoretical simulation and Cao *et al* [22] by experimental measurement obtained the magnetization plateaus, respectively. Ciftja and co-workers [23, 24] also obtained a magnetization plateau at low temperature. Moreover, they found that there was an absence of rapid changes in the magnetic moment of the system that occur for $S = 1/2$ when they studied dimer, equilateral triangle, square, and regular tetrahedron arrays of spins by using an analytical method. They concluded that the rapid changes in magnetic moment versus applied magnetic field at low temperature that occurred for the case $S = 1/2$ were a direct consequence of ground-state level crossing. Blankschtein *et al* [25] by using Landau–Ginzburg–Wilson (LGW) theory and Netz *et al* [26] by using mean-field (MF) theory studied the influence of frustration in 3D and two-dimensional (2D) systems on specific heat in zero external field. They observed two peaks in the curve of the specific heat as a function of temperature. However, in their models the interlayer frustration effect was not introduced. The 'spin ice' compound with pyrochlore structure is another kind of frustrated magnetic material [27] which shows novel and complex phenomena. Using the Heisenberg and Ising models with exchange couplings, dipolar interactions and a strong easy-axis anisotropy, Moessner and Sondhi *et al* found that the application of a magnetic field along the [111] direction led to two magnetization plateaus, and between them the entropy exhibited a giant spike [28, 29].

At present, geometrically frustrated magnets are considered to be in a separate class from both unfrustrated and disordered magnets (spin glasses and the like). Therefore, an investigation of the magnetism properties of geometrically frustrated magnets will be significant. In this paper, we investigate the 'fully' frustrated antiferromagnets, and study the influence of frustration on magnetization behavior and the magnetic entropy change. The hcp lattice will be used here because it is one of the commonest and most important crystalline structures in metals [30]. We find exotic magnetization plateaus on the hcp lattice, and establish a relation between the magnetization plateaus and the magnetic entropy changes. We introduce our model in section 2, and present simulation results and a discussion in section 3. Finally, we present a summary in section 4.

2. Model and simulation method

An Ising AF model on layered triangular lattices with hcp structure is considered. Thus we can study the effect of frustration from 2D triangular to 3D hcp lattices. The Hamiltonian in an external field is,

$$H = -J_{12} \sum_{\langle i,j \rangle}^{xy} s_i s_j - J_3 \sum_{\langle i,j \rangle}^{z} s_i s_j - H \sum_i s_i, \quad (1)$$

where $\langle i, j\rangle$ indicates summation over the nearest-neighbor pairs, $s_i(s_j) = \pm 1/2$. The first term is the exchange interaction between the nearest-neighbor spins in the xy plane, J_{12} is the exchange integral and $J_{12} < 0$. The second term is the exchange interaction between the nearest-neighbor spins along the z direction, J_3 is the exchange integral and $J_3 \leqslant 0$. As $J_3 = 0$, the system is just a 2D triangular lattice model. The last term is the Zeeman energy, and H is the external field.

In the calculation, let $j_{12} = J_{12}/|J_{12}| = -1$, $j_3 = J_3/|J_{12}|$ and $h = H/|J_{12}|$ represent the reduced intralayer exchange integral, the interlayer exchange integral and the external field, respectively, and let $T_R = k_B T/|J_{12}|$ represent the reduced temperature. We will employ a single spin-flip Monte Carlo (MC) metropolis algorithm [31] with periodic boundary conditions to study the system described in equation (1). In order to check the effects of the simulation size and MC step on the magnetization, in figure 1 we give the simulation results for the magnetization as a function of external field at low temperature with different simulation sizes and MC steps.

It is found that the magnetizations are almost independent of the numbers of spins in the present size of system in figure 1(a). The reason may be that short-range interaction is considered and periodic boundary conditions are used. As shown in figure 1(b), when the number of MC steps is above 10 000, the magnetization is almost unchanged. Although finite steps in the simulation are adopted, the impact on spin states of the MC step number is very weak in the frustrated Ising AF model when the step number is sufficient [32]. Thus

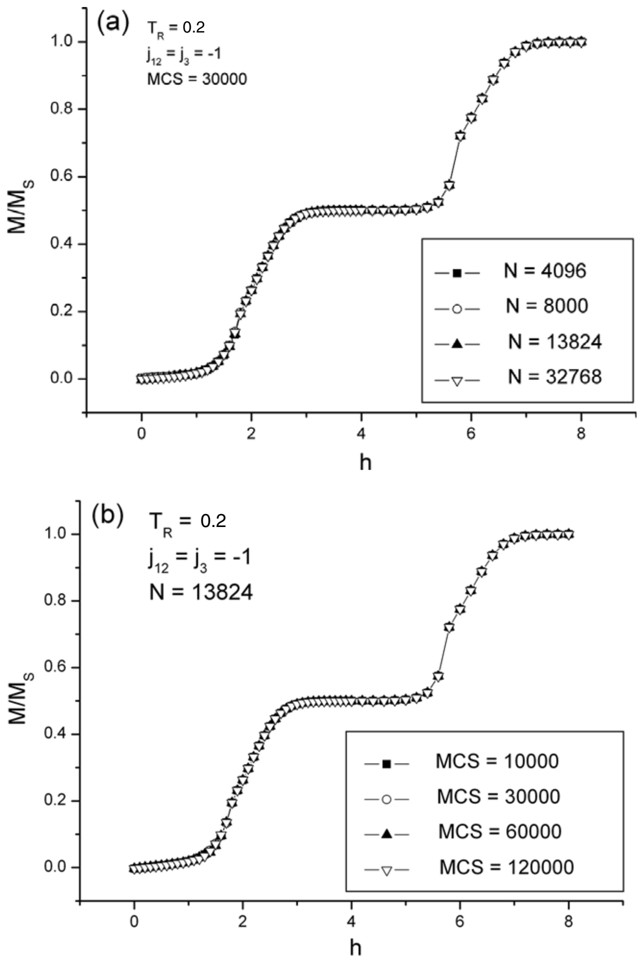

Figure 1. The normalized magnetization (M/M_S) as a function of reduced external field in the antiferromagnetic Ising model on a 3D hcp lattice with different numbers (N) of spins (a) and MC steps (b).

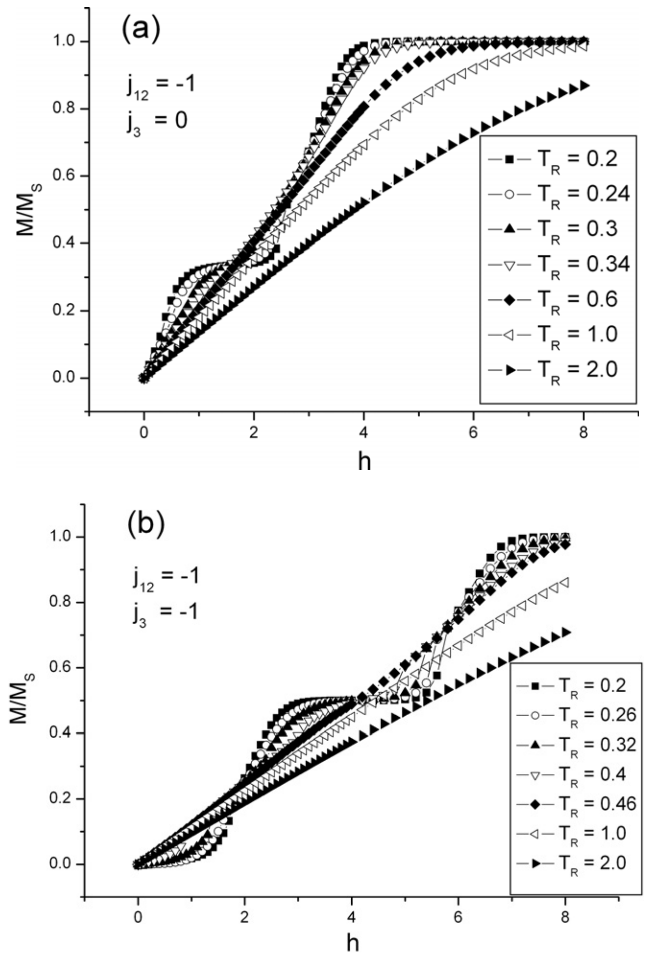

Figure 2. The normalized magnetization (M/M_S) as a function of reduced external field in the antiferromagnetic Ising model on 2D triangular (a) and 3D hcp (b) lattices at different temperatures.

in the present simulation we take the sizes of the system to be $24 \times 24 \times 24$ spins on 3D hcp and $80 \times 80 \times 2$ spins on 2D triangular lattices with periodic boundary conditions. The first 15 000 MC steps per spin are discarded for equilibrium and thermal averages are taken with the next 15 000 MC steps.

3. Results and discussion

3.1. Magnetization behavior

Now, we investigate the 2D and 3D AF Ising models by using the standard MC simulation method. Figures 2(a) and (b) show the normalized magnetization as a function of reduced external field on 2D and 3D lattices at different temperatures, respectively. From figure 2, it is found that the magnetization as a function of external field shows a plateau at 1/3 on the 2D lattice, and two plateaus at 0 and 1/2 on the 3D lattice at low temperature. With increasing temperature, the magnetization plateaus gradually shorten. Finally, the magnetization plateau on the 2D lattice vanishes at $T_R^C \approx 0.34$ and the two magnetization plateaus vanish simultaneously at $T_R^C \approx 0.46$ on the 3D lattice (figure 2). Furthermore, the higher the temperature, the stronger is the field needed to make the system saturate. The results are in good agreement with those obtained by Ciftja *et al* using the analytical method [23]. Commonly, a frustrated system is in magnetic disorder in zero external field at finite temperature, although the frustration effect can produce extra restraint on thermal fluctuation. Only in an external field can the system show an ordered structure, and the magnetization plateau phenomena may appear [21, 22].

In order to analyze directly the reason why magnetization plateaus exist, we explore the local spin configuration of an arbitrary point on the plateaus at low temperature. The spin configuration of an AF Ising model on a 2D triangular lattice at low temperature is shown in figure 3. It is observed that the lines of spins corresponding to sublattices B and C are fully frustrated in the plane, since they have three satisfied and three unsatisfied bonds. Therefore, when the system is magnetized, only two sublattices of A, B, and C will be along the direction of the external field, that is, the average magnetization value of a unit cell is 1/3. There is a period with a transverse 3 lattice constant, and a vertical $\sqrt{3}$ lattice constant. Therefore the normalized magnetization produces a 1/3 plateau macroscopically. The result is the same as one of the ground-state configurations pointed out by Coppersmith [33]. She proposed that the appearance of

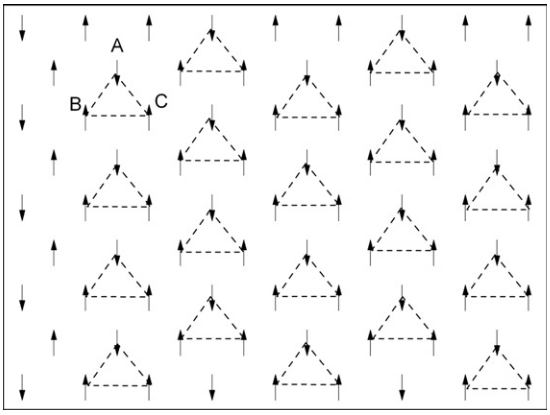

Figure 3. The spin configuration of the Ising antiferromagnet on the 2D triangular lattice at $T_R = 0.2$ in $h = 1.5$; ABC is a unit cell.

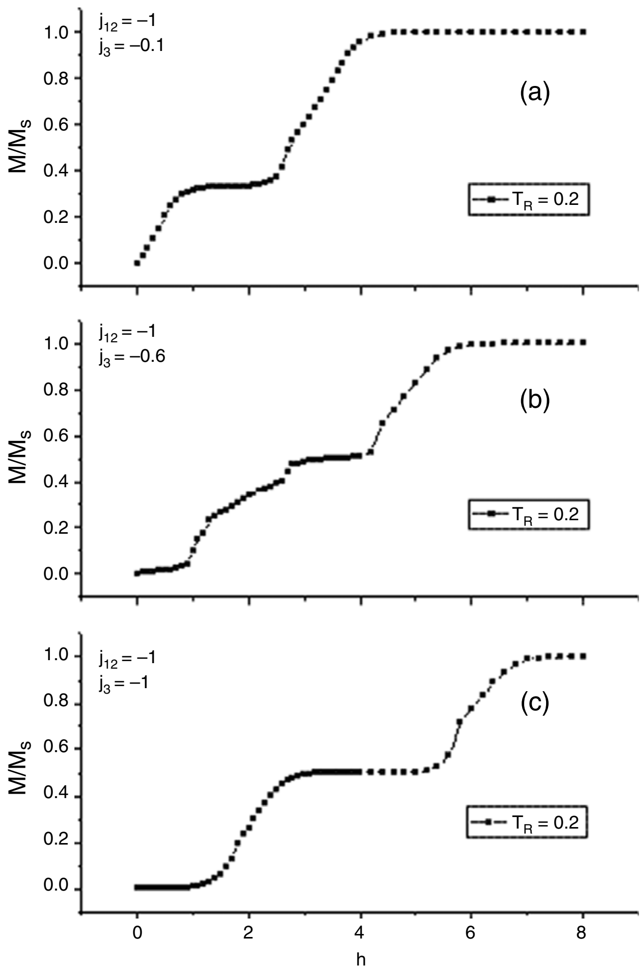

Figure 4. The normalized magnetization (M/M_S) as a function of reduced external field in the antiferromagnetic Ising model on the 3D hcp lattice with different j_3 at $T_R = 0.2$.

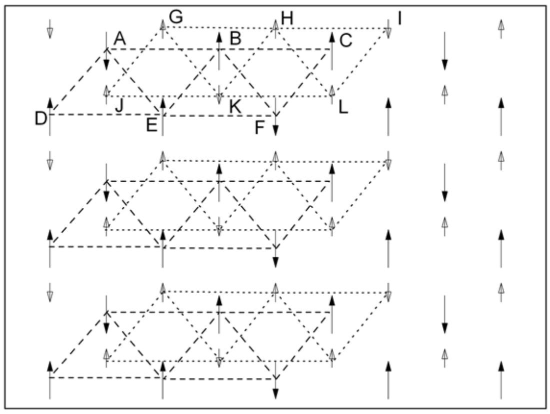

Figure 5. The spin configuration of the Ising antiferromagnet on the 3D hcp lattice with $j_3 = -0.1$ at $T_R = 0.2$ in $h = 1.5$; A–L is a unit cell, and solid and open arrows represent the first- and second-layered spins perpendicular to the paper plane, respectively.

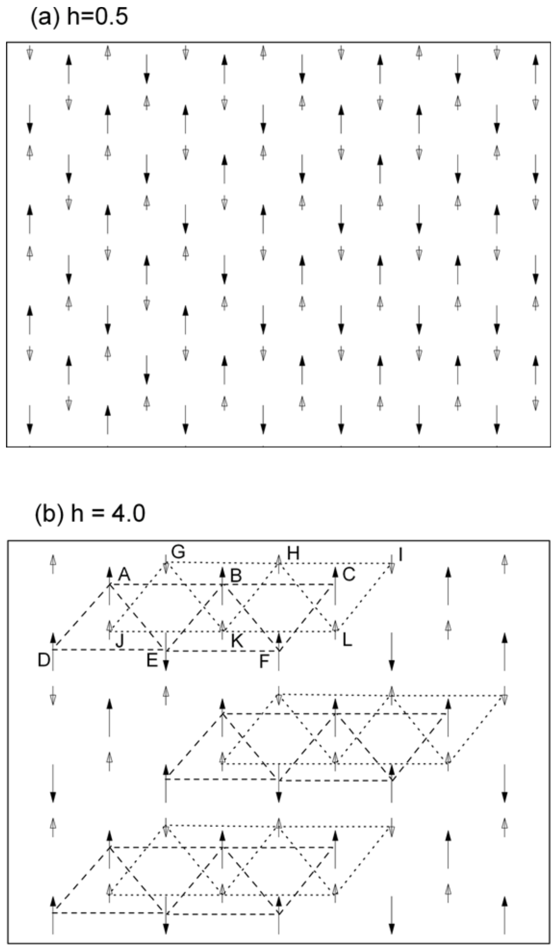

Figure 6. The spin configurations of the Ising antiferromagnet on the 3D hcp lattice with $j_3 = -1$ at $T_R = 0.2$ in weak (a) and strong (b) external fields; A–L is a unit cell in (b), and solid and open arrows represent the first- and second-layered spins perpendicular to the paper plane, respectively.

such a configuration was favored at low temperature because of entropy considerations. This state is stable within a range of external fields due to the influences of exchange interaction and frustration.

For the sake of analyzing the phenomena of magnetization plateaus on the 3D lattice, we study the magnetization behaviors of a system with different J_3 at low temperature, and the results are given in figure 4. When the interlayer exchange interaction is weak, the magnetization is still similar

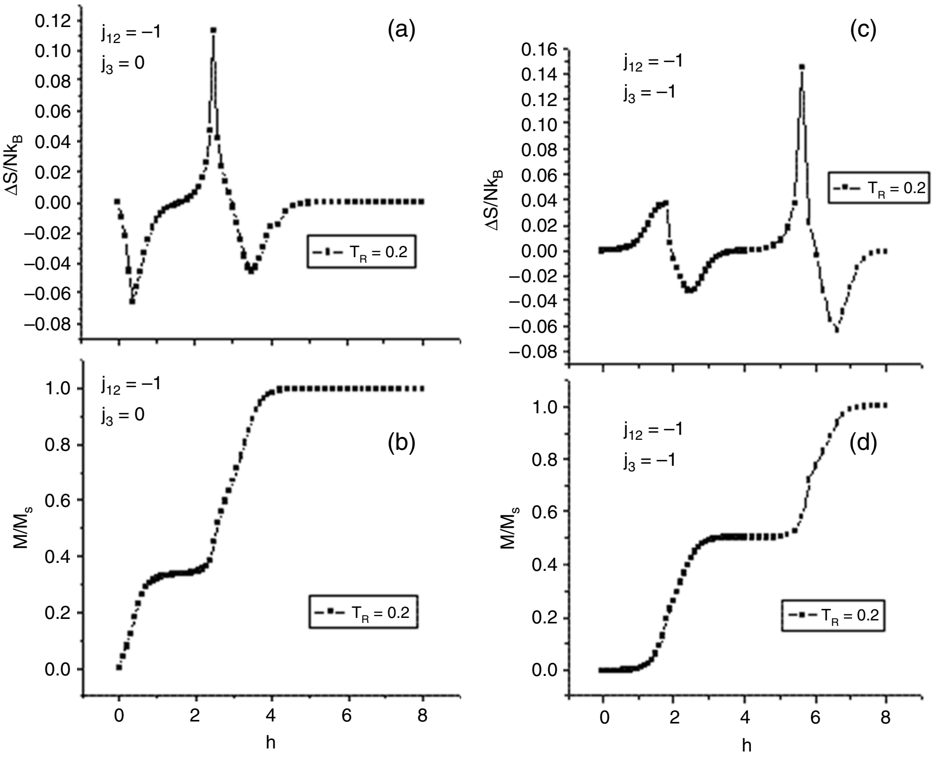

Figure 7. The magnetic entropy change per spin and normalized magnetization (M/M_S) as functions of reduced external field in the antiferromagnetic Ising models on 2D triangular (a), (b) and 3D hcp (c), (d) lattices.

to that on the 2D lattice, that is, it exhibits a magnetization plateau. Its spin configuration is shown in figure 5. There is a period with a transverse 3 lattice constant, a vertical $\sqrt{3}$ lattice constant and a $2\sqrt{6}/3$ lattice constant perpendicular to the paper plane. Since the average magnetization value of a unit cell A–L is also 1/3, the normalized magnetization shows a 1/3 plateau macroscopically. It is observed that the magnetization plateau at 1/3 becomes obscure and two new plateaus at 0 and 1/2 gradually appear with an increase in $|j_3|$. Finally, the magnetization plateau at 1/3 vanishes entirely. Instead, the two new magnetization plateaus at 0 and 1/2 are stabilized.

The local spin configurations of two arbitrary points on the magnetization plateaus on a 3D lattice with $j_3 = -1$ at low temperature are given in figure 6. From figure 6(a), it is found that the spin configuration is disordered in a weak external field, leading the value of normalized magnetization being zero. The reason may be that the 3D AF exchange interaction is strong enough, which makes the Zeeman energy unavailable to overcome it to make the spins align along the direction of the external field. The system is still in the frustrated state, that is, the spin configuration is disordered in the weak external field, and the magnetization is zero, forming the first plateau at 0 macroscopically. However, in a strong external field the spin configuration becomes ordered again, as shown in figure 6(b), and forms a period with a transverse 2 lattice constant, a vertical $2\sqrt{3}$ lattice constant and a $2\sqrt{6}/3$ lattice constant perpendicular to the paper plane. The average magnetization value of a unit cell A–L is 1/2, and as a result the value of normalized magnetization is 1/2, that is, producing the second plateau at 1/2 macroscopically. Based on the analyses above, the pattern of field-induced metamagnetic transition has been changed due to the introduction of an interlayer exchange integral. There are two phase-transition points in the range of fields with a strong interlayer exchange integral at low temperature, and the feature is analogous to phase versus field at low temperature in the ANNNI models [18].

The phenomena of magnetization plateaus on 2D and 3D antiferromagnets show that different magnetic phases are formed with an increase in external field at low temperature. However, the field-induced magnetic-phase transitions vanish when the temperature is high enough, as shown in figure 2.

3.2. Magnetic entropy change

In order to investigate the influence of frustration on the magnetic entropy change, we use the thermodynamic Maxwell equation,

$$\left(\frac{\partial S}{\partial H}\right)_T = \left(\frac{\partial M}{\partial T}\right)_H, \quad (2)$$

where S denotes magnetic entropy, T denotes temperature, and M denotes the total magnetic moment in the external field H. Thus the magnetic entropy change $\Delta S(T, H)$ of the system from zero field to a field of H can be derived from the above equation by integrating over the magnetic field:

$$\Delta S(T, H) = \int_0^H \left(\frac{\partial M}{\partial T}\right)_H \mathrm{d}H \approx \sum_i \left(\frac{\partial M}{\partial T}\right)_{H^i} \Delta H^i$$
$$= k_B \sum_i \left(\frac{\partial M}{\partial T_R}\right)_{h^i} \Delta h^i. \quad (3)$$

It is found from equation (3) that the greatest magnetic entropy change is related to the speed of variation of magnetization with temperature. Because materials with a considerable magnetic entropy change over a temperature span can be considered as practical refrigerants, research on magnetic entropy change is of great importance to finding potential magnetic refrigerant materials.

Figures 7(a) and (c) show the magnetic entropy changes per spin as a function of reduced external field at low temperature on 2D and 3D lattices, respectively; the corresponding magnetization behaviors are shown in figures 7(b) and (d). In a common sense, the thermal fluctuation energy of spins increases with increasing temperature, making the system difficult to magnetize in an external field, that is, ΔM is usually negative as ΔT is positive in a finite external field. Therefore the magnetic entropy change is usually negative; in other words, the system is exothermic as an external field is applied. However, in the present system the magnetic entropy change as a function of external field may be positive, as shown in figures 7(a) and (c); an endothermic phenomenon appears in an external field. We can find clues from the magnetization process to analyze this anomalous phenomenon. As shown in figure 7, it is found that the value of external field at which the magnetization plateau begins just corresponds to the place where the magnetic entropy change achieves a negative maximum, and the value at which the magnetization plateau ends just corresponds to the place where the positive magnetic entropy change begins. The reason may be that the variations in magnetization as a function of external field become extrema in the vicinity of the beginning and end of the magnetization plateaus. In helimagnetic dysprosium, a positive magnetic entropy change was found at 174 K in a weak magnetic field [4], and in 'spin ice' it has also been found that the entropy exhibits a giant spike between two plateaus [29]. However, in [29], the intuitive corresponding relation between plateaus and magnetic entropy changes was not presented theoretically. That is, the beginning and end of magnetization plateaus on 2D or 3D lattices map on to the negative maximum and positive starting points of magnetic entropy changes, respectively. Du *et al* in their letters discussed that the occurrence of a magnetically inhomogeneous state near the transition temperature is because the first-order magnetic transition leads to the presence of mixed magnetic exchange interaction [9]. According to the discussion above, we have deduced that the phenomena of magnetization plateaus and positive magnetic entropy changes exist due to frustration, and that the magnetization plateau and the magnetic entropy change have a corresponding relationship. Moreover, we conclude that, due to the presence of frustration and magnetic exchange interaction, the application of an external magnetic field also leads to further spin disorder in our systems, which makes the magnetic entropy increase.

The magnetic entropy changes per spin as a function of temperature in different external fields on 2D and 3D lattices are shown in figures 8(a) and (b), respectively. It is found that in different ranges of external fields the magnetic entropy change exhibits a positive or negative maximum in the frustrated system at low temperature. However, as the temperature is elevated by some extent,

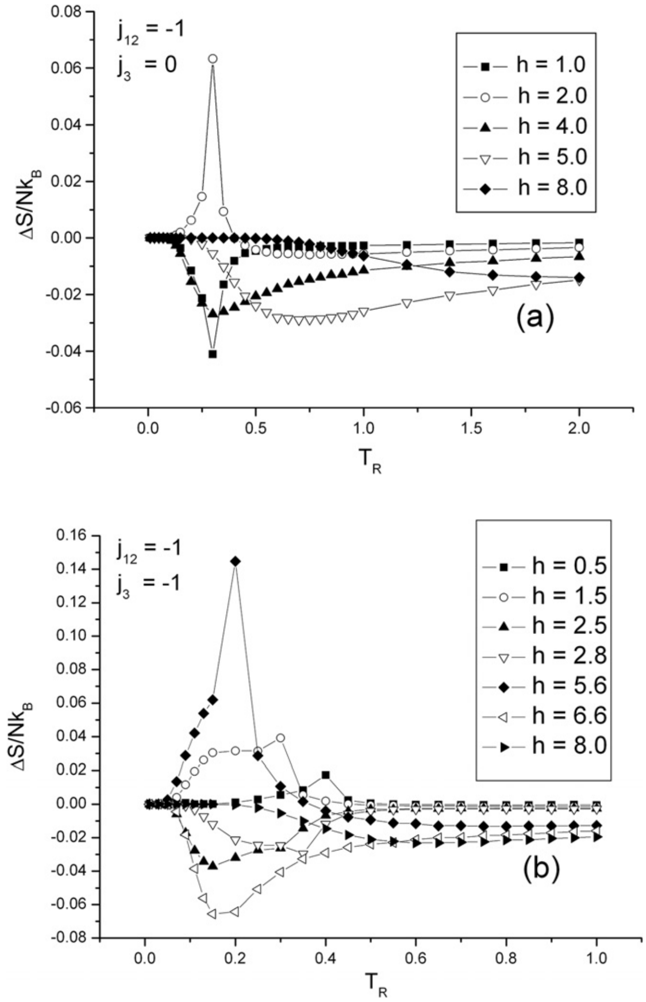

Figure 8. The magnetic entropy change per spin as a function of reduced temperature in the antiferromagnetic Ising model on the 2D triangular (a) and 3D hcp (b) lattices.

the positive magnetic entropy change no longer appears. The extrema of the magnetic entropy change appear here due to the field-induced transition from a frustrated phase to a paramagnetic phase entirely, which is different from the common temperature-induced phase-transition from ferromagnetism/antiferromagnetism to paramagnetism.

4. Conclusions

In conclusion, the Ising antiferromagnets on 2D triangular and 3D hcp lattices with geometrical frustration have been studied. We discuss the magnetization behavior at low temperature. The results indicate that the normalized magnetization as a function of reduced external field shows a $1/3$ plateau on the 2D system, while it shows 0 and $1/2$ plateaus on the 3D system at low temperature. In order to explain the phenomena, we present and analyze the microstructure of spins of the system, and study the change in magnetization behavior and the spin configuration, as the system varies from two dimensions to three dimensions. In the next part, we investigated the magnetic entropy change as a function of external field and temperature. It is observed that the values of magnetic entropy change may be positive at low temperature, which is contrary to common sense. The anomalous phenomena are related to the magnetization plateaus; the beginning of a magnetization plateau corresponds to the negative maximum of a magnetic entropy change, and the end of a magnetization plateau just corresponds to the start of a positive magnetic entropy change. The mapping of the magnetization plateaus on to magnetic entropy changes indicates that frustration plays a crucial role. At zero temperature, the ground state of a frustrated system is degenerate, and the entropy is finite, thus spontaneous magnetization is absent. However, at finite temperature, the specific heat exhibits two peaks. In an external field, the magnetization exhibits plateaus and a positive magnetic entropy change appears. The degeneracy of the state may be the main reason. The study of systems with frustrated antiferromagnetic phases may open up an important field in searching for new materials for magnetic refrigeration.

Acknowledgments

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