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Slow relaxation process in Ising-like Heisenberg kagome antiferromagnets due to macroscopic degeneracy in the ordered state

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Abstract
We study relaxation phenomena in the ferromagnetically ordered state of the Ising-like Heisenberg kagome antiferromagnets. We introduce the *weathervane loop* in order to characterize macroscopic degenerate ordered states and study the microscopic mechanism of the slow relaxation from a viewpoint of the dynamics of the weathervane loop configuration. This mechanism may give a possible origin of the slow relaxation reported in recent experiments.

1. Introduction

Due to the frustration among the magnetic interactions, antiferromagnetic spin systems on triangular, kagome, or pyrochlore lattices show very interesting properties of magnetic ordering; this is the case in particular in the kagome lattice and the pyrochlore lattice, which are so-called corner-sharing lattices in which neighbouring local triangles share a spin but not a bond. There are various candidate materials of this type, for example, $Rb_2M_3S_4$ [2–5], (M is a magnetic ion such as Ni, Co, Mn), $AM_3(OH)_6(SO_4)_2$, (A^+ is a cation such as K^+, Rb^+, NH_4^+, Tl^+, Ag^+ or Na^+, and M^{3+} is a magnetic ion such as Fe^{3+}, or Cr^{3+}) [6–12], $NaFe_3(SeO_4)_2(OH)_6$ [13], $SrCr_{9x}Ga_{12-9x}O_{19}$ (SCGO) [7], and ^3He on a sheet of graphite [14]. In such systems frustration causes macroscopic degenerate states in the ground state even for the continuous spin systems, e.g. XY, and Heisenberg systems. Thus, there is no phase transition at finite temperatures in the kagome antiferromagnets with Ising, XY and Heisenberg spin systems. However, Kuroda and Miyashita pointed out that the kagome antiferromagnet has a magnetic phase transition of the universality class of the ferromagnetic Ising model [1], when the system has finite easy-axis anisotropy, namely Ising-like Heisenberg interaction. There the ordered state is still macroscopically degenerated, and it is called an exotic ferromagnetic phase. Recently slow relaxation phenomena have been found in the kagome and pyrochlore lattices [6, 7]. In this paper we consider the origin of the slow relaxation

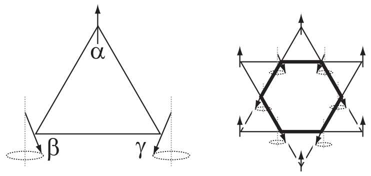

Figure 1. The spin configuration of the ground state in a triangle cluster and an example of the weathervane loop (bold line).

from the viewpoint of structural change of the macroscopically degenerate ordered states of the Ising-like Heisenberg kagome antiferromagnet. Maegawa *et al* discovered the two cusps in the temperature dependence of the susceptibility in $NH_4Fe_3(OH)_6(SO_4)_2$ [8]. They concluded that the successive phase transition may be caused by a small Ising-like anisotropy in the Heisenberg kagome antiferromagnet.

In recent experimental studies, the antiferromagnetic kagome compounds show slow relaxation of magnetization and dynamical susceptibility. Usually, the slow dynamics is caused by random interaction of the systems. However, slow relaxation appears in non-random spin systems such as $SrCr_{9x}Ga_{12-9x}O_{19}$ (SCGO) and $AM_3(OH)_6(SO_4)_2$, which are corner-sharing structures.

In the present paper, as a candidate of the origin of the slow relaxation in non-random systems, we study relaxation phenomena in kagome antiferromagnetic systems in the macroscopically degenerate ordered state of the Ising-like Heisenberg antiferromagnetic kagome systems. In order to characterize the degenerate state we introduce a *weathervane loop*, and investigate the microscopic mechanism of the slow relaxation by considering the relaxation of the configuration of the weathervane loop structure, which is much slower than the relaxation of the total magnetization which is the macroscopic order parameter of the model.

In section 2, we review the phase transition and features of the Ising-like Heisenberg antiferromagnetic kagome systems. In section 3, we consider relaxation processes of a number of 'weathervane loops'. In section 4, we conclude our research.

2. Model

We consider the Ising-like Heisenberg kagome antiferromagnetic system,

$$\mathcal{H} = J\left(\sum_{\langle i,j\rangle} S_i^x S_j^x + S_i^y S_j^y + A S_i^z S_j^z\right), \qquad J > 0 \text{ and } A > 1, \tag{1}$$

where $\langle i, j\rangle$ and A denote the nearest neighbour in the kagome lattice and Ising-like anisotropy, respectively. The kagome system consists of triangle units that share one corner. The ground state of an Ising-like Heisenberg triangle unit is given by $S_\alpha = (0, 0, 1)$, $S_\beta = (s, 0, -c)$, $S_\gamma = (-s, 0, -c)$, where $c = \frac{A}{A+1}$ and $s = \sqrt{1-c^2}$. The freedom of rotation 2π in the xy-plane remains in S_β and S_γ. Because the antiferromagnetic kagome lattice system has a large number of degenerate states, it is important to consider the entropy of the spin configuration. If we connect $\{S_\beta\}$ and $\{S_\gamma\}$ in the lattice, we find a closed loop as shown in figure 1. We call the line a *weathervane loop*.

This system has nonzero magnetization in the ground state, the value of which is $1 - 2c$ in each triangle unit. It is important to note that no sublattice long-range order exists in this system in spite of the existence of the magnetic phase transition.

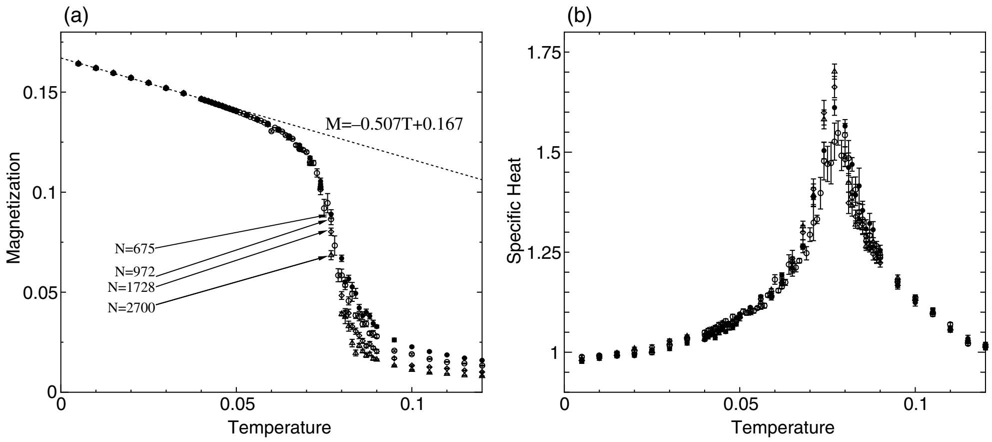

Figure 2. Temperature dependence of (a) the magnetization and (b) the specific heat for $A = 3$. (● for $N = 675$, ○ for $N = 972$, ◊ for $N = 1728$, and △ for $N = 2700$.)

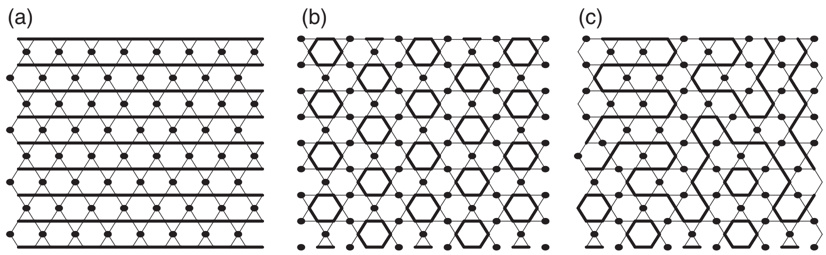

Figure 3. A typical example of the ground state of the anisotropic Heisenberg kagome system. (a) $q = 0$ state, (b) $\sqrt{3} \times \sqrt{3}$ state, and (c) the random ground state. The circles and thick lines denote S_α and the weathervane loop, respectively.

In order to study the equilibrium properties, we use the heat bath method of Monte Carlo simulations with 100 000 Monte Carlo steps (MCSs) for initial relaxation and 100 000 MCSs for collecting data. For the region near the critical point, we perform 200 000 MCSs for initial relaxation and 500 000 MCSs for measurement. Figures 2(a) and (b) show the temperature dependence of the magnetization and the specific heat for $A = 3$, respectively.

In figure 2(a) we see that, when $T \to 0$, the magnetization approaches the value of the ground state, $\frac{1}{3}\frac{-A+1}{A+1} = -\frac{1}{6}$. Around $T \simeq 0.078$, the magnetization changes suddenly and the specific heat diverges, which indicates a second-order phase transition. This is the phase transition which belongs to the two-dimensional Ising ferromagnetic universality class [1].

3. Weathervane loop and defects

In the ground state, there are macroscopic degenerate configurations of the weathervane loop as depicted in figures 3(a)–(c). Figures 3(a) and (b) show the ground states which are called the $q = 0$ and $\sqrt{3} \times \sqrt{3}$ structure, respectively. On the other hand, in figure 3(c), we show a configuration which is obtained by quenching the system from a high temperature; we call it a 'random structure'. Let us consider the degeneracy of the ground state. Each loop has a rotation degree of freedom 2π. We denote the number of the degeneracy of the ground state $(2\pi)^{n_{\rm loop}}$, where $n_{\rm loop}$ denotes the number of weathervane loops. In the spin configuration of

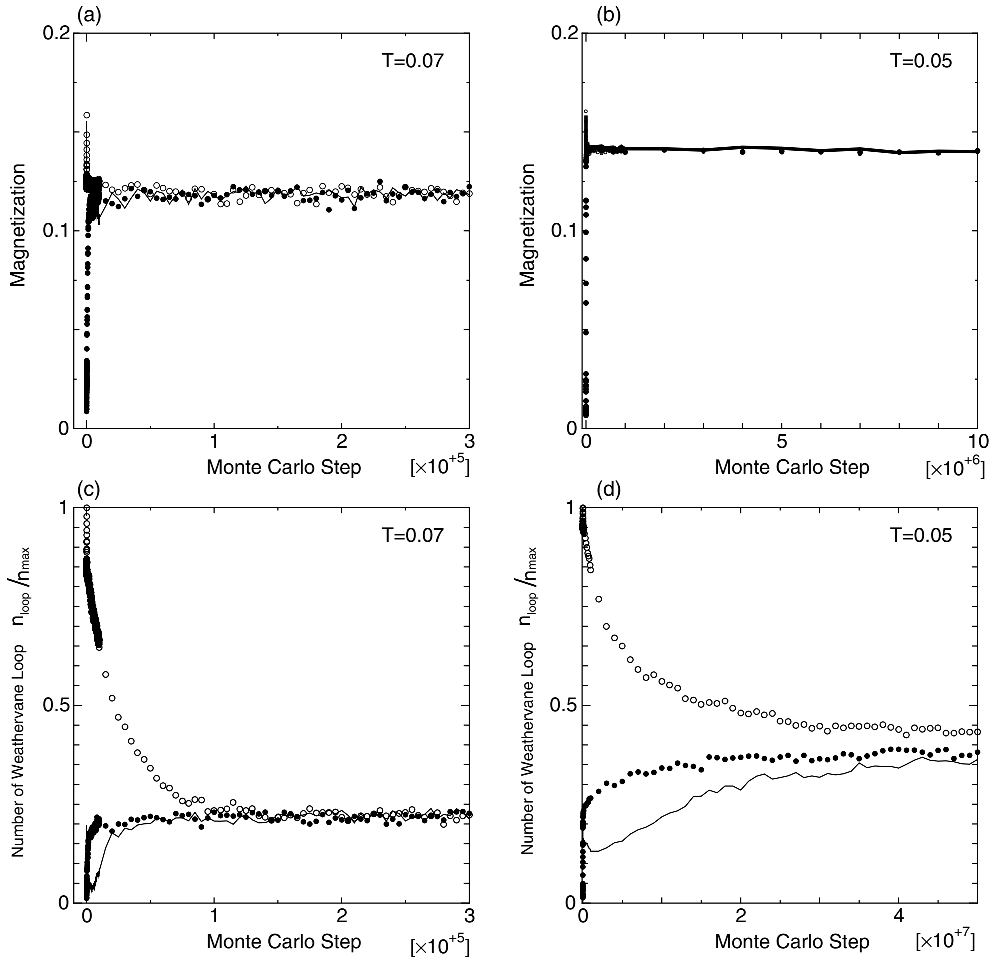

Figure 4. Time evolution of the magnetization (a) at $T = 0.07$ and (b) at $T = 0.05$, and n_{loop} (c) at $T = 0.07$ and (d) at $T = 0.05$. Symbols denote the initial conditions (○ for $\sqrt{3} \times \sqrt{3}$, a solid line for $q = 0$, and ● for the random pattern).

the $\sqrt{3} \times \sqrt{3}$ structure, n_{loop} takes the maximum value $n_{\max} = N/9$, where N is the number of sites.

Next, we consider the time evolutions of the number of the weathervane loops and the magnetization. Here we study the system of $A = 3$ and $N = 675$. In figure 4(a), we show time evolution of the magnetization at $T = 0.07$ starting from three types of initial spin states, i.e. random, $q = 0$, and $\sqrt{3} \times \sqrt{3}$. Here the data are obtained by averaging over 36 independent runs. At $T = 0.07$, though the magnetization relaxes to the equilibrium value in a time, $\tau_{\mathrm{mag}}^{(T=0.07)} \sim 10^3$ MCSs, it takes $\tau_{\mathrm{loop}} \sim 10^5$ MCSs for n_{loop} to relax (figure 4(c)). Depending on the initial states, we find the different relaxation processes in the three cases. In the case of a random pattern, we find that the number of loops increases monotonically. When we start from the $\sqrt{3} \times \sqrt{3}$ structure, n_{loop} reduces from the maximum value to the equilibrium value monotonically. On the other hand, when we start from the $q = 0$ structure, n_{loop} behaves non-monotonic.

At $T = 0.05$, the relaxation time of the magnetization is $\tau_{\rm mag}^{(T=0.05)} \sim 10^3$ MCSs (figure 4(b)). In this case, it should be noted that the relaxation time of the weathervane loop is much longer than the case of figure 4(c) and it is more than 5×10^7 MCSs (figure 4(d)). This slow relaxation is attributed to the energy cost that is necessary for rearrangement of the weathervane loops. The difficulty in the rearrangement of the weathervane loops is the origin of the slow relaxation in easy-axis type anisotropic Heisenberg kagome antiferromagnets.

4. Conclusion

We consider the origin of the slow dynamics in Ising-like Heisenberg kagome antiferromagnets. In this study, the main stress falls on the difficulty of the rearrangement of the weathervane loops, which characterizes the macroscopic degeneracy of the system. The dynamics of the weathervane loops is the origin of the slow relaxation in this system. In this paper, we have considered two-dimensional kagome antiferromagnetic classical spin systems with Ising-like anisotropic interaction. Below the critical temperature, the relaxation of the $n_{\rm loop}$ is slower than that of the magnetization, which is the macroscopic order parameter of this model. We expect that the present mechanism is realized in easy-axis-type anisotropic kagome Heisenberg antiferromagnets in their ferromagnetic ordered state. When interplane interaction between kagome planes exists, we have to study the present mechanism in three dimensions. There the weathervane loop becomes a weathervane plane and the recombination will have more significant effect on the relaxation, which will be reported elsewhere [15].

Acknowledgments

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