Monte Carlo study of the ordering of the pyrochlore Ising model with the long-range RKKY interaction

Atsushige Ikeda and Hikaru Kawamura
Department of Earth and Space Science, Faculty of Science, Osaka University, Toyonaka 560-0043, Japan
E-mail: kawamura@ess.sci.osaka-u.ac.jp

Abstract. The ordering of the Ising model on a pyrochlore lattice interacting via the long-range RKKY interaction, which is intended to model a metallic pyrochlore magnet such as Pr$_2$Ir$_2$O$_7$, is studied by Monte Carlo simulations. Depending on the parameter $k_F$ representing the Fermi wavevector, the model exhibits rich ordering behaviors. When the $k_F$-value is close to the experimental value, the model exhibits a “2-in 2-out” local spin structure reminiscent of the spin ice accompanied by the antiferromagnetic Curie-Weiss constant. This observation is consistent with experiment. The model exhibits a first-order transition into the magnetic long-range ordered state without showing a spin-liquid-like behavior.

1. Introduction
Pyrochlore magnets consisting of corner-sharing network of tetrahedra are typical examples of geometrically frustrated magnets. Intensive studies were performed on “spin-ice” magnets, e.g., Ho$_2$Ti$_2$O$_7$ and Dy$_2$Ti$_2$O$_7$, which possesses a strong Ising-like $<111>$ magnetic anisotropy and exhibits an intriguing glassy behavior [1, 2, 3]. In spin-ice magnets, the local spin configuration at each tetrahedron is known to be the so-called “2-in 2-out” configuration, which is stabilized by an effective ferromagnetic interaction. The dominant interaction in real spin-ice magnets is the long-range dipolar interaction, which falls off as $1/r^3$ with distance $r$ [3, 4, 5, 6]. Thanks to extensive experimental and theoretical studies, the experimental results on spin ice magnets now seem to be relatively well understood on the basis of the dipolar spin-ice model, i.e., the pyrochlore Ising model with the long-range dipolar interaction.

Typical spin-ice magnets Ho$_2$Ti$_2$O$_7$ and Dy$_2$Ti$_2$O$_7$ are insulators. Ordering properties of the corresponding metallic pyrochlore magnets are then of special interest [3]. Recently, Nakatsuji and collaborators reported that a metallic Ising-like pyrochlore magnet Pr$_2$Ir$_2$O$_7$ exhibits an interesting behavior at low temperatures [7]. The magnetism of this compound is borne by the localized moment of Pr$^{3+}$ which possesses a strong $<111>$ Ising-like magnetic anisotropy. Reflecting the metallic nature of the compound, the dominant magnetic interaction is expected to be the long-range RKKY interaction mediated by the Ir$^{4+}$ conduction electrons, rather than the dipolar interaction. Although the Curie-Weiss constant of Pr$_2$Ir$_2$O$_7$ determined from the high-temperature susceptibility measurement is antiferromagnetic $\theta_{CW} \simeq -20K$, and the antiferromagnetic interaction usually prefers “all-in all-out” structure accompanying no frustration, the short-range magnetic order of this compound realized at lower temperatures appears to be “2-in 2-out” similarly to that of spin ice, characteristic of the effective
ferromagnetic interaction. On decreasing the temperature, Pr$_2$Ir$_2$O$_7$ exhibits a spin-liquid-like behavior accompanied by the Kondo-like effect and an unconventional anomalous Hall effect observed in its longitudinal and transverse resistivities, until it eventually exhibits a weak spin freezing at a low temperature $T \simeq 0.12$K [7]. The possible occurrence of the spin-liquid ground state in this compound might then be of special interest.

Under such circumstances, to better understand the ordering of metallic pyrochlore magnets, it would be desirable to study the nature of the magnetic ordering of the pyrochlore Ising model interacting via the long-range RKKY interaction, and compare it with the property of the dipolar spin-ice model [8].

2. The model and the method
We consider the Hamiltonian

$$\mathcal{H} = -\sum_{ij} J(r_{ij}) \vec{S}_i \cdot \vec{S}_j,$$

where $\vec{S}_i$ denotes a three-component unit vector along the $<111>$ direction located at the site $i$ of the pyrochlore lattice, pointing either parallel or antiparallel with the vector connecting the site $i$ and the center of a tetrahedron. The coupling between the spins at the sites $i$ and $j$ is the long-range RKKY interaction given by

$$J(r_{ij}) = -J_0 a^3 \left( \frac{\cos(2k_F r_{ij})}{r_{ij}^3} - \frac{\sin(2k_F r_{ij})}{2k_F r_{ij}^2} \right),$$

where $r_{ij}$ is the distance between the sites $i$ and $j$, $k_F$ the Fermi wavevector, and the sum is taken over all spin pairs on the pyrochlore lattice. Our length unit is taken to be the spacing of the cubic unit cell $a$. The lattice contains $N = 16L^3$ spins (the cubic unit cell contains 16 sites). Periodic boundary conditions are applied.

The model contains one parameter, the Fermi wavevector $k_F$. In Pr$_2$Ir$_2$O$_7$, experimentally determined carrier density $n \simeq 4.13 \times 10^{21}$ cm$^{-3}$ and the lattice constant $a \simeq 10.39 \times 10^{-8}$ cm [7] yield the dimensionless Fermi wavevector $k_F \simeq 2\pi/1.218$ in units of $a^{-1}$.

In order to study the ordering properties of the model, we perform Monte Carlo simulations on finite lattices, with $L$ in the range of $2 \leq L \leq 6$. The long-range nature of the RKKY interaction has been taken into account via the Ewald sum technique. Typically, each run consists of $5 \times 10^4$ Monte Carlo sweeps, several independent runs being made to guarantee the statistical accuracy of the data.

3. The “experimental” case: $k_F = 2\pi/1.218$
First, we study the “experimental” case of $k_F = 2\pi/1.218$. In Fig.1(left), we show the temperature dependence of the energy per spin. The data exhibit a jump at $T = T_c \simeq 16$ (in units of $J_0$), which becomes more eminent with the size $L$ suggesting that the transition is of first-order.

Contrary to a naive expectation, the observed antiferromagnetic $\Theta_{CW}$ does not mean here the occurrence of the “all-in all-out” local spin structure at low temperatures. Fig.2 exhibits the temperature dependence of the ratios of the “2-in 2-out” structure, the “3(1)-in 1(3)-out” structure and the “all-in all-out” structure, which represent the probability that each type of local spin structure is realized at each tetrahedron. As can be seen from the figure, the local spin structure below $T_c$ is “2-in 2-out”, not “all-in all-out”. Indeed, the appearance of the ferromagnetic “2-in 2-out” spin structure under the antiferromagnetic $\Theta_{CW}$ is consistent with the experimental observation for Pr$_2$Ir$_2$O$_7$ [7].
The temperature dependence of the energy per spin (left) and of the inverse susceptibility per spin $\chi^{-1}$ (right) for the parameter $k_F = 2\pi/1.218$.

In Figs.3, we show the calculated spin structure factor $F(\vec{q})$, i.e., the thermal average of the squared Fourier amplitude defined by

$$F(\vec{q}) = \frac{1}{N} \langle |\sum_j \vec{S}_j e^{i\vec{q}\vec{r}_j}|^2 \rangle,$$

where the wavevector is given by $\vec{q} = \frac{2\pi}{a}(h, k, l)$ with $-2 \leq h, k, l \leq 2$. In Figs.3, we show the structure factor in the $(h, h, l)$ plane calculated at a low temperature $T = 2$, which has been averaged over all equivalent directions of $(\pm h, \pm h, \pm l), (\pm l, \pm h, \pm h)$ and $(\pm h, \pm l, \pm h)$. As can be seen from the figure, the spin structure factor exhibits a sharp Bragg peak at $(0, 0, 1)$, or at the equivalent ones, indicating that the magnetic long-range order sets in below $T_c$, the associated spin order being characterized by the wavevector $(0, 0, \frac{2\pi}{a})$. As is evident from the figure, the Bragg peaks at these spots sharpen with increasing $L$.

In fact, such an ordered state is the same as the one observed in the dipolar spin-ice model in its equilibrium [9]. In the case of the dipolar spin-ice model, the $(0, 0, \frac{2\pi}{a})$ ordered state is observed only in a special type of simulation devised to promote equilibration (loop algorithm), not in the standard single spin-flip simulation [6]: In the latter case, the system gets out of equilibrium leading to the glassy, spin-ice behavior. In our present RKKY model, by contrast, the $(0, 0, \frac{2\pi}{a})$ long-range order is achieved more readily, even without using the loop algorithm. Such a difference might be understandable by noting the fact that a significant portion of the “3-in, 1-out” local structures still remains in the ordered state of the RKKY model, which serves to keep the intermediate path for the transition among various different local “2-in, 2-out” structures.
4. More general case: $k_F = 1.13, 1.15, 1.4, 1.7$

In order to examine the stability of the ordering against the possible change of the parameter $k_F$, we also simulate the model for other values of $k_F$. The calculated energy and the inverse susceptibility are shown in Figs.4 and Figs.5, respectively, for $k_F = 1.13, 1.15, 1.4$ and 1.7. As can be seen from Figs.4, the model exhibits a first-order transition at a finite temperature for all $k_F$-values studied. The local spin structure stabilized at lower temperatures turns out to be “2-in, 2-out” for these $k_F$-values.

![Diagram](image1)

**Figure 3.** The spin structure factor $F(\vec{q})$ in the $(h,l)$ plane at a temperature $T = 2$ for $k_F = 2\pi/1.218$. The lattice size is $L = 2$ (left) and $L = 6$ (right).

![Diagram](image2)

**Figure 4.** The temperature dependence of the energy per spin for the wavevectors $k_F = 2\pi/1.13, 1.15, 1.4$ and 1.7.

By contrast, the inverse susceptibility shown in Figs.5 reveals an interesting $k_F$ dependence. With increasing the $k_F$-value from the “experimental” value $k_F = 2\pi/1.218$, $|\Theta_{CW}|$ decreases its magnitude, eventually changing its sign from negative to positive at around $k_F \approx 2\pi/1.14$. At
$k_F = 2\pi/1.13$, $\Theta_{CW}$ becomes positive (ferromagnetic), while the model still exhibits a first-order transition into the long-range magnetic ordered state with keeping the “2-in 2-out” local spin structure. While the type of the magnetic long-range order at the wavevectors $k_F = 2\pi/1.15$ is the same as the one at $k_F = 2\pi/1.218$, i.e., the $(0, 0, 2\pi)$ state, as $\Theta_{CW}$ changes its sign, it changes into a distinct one characterized by the wavevector $(\frac{\pi}{6}, \frac{\pi}{6}, \frac{\pi}{6})$. The computed spin structure factors at $k_F = 2\pi/1.13$ are shown in Figs.6.

On the other hand, with decreasing the $k_F$-value from $k_F = 2\pi/1.218$, the sign of $\Theta_{CW}$ is kept to be negative. The type of the magnetic long-range order remains to be the $(0, 0, 2\pi)$ one at $k_F = 2\pi/1.4$, while at $k_F = 2\pi/1.7$ it changes into the more complex state characterized by the wavevectors with all three spin components different from each other, $h \neq k \neq l$.

![Graphs showing the temperature dependence of the inverse susceptibility per spin for the wavevectors $k_F = 2\pi/1.13, 1.15, 1.4$ and 1.7.](image)

**Figure 5.** The temperature dependence of the inverse susceptibility per spin for the wavevectors $k_F = 2\pi/1.13, 1.15, 1.4$ and 1.7.

It should be noticed that there is no direct correspondence between the sign of the Curie-Weiss constant, i.e., antiferromagnetic $\Theta < 0$ or ferromagnetic $\Theta > 0$, and the type of the local spin structure realized at low temperatures, i.e., antiferromagnetic “all-in, all-out” or ferromagnetic “2-in, 2-out”. This is due to the fact that the type of the local spin order is primarily dictated by the sign of the nearest-neighbor part of the RKKY interaction, while distant-neighbor interactions also contribute to the Curie-Weiss constant, as deduced from the high-temperature expansion expression of the susceptibility [8].

We note in passing that, when one further extends the $k_F$ values toward $k_F < 2\pi/1.7$ or $k_F > 2\pi/1.13$, still other types of magnetic ordered state arise. For example, at small enough values of $k_F \lesssim 2\pi/2.5$, the ordered state turns out to be a simple $q = 0$ state consisting of the “2-in 2-out” structure, while “all-in, all-out” structure is stabilized for larger $k_F$-values of $k_F \gtrsim 2\pi/0.95$, either with a $q = 0$ state ($k_F = 2\pi/0.95$ and $2\pi/0.9$) or a $q \neq 0$ state ($k_F = 2\pi/0.81$ and $2\pi/0.7$). Yet, at any $k_F$-value studied, we have never observed a spin-liquid-like behavior in which the spin long-range order is suppressed down to $T = 0$ or temperatures much lower than $\Theta_{CW}$. 


Figure 6. The spin structure factor $F(q)$ in the $(h,l)$ plane at a temperature $T = 2$ for $k_F = 2\pi/1.218$. The lattice size is $L = 2$ (left) and $L = 6$ (right).

5. Summary and discussion
The ordering of the Ising model on a pyrochlore lattice interacting via the long-range RKKY interaction is studied by Monte Carlo simulations. Depending on the parameter $k_F$ representing the Fermi wavevector, the model exhibits rich ordering behaviors. The model is intended to mimic a metallic pyrochlore magnet such as Pr$_2$Ir$_2$O$_7$. When the $k_F$-value is close to the experimental value, the model exhibits a “2-in 2-out” local spin structure reminiscent of spin ice accompanied by the antiferromagnetic Curie-Weiss constant, which is consistent with experiment. The present RKKY model exhibits a first-order transition into the magnetic long-range ordered state, without showing the spin-liquid-like behavior.

A first-order transition into the $(0, 0, \frac{2\pi}{a})$ ordered state as observed in our simulation has not been observed experimentally. If one deduces the transition temperature by fitting the calculated $\Theta_{CW}$ to the experimental $\Theta_{CW}$, one gets an estimate of $T_c \simeq 10K$. Experimentally, below 10K, Pr$_2$Ir$_2$O$_7$ gets into the Kondo regime. This coincidence strongly suggests that the hybridization effect between the itinerant Ir conduction electrons and the localized Pr moments, not just the effect of magnetic frustration, is essential in realizing the spin-liquid-like behavior observed in Pr$_2$Ir$_2$O$_7$ at low temperatures.

The authors are thankful to Dr. S. Nakatsuji, Dr. Y. Machida, Dr. S. Onoda, Dr. M. Gingras, Dr. P. Holdsworth and Dr. C. Broholm for useful discussion. This study was supported by Grant-in-Aid for Scientific Research on Priority Areas “Novel State of Matter Induced by Frustration” (19052006).

[1] Harris M J, Bramwell S T, McMorrow D F, Zeiske T and Godfrey K W 1997 Phys. Rev. Lett. 79 2554
[2] Ramirez A P, Hayashi A, Cava R J, Siddharthan R and Shastry B S 1999 Nature 399 333
[3] Bramwell S T and Gingras M J P 2001 Science 291 1495
[4] For review, see Melko R G and Gingras M J P 2004 J. Phys. Condens. Matter 16 R1277
[5] Siddharthan R, Shastry B S, Ramirez A P, Hayashi A, Cava R J and Rosenkranz S 1999 Phys. Rev. Lett. 83 1584
[6] den Hertog B C and Gingras M J P 2000 Phys. Rev. Lett. 84 3430
[7] Nakatsuji S, Machida Y, Maeno Y, Tayama T, Sakakibara T, van Duijn J, Balicas L, Milican J N, Macaluso R T and Chan J Y, 2007 Phys. Rev. Lett. 98 057203
[8] Ikeda A and Kawamura H, 2008 J. Phys. Soc. Jpn. 77 073707
[9] Melko R G, den Hertog B C and Gingras M J P 2001 Phys. Rev. Lett. 87 067203