

Observation of Magnetic Monopoles in Spin Ice

Hiroaki KADOWAKI¹, Naohiro DOI¹, Yuji AOKI¹, Yoshikazu TABATA², Taku J. SATO³, Jeffrey W. LYNN⁴, Kazuyuki MATSUHIRA⁵, and Zenji HIROI⁶

¹*Department of Physics, Tokyo Metropolitan University, Hachioji-shi, Tokyo 192-0397*

²*Department of Materials Science and Engineering, Kyoto University, Kyoto 606-8501*

³*NSL, Institute for Solid State Physics, University of Tokyo, Tokai, Ibaraki 319-1106*

⁴*NCNR, National Institute of Standards and Technology, Gaithersburg, Maryland 20899-6102, USA*

⁵*Department of Electronics, Faculty of Engineering, Kyushu Institute of Technology, Kitakyushu 804-8550*

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 $\text{Dy}_2\text{Ti}_2\text{O}_7$ 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,^{1,2)} many experimental searches have been performed, ranging from a monopole search in rocks of the moon to experiments using high energy accelerators.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,⁴⁻⁶⁾ and one prediction⁵⁾ is for an emergent elementary excitation in the spin ice^{7,8)} compound $\text{Dy}_2\text{Ti}_2\text{O}_7$.

In solid water, the protons are disordered even at absolute zero temperature and thus retain finite entropy,⁹⁾ and spin ice exhibits the same type of disordered ground states.^{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.^{9,10)} In addition to this remarkable observation, there is the more intriguing possibility⁵⁾ 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.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.¹²⁾ This is referred to as the kagomé ice state^{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” (antimonopole) tetrahedral neighbors. From the viewpoint of the dumbbell model,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,⁵⁾ which is brought about by the dipolar interaction¹⁶⁾ between spins in $\text{Dy}_2\text{Ti}_2\text{O}_7$. 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,^{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

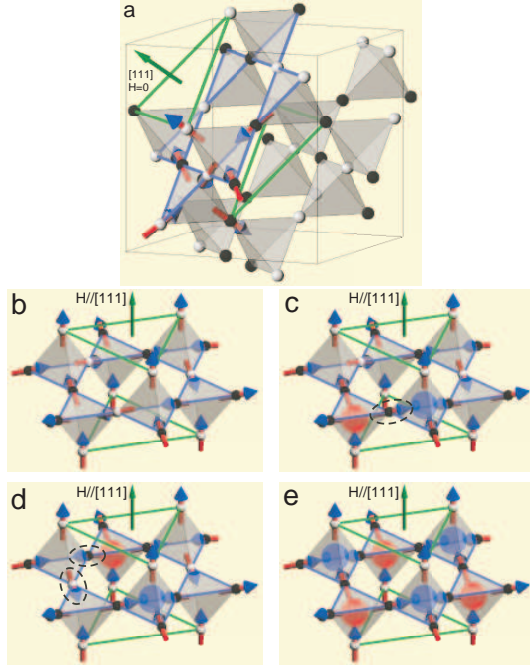


Fig. 1. (Color) Magnetic moments of $\text{Dy}_2\text{Ti}_2\text{O}_7$ reside on the pyrochlore lattice.⁸⁾ At low temperatures, four magnetic moments on each tetrahedron obey the ice rule (2-in, 2-out). The resulting spin ice state is shown in (a). The pyrochlore lattice consists of stacked triangular 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.¹¹⁾ (c) 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.

scattering techniques and thermodynamic specific heat measurements.

Single crystals of $\text{Dy}_2\text{Ti}_2\text{O}_7$ were prepared by the floating-zone method.¹¹⁾ Specific heat was measured by a quasi-adiabatic method. Neutron scattering experiments on a single crystal under a [111] field were performed on the triple-axis spectrometers BT-9 at the NIST Center for Neutron Research and the ISSP-GPTAS at the Japan Atomic Energy Agency. The sample was mounted in dilution refrigerators so as to measure the scattering plane perpendicular to the [111] direction.

A straightforward signature of monopole-pair creation is an Arrhenius law in the temperature (T) dependence of the specific heat (C), $C(T) \propto \exp(-\Delta E/k_B T)$, where ΔE is a field (H) dependent creation energy. One can simply expect $\Delta E = E_0 - \mu H$ owing to the Zeeman effect. Figure 2 shows the measured $C(T)$ of $\text{Dy}_2\text{Ti}_2\text{O}_7$ under a [111] applied field as a function of $1/T$. The Arrhenius law is clearly seen at low temperatures, indi-

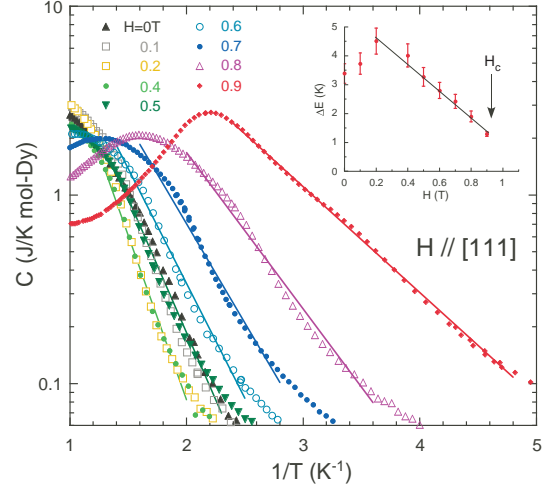


Fig. 2. (Color) Specific heat under [111] fields is plotted as a function of $1/T$. In the intermediate temperature range these data are well represented by the Arrhenius law denoted by solid lines. The inset shows the field dependence of the activation energy.

catating that monopole–antimonopole pairs are thermally activated from the ground state. We remark that all the measurements were performed under field cooling conditions, which are important to avoid complications due to spin freezing^{8, 10, 18)} among the ground state manifolds, whose degeneracy are slightly lifted. We think that the deviation from the Arrhenius law at the lowest temperatures is attributable to these perturbative effects. The observed activation energy ΔE depends linearly on H (Fig. 2 inset) between 0.2 and 0.9 T, i.e., in the kagomé ice state. The deviation from linearity below 0.2 T (spin ice regime) suggests that the nearest-neighbor effective bond energy J_{eff} slightly changes between the two states. The zero-field value $\Delta E(H = 0) = 3.5$ K reasonably agrees with an estimation $\Delta E(H = 0) = 4J_{\text{eff}} = 4.5$ K using J_{eff} in ref. 16. The observed Arrhenius law of $C(T)$, which is attributable to variation of density of the monopole pairs, implies that the number of monopoles can be tuned by changing T and H .

A microscopic experimental method of observing monopoles 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^{8, 14)} of the ground states. A theoretical idea⁵⁾ which 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

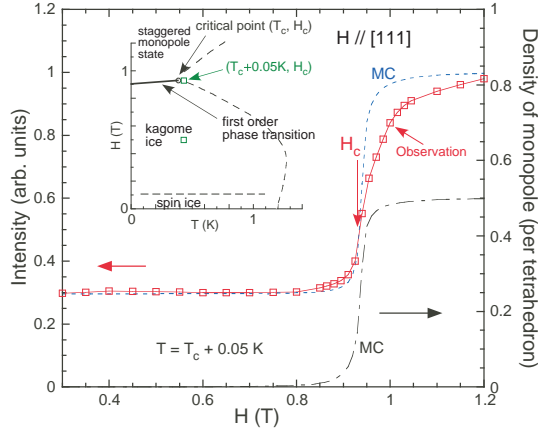


Fig. 3. (Color) The magnetic Bragg intensity at $T = T_c + 0.05$ K is plotted as a function of the [111] field. The open squares and dashed curves represent the measurements at $(2\bar{2}0)$ and corresponding MC simulations, respectively. The dot-dashed curve is the density of positively charged monopoles obtained by the simulation. The inset shows the HT phase diagram under the [111] field. The solid line represents the first-order phase transition with the critical point shown by an open circle.¹³⁾ The dashed lines are crossovers.¹⁵⁾ The intensity maps shown in Fig. 4 were measured at the two points depicted by open squares.

best temperature and field region to observe monopoles in $\text{Dy}_2\text{Ti}_2\text{O}_7$ is close to the liquid-gas type critical point¹³⁾ (T_c, H_c) (Fig. 3 inset). In the monopole picture,⁵⁾ where they are interacting via the magnetic Coulomb force, the first-order phase transition¹³⁾ is ascribed to phase separation between high- and low-density states. We naturally anticipate that neutron scattering close to the critical point is a superposition of the scattering pattern by the (low-density) kagomé ice state¹⁴⁾ and that by high-density monopoles, which is diffuse scattering around magnetic Bragg reflections, i.e., ferromagnetic fluctuations.

The neutron measurements were performed under a [111] field, and Monte Carlo (MC) simulations¹⁴⁾ were also carried out for the dipolar spin ice model^{16,18)} to quantify our observations. Figure 3 shows the field dependence of the magnetic intensity of the $(2\bar{2}0)$ Bragg reflection at $T = T_c + 0.05 = 0.43$ K. The intensity plateau for $H < 0.8$ T corresponds to the kagomé ice state with low density monopoles. The deviation from the plateau as H exceeds 0.8 T indicates that monopoles are being created gradually, while the saturation of the intensity for $H \gg H_c$ denotes the staggered monopole state (Fig. 1(e)). In Fig. 3 we also show the Bragg intensity and the density of the positively charged monopoles obtained by the simulation. The observation shows good agreement with the simulation for $H < H_c$. On the other hand, above H_c there are substantially less monopoles than expected from the simulation, which will be discussed below.

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 H, T 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 structure¹⁴⁾ at $(2/3, -2/3, 0)$ and the pinch point¹⁹⁾ 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 $(2\bar{2}0)$ (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 $\Delta q = 0.25 \text{ \AA}^{-1}$ (full width at half maximum, FWHM). We carried out the same measurement on the BT-9 spectrometer. It has a smaller vertical resolution of $\Delta q = 0.1 \text{ \AA}^{-1}$ (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 $(2\bar{2}0)$. 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,¹⁴⁾ 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 explanation⁵⁾ of the puzzling liquid-gas type critical point¹³⁾ using the similarity argument to phase transitions of ionic particle systems on lattices.²⁰⁾ Consequently they strongly suggest existence of magnetic monopoles interacting via the magnetic Coulomb force. Further investigations of critical phenomena,¹⁵⁾ screening of the Coulomb interaction, and effects of the

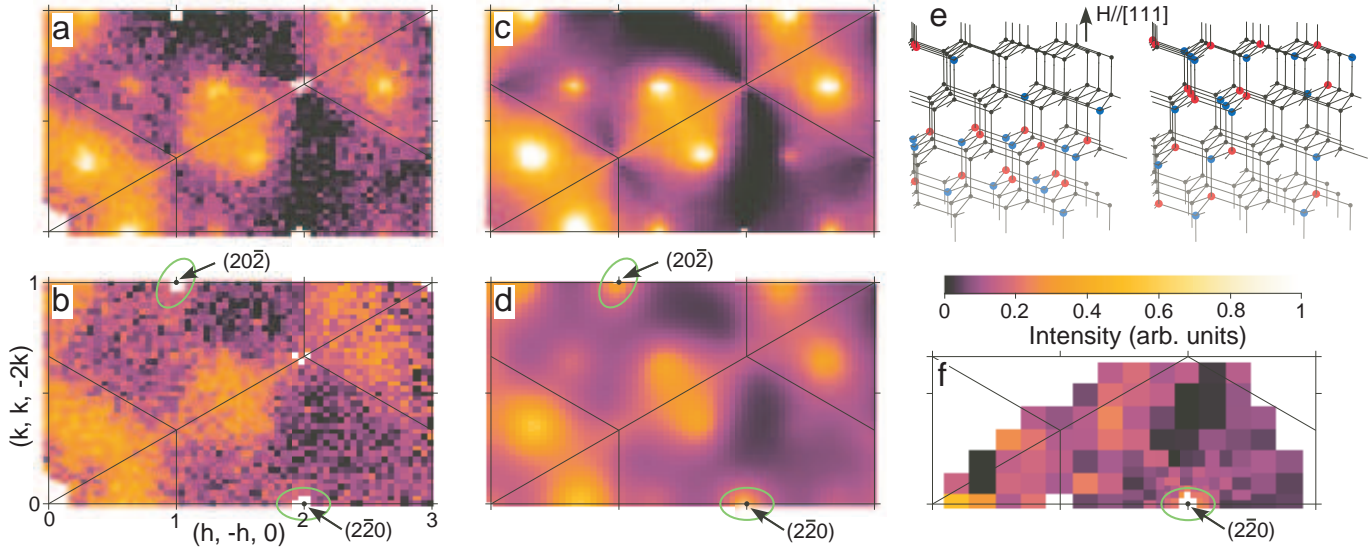


Fig. 4. (Color) Intensity maps measured at $T = T_c + 0.05$ K in the scattering plane are shown for two field values (Fig. 3 inset): the kagomé ice state at 0.5 T (a, c) and the fluctuating high- and low-density monopoles at H_c (b, d, f). (a, b) and (f) were measured on the GPTAS and BT-9 spectrometers, respectively. (c, d) are simulated intensities. (e) Two snapshots of the monopoles of the simulation corresponding to (b, d, f) are shown on the diamond lattice, in which blue, red, and black points represents $+$, $-$ and 0 magnetic charges, respectively. The light green circles in (b, d, f) show the high intensity regions caused by the high-density monopoles, i.e., ferromagnetic fluctuations.

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 study²¹⁾ of the diffusive motion of monopoles in spin ice state, it seems that the interesting temperature ranges where unconfined monopoles move diffusively are roughly $T > 1.5$ K ($H = 0.5$ T) and $T > 0.7$ K ($H = 0.9$ T). There may be another interesting issue in the discrepancy between observed Bragg intensity and the classical MC simulation shown in Fig. 3 ($H > H_c$). We speculate that it may indicate the existence of quantum mechanical effects neglected in the computation. Puzzling experimental facts at low T were also noticed by the slow saturation of magnetization¹³⁾ and the non-zero specific heat¹⁵⁾ above H_c down to very low temperatures, $T < 0.1$ K. For example, if the double spin flips shown in Fig. 1(d) can occur by tunneling,²²⁾ monopoles (or holes in the staggered monopole state) might move more easily than classical diffusion.²¹⁾

Typical elementary excitations in condensed matter, such as acoustic phonons and magnons, are Nambu-Goldstone modes where a continuous symmetry is spontaneously broken when the ordered state is formed. This contrasts with the monopoles in spin ice, which are point defects that can be fractionalized in the frustrated ground states. Such excitations are unprecedented in condensed matter, and will now enable conceptually new

emergent phenomena to be explored experimentally.

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