Observation of Magnetic Monopoles in Spin Ice

Hiroaki Kadowaki\textsuperscript{1}, Naohiro Doi\textsuperscript{1}, Yuji Aoki\textsuperscript{1}, Yoshikazu Tabata\textsuperscript{2}, Taku J. Sato\textsuperscript{3}, Jeffrey W. Lynn\textsuperscript{4}, Kazuyuki Matsuhira\textsuperscript{5}, and Zenji Hiroi\textsuperscript{6}

\textsuperscript{1}Department of Physics, Tokyo Metropolitan University, Hachioji-shi, Tokyo 192-0397
\textsuperscript{2}Department of Materials Science and Engineering, Kyoto University, Kyoto 606-8501
\textsuperscript{3}NSL, Institute for Solid State Physics, University of Tokyo, Tokai, Ibaraki 319-1106
\textsuperscript{4}NCNR, National Institute of Standards and Technology, Gaithersburg, Maryland 20899-6102, USA
\textsuperscript{5}Department of Electronics, Faculty of Engineering, Kyushu Institute of Technology, Kitakyushu 804-8550
\textsuperscript{6}Institute for Solid State Physics, University of Tokyo, Kashiwa 277-8581

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Excitations from a strongly frustrated system, the kagomé ice state of the spin ice Dy\textsubscript{2}Ti\textsubscript{2}O\textsubscript{7} under magnetic fields along a [111] direction, have been studied. They are theoretically proposed to be regarded as magnetic monopoles. Neutron scattering measurements of spin correlations show that close to the critical point the monopoles are fluctuating between high- and low-density states, supporting that the magnetic Coulomb force acts between them. Specific heat measurements show that monopole-pair creation obeys an Arrhenius law, indicating that the density of monopoles can be controlled by temperature and magnetic field.

KEYWORDS: magnetic monopole, spin ice, kagomé ice, neutron scattering, specific heat

Since the quantum mechanical hypothesis of the existence of magnetic monopoles proposed by Dirac\textsuperscript{1,2} and many experimental searches have been performed, ranging from a monopole search in rocks of the moon to experiments using high energy accelerators.\textsuperscript{3} But none of them was successful, and the monopole is an open question in high energy physics. Recently, theoretical attention has turned to condensed matter systems where tractable analogs of magnetic monopoles might be found.\textsuperscript{4–6} and one prediction\textsuperscript{5} is for an emergent elementary excitation in the spin ice\textsuperscript{7,8} compound Dy\textsubscript{2}Ti\textsubscript{2}O\textsubscript{7}.

In solid water, the protons are disordered even at absolute zero temperature and thus retain finite entropy,\textsuperscript{9} and spin ice exhibits the same type of disordered ground states.\textsuperscript{8,10} The Dy spins occupy a cubic pyrochlore lattice, which is a corner sharing network of tetrahedra (Fig. 1(a)). Each spin is parallel to a local [111] easy axis, and interacts with neighboring spins via an effective ferromagnetic coupling. This brings about a geometrical frustration where the lowest energy spin configurations on each tetrahedron follow the ice rule, “2-in, 2-out” structure, and the ground states of the entire tetrahedral network are macroscopically degenerate in the same way as the disordered protons in water ice.\textsuperscript{9,10} In addition to this remarkable observation, there is the more intriguing possibility\textsuperscript{5} that the excitations from these highly degenerate ground states are topological in nature and mathematically equivalent to magnetic monopoles.

The macroscopic degeneracy of the spin ice state can be partly lifted by applying a small magnetic field along a [111] direction.\textsuperscript{11} Along this direction the pyrochlore lattice consists of a stacking of triangular and kagomé lattices (Fig. 1(a)). In this field-induced ground state, the spins on the triangular lattices are parallel to the field and consequently drop out of the problem, while those on the kagomé lattices retain disorder under the same ice rules, only with a smaller zero-point entropy.\textsuperscript{12} This is referred to as the kagomé ice state\textsuperscript{11,13–15} (Fig. 1(b)).

In Fig. 1(c) we illustrate creation of a magnetic monopole and antimonopole pair in the kagomé ice state. An excitation is generated by flipping a spin on the kagomé lattice, which results in ice-rule-breaking “3-in, 1-out” (magnetic monopole) and “1-in, 3-out” (anti-monopole) tetrahedral neighbors. From the viewpoint of the dumbbell model,\textsuperscript{5} where a magnetic moment is replaced by a pair of magnetic charges, the ice-rule-breaking tetrahedra simulate magnetic monopoles, with net positive and negative charges sitting on the centers of tetrahedra. The monopoles should interact via the magnetic Coulomb force,\textsuperscript{5} which is brought about by the dipolar interaction\textsuperscript{16} between spins in Dy\textsubscript{2}Ti\textsubscript{2}O\textsubscript{7}. They can move and separate by consecutively flipping spins, but are confined to the two-dimensional kagomé layer (e.g. Fig. 1(d)). This possibility of separating the local excitation into its constituent parts is a novel fractionalization in a frustrated system in two or three dimensions,\textsuperscript{5,17} and enables many new aspects of these emergent excitations to be studied experimentally, such as pair creation and interaction, individual motion, currents of monopoles, correlations and cooperative phenomena.

In the present study, inspired by the theoretical prediction of the monopoles, we have investigated two aspects of magnetic monopoles in spin ice using direct neutron
scattering techniques and thermodynamic specific heat measurements.

Single crystals of Dy$_2$Ti$_2$O$_7$ were prepared by the floating-zone method. At low temperatures, four magnetic moments on each tetrahedron obey the ice rule (2-in, 2-out). The resulting spin ice state is shown in (a). The pyrochlore lattice consists of stacked triangular and kagomé lattices, shown by green and blue lines, respectively, along a [111] direction. (b) Under small [111] magnetic fields, spins on the kagomé lattice remain in the disordered kagomé ice state. An excited state is induced by flipping a spin from (b), enclosed by a dashed circle, where neighboring tetrahedra have 3-in, 1-out and 1-in, 3-out configurations. These ice-rule-breaking tetrahedra are represented by magnetic monopoles with opposite charges depicted by spheres. (d) By consecutively flipping two spins from (c), the monopoles are fractionalized. (e) As the magnetic field is increased, $H \gg H_c$, spins realize a fully ordered, staggered arrangement of monopoles. The inset shows the field dependence of the activation energy. 

Fig. 2. (Color) Specific heat under [111] fields is plotted as a function of $1/T$. In the intermediate temperature range these data are well represented by the Arrhenius law denoted by solid lines. 

A straightforward signature of monopole–antimonopole pairs is magnetic neutron scattering. One challenge to the experiments is to distinguish the relatively weak scattering from the small number of monopoles from the very strong magnetic scattering of the ground states. A theoretical idea [5] is helpful for identifying the monopole scattering is that the [111] field acts as chemical potential of the monopoles, enabling us to control their density as shown by the present specific heat measurements. As the field is increased, the kagomé ice state with low monopole density changes continuously to the maximum density state, the staggered monopole state (Fig. 1(e)), where all spin configurations become “3-in, 1-out” or “1-in, 3-out” to minimize the Zeeman energy. For the present neutron scattering experiments, the
The magnetic Bragg intensity at $T = T_c + 0.05$ K is plotted as a function of the [111] field. The open squares and dashed curves represent the measurements at (220) and corresponding MC simulations, respectively. The dot-dashed curve is the density of positively charged monopoles obtained by the simulation. The inset shows the $HT^c$ phase diagram under the [111] field. The solid line represents the first-order phase transition with the critical point shown by an open circle. The dashed lines are crossovers. The intensity maps shown in Fig. 4 were measured at the two points depicted by open squares.

We selected $T = T_c + 0.05$ K and $H = H_c$ (Fig. 3 inset) for observation of the fluctuating high- and low-density monopoles. At this $HT$ point, we measured intensity maps in the scattering plane. An intensity map of the kagomé ice state at $T = T_c + 0.05$ K and $H = 0.5$ T was also measured for comparison. Figure 4 compares the measured and simulated intensity maps. The observed scattering pattern of the kagomé ice state (Fig. 4(a)) is in excellent agreement with the simulation (Fig. 4(c)), showing the peaked structure at $(2/3, -2/3, 0)$ and the pinch point at $(4/3, -2/3, -2/3)$. These structures reflect the kagomé ice state.

The observed (Fig. 4(b)) and simulated (Fig. 4(d)) intensity maps close to the critical point show a weakened kagomé-ice scattering pattern (by the low-density state) and diffuse scattering around (220) (by the high-density state). The observation agrees fairly well with the simulation. However, the diffuse scattering is less pronounced for the observation. We found that this discrepancy originated from an instrumental condition of the GPTAS spectrometer, which has a large vertical resolution of $\Delta q = 0.25$ Å$^{-1}$ (full width at half maximum, FWHM). We carried out the same measurement on the BT-9 spectrometer. It has a smaller vertical resolution of $\Delta q = 0.1$ Å$^{-1}$ (FWHM), which does not affect the diffuse scattering. The resulting data are shown in Fig. 4(f), which are in better agreement with the simulation (Fig. 4(d)) around (220). An interesting point suggested by this resolution effect is that correlations of the high-density monopoles are three dimensional in space, although the monopoles can only move in the two dimensional layers (Fig. 1(d)). The three dimensional correlations are consistent with the isotropic Coulomb interaction between monopoles. We note that the kagomé-ice scattering pattern is two dimensional in nature, and thus is not affected by the vertical resolution.

To illustrate the high- and low-density monopoles yielding the scattering patterns in Figs. 4(b), 4(d), and 4(f), two typical snapshots of the monopoles of the MC simulation are shown in Fig. 4(e), where we depict magnetic charges at centers of the tetrahedra. Lines connecting the centers of tetrahedra form a diamond lattice, and magnetic charges reside on its lattice points. Regions of the low-density monopoles (where black points dominate) produce the kagomé-ice scattering pattern, while those of the high-density monopoles (where red and blue points dominate) produce the diffuse scattering around the Bragg reflections. These critical fluctuations between high- and low-density phases reinforce the proposed explanation of the puzzling liquid-gas type critical point using the similarity argument to phase transitions of ionic particle systems on lattices. Consequently they strongly suggest existence of magnetic monopoles interacting via the magnetic Coulomb force. Further investigations of critical phenomena, screening of the Coulomb interaction, and effects of the
anisotropic motion of the monopoles within the kagomé lattices are of interest.

A question, which is not pursued in the present study, is how monopoles unbound by the fractionalization move in the kagomé lattice. Comparing the observed Arrhenius law with that of a study of the monopoles unbound by fractionalization, it seems that the interesting temperature ranges where unconfined monopoles move diffusively are roughly that the interesting temperature ranges where unconstrained motion of monopoles in spin ice state, it seems that the interesting temperature ranges where unconstrained motion of monopoles in spin ice state, it seems

emergent phenomena to be explored experimentally.

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