during chain packing act as if they are large hollow monomers that can pack together to form a jammed (shear-resisting) configuration. A two-dimensional simulation of granular chains at low chain density, where the system is unjammed (has no resistance to shear) (see the figure, panel A), contains randomly configured chains. At high chain density when the system is jammed (see the figure, panel B), semiloop structures appear. Zou et al. revealed that semiloop formation is crucial to the behavior of the granular chain system. The presence of semiloops lowers the jamming density of the system, and this effect is more pronounced in longer chains where there are fewer chains per unit volume.

Real polymer systems undergo a transition to a glassy state below some temperature. It is known that this glass transition temperature increases with polymer length and saturates for long polymers (4). In the granular chain polymer model system, there is no temperature; instead, the inverse density serves as a quantity analogous to temperature. When Zou et al. compare the behavior of the inverse density of the granular chains as a function of chain length to the glass transition temperature of a real polymer as a function of polymer length, they find that the behaviors are nearly identical. This remarkable result could imply that the glass transition in real polymers might be a jamming transition governed by the formation of rigid polymer semiloops. It would be extremely interesting to determine whether this type of semiloop actually occurs in real polymer systems. Semiloops should form much more easily than complete loops because the two ends of the polymer do not need to be near each other. If semiloops play a role in real polymer systems, this would be one of the most beautiful examples of where a macroscopic toy system explains the physics on a molecular scale. Such a result would also confirm that the ideas of jamming originally proposed for granular assemblies (5-7)are applicable to a wide variety of molecular systems.

Granular polymers also represent a new form of granular matter and most likely will exhibit new kinds of phenomena that do not occur in real polymers or in granular systems composed of individual grains. As the jamming density of the granular polymer system can be controlled readily by adjusting the length of the granular chains, introducing chains into other systems that exhibit jamming may allow for a much more precise control of the jamming of these materials.

The experimental demonstration by Zou *et al.* that granular polymers can give insight into molecular polymer systems potentially opens

a new avenue of research for polymer science. For example, polymer systems under shear often exhibit very complex dynamics. Perhaps by shearing granular polymers, the microscopic motions of the shearing process can be studied directly, along with other analogous phenomena such as entanglement and polymer mechanical properties (1, 2). One can imagine going even further and creating increasingly intricate granular polymers to model biological polymers, such as proteins or DNA.

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## PHYSICS

# Observing Monopoles in a Magnetic Analog of Ice

### Michel J. P. Gingras

Experimental evidence has been found that magnetic poles within metal oxide magnets can be separated.

bar magnet has a north and south pole, and cutting it in half just creates two new poles, not two separated monopoles. However, a recent theoretical proposal suggested that defects in the spin alignment of certain oxide magnets can create separated effective magnetic monopoles (1). These materials are called spin ices because the lowest-energy orientation of the magnetic spins closely mimics the most stable arrangement of protons in water ice (2). On pages 415 and 411 of this issue, Fennell et al. (3) and Morris et al. (4) report measurements from neutron-scattering experiments showing that the low-energy excitations in spin ices are reminiscent of Dirac's elementary magnetic monopoles (5) that have so far eluded the searches of high-energy physicists. These dissociated north and south poles diffuse away from each other (6) in these oxides and leave behind a "Dirac string" of reversed spins that can be seen as patterns in the intensity of scattered neutrons.

The materials studied, holmium titanate (Ho<sub>2</sub>Ti<sub>2</sub>O<sub>7</sub>) (3) and dysprosium titanate (Dy<sub>2</sub>Ti<sub>2</sub>O<sub>7</sub>) (4), are geometrically frustrated ferromagnets (7, 8); they are unusual paramagnets with strong spin-spin correlations (8) that become magnetized in a magnetic field. These oxides have a pyrochlore structure; the Ho<sup>3+</sup> and Dy<sup>3+</sup> ions within these crystals form a lattice of corner-sharing tetrahedra (see the figure, panel A). The magnetic moments of Ho<sup>3+</sup> and Dy<sup>3+</sup> act like two states, or Ising model spins, and are constrained by anisotropic forces to point "in" or "out" of the tetrahedra. The minimum energy condition, or rule, is that there must be two spins pointing "in" and two spins pointing "out" on each tetrahedron (7, 8).

The "two-in, two-out" rule is analogous to the Bernal-Fowler ice rule, which states that for energetic reasons, two protons must be "close" and two protons must be "far" from any given oxygen in common water ice (2). As explained by Pauling (2), the very large number of two-in, two-out configurations in a macroscopic ice sample leads to a measurable residual entropy in ice at low temperatures (9, 10). Correspondingly, Ho<sub>2</sub>Ti<sub>2</sub>O<sub>7</sub> and Dy<sub>2</sub>Ti<sub>2</sub>O<sub>7</sub> also exhibit a residual low-temperature "Pauling entropy" (11).

A tetrahedron fulfilling the two-in, twoout rule amounts to an effective "magnetic charge neutrality" where four (two positive, two negative) magnetic charges cancel out at the center of the tetrahedron (1). A spin flipped by thermal fluctuations creates two defective adjoining tetrahedra—a monopoleantimonopole pair (see the figure, panel B). Once formed, these particles can diffuse away from each other by reversing spins along the path they trace as they separate, which recon-

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stitutes tetrahedra that obey the ice rule along the path ( $\delta$ ). In his effort to alter the standard theory of electromagnetism as little as possible (12), Dirac (5) postulated that a monopoleantimonopole pair is connected by an unobservable "string" or tightly wound magnetic solenoids. In spin ices, the "Dirac strings" of reversed spins have direct consequences (3) and are observable (4). Because all tetrahedra, except the two adjoining the north and south poles, fulfill the ice rule, the string is without tension and the energy to separate the poles by an infinite distance is finite (1)—the defects are dissociated, or deconfined.

The monopoles in spin ice act like magnetic charges: They obey analogous electric field laws and exhibit an effective Coulomb's law for their interaction strength. At zero temperature, the spin-ice state can be viewed as a "vacuum" free of monopoles and is referred to as a "magnetic Coulomb phase" (3). This analogy affords a mathematical framework for calculating the underlying spin correlations of the rare-earth magnetic moments. Thermal fluctuations that create dissociated monopoles are sources of the analogous electric field that modifies the spin correlations (3, 4) and the dissociated monopoles can be used to describe the low-temperature thermodynamic properties of the material (1, 4).

Neutron scattering can probe the "Coulomb phase" nature of the spin-ice state by measuring the spin-spin correlation function, C(r), where r is the distance between spins. In the absence of thermally induced monopoles, C(r) does not decay exponentially with r, as would be the case for a conventional thermally disordered paramagnet. Rather, C(r) is theo-

Spin excitations creating magnetic monopoles. (A) Spins on two adjacent tetrahedra for magnetic ions in the pyrochlore lattice satisfy a rule that requires two spins pointing in and two spins pointing out, similar to the arrangement of protons in water ice. (B) The reversal of a spin connecting two tetrahedra amounts to the creation of a "monopole" and an "antimonopole" that differ in magnetic "charge." At finite temperature, monopole-antimonopole pairs are created by thermal fluctuations. The monopoles can separate, leaving behind a "Dirac string" of reversed spins. Their signature was deduced in spin-polarized neutron scattering by Fennell *et al.* in zero applied magnetic field, where the Dirac strings have no preferred orientation. (C) For a magnetic field applied along the crystallographic [100] direction shown, the ground state has the magnetic moments on each tetrahedron pointing with the field, which still maintains the two-in, two-out rule. Two separated tetrahedra, each with a flipped spin (three in, red; three out, blue) leads to a pair of monopoles connected by a string (green) of spins reversed against the field. Morris et al. observed a signature for this string in their neutron-scattering data by carefully tuning the applied magnetic field.

retically expected to display the same spatial anisotropy and  $r^{-3}$ decay as a dipolar interaction. These correlations are manifest in the neutron scattering

as "bow-tie" pinch-point singularities at particular neutron-scattering directions of wave vectors Q, which correspond to a "reciprocal space" of the real-space lattice in the crystals. The theoretical argument for the magnetic Coulomb phase (1) is highly compelling, but all previous neutron-scattering experiments, such as those on Ho<sub>2</sub>Ti<sub>2</sub>O<sub>7</sub> (13) and Dy<sub>2</sub>Ti<sub>2</sub>O<sub>7</sub> (14), failed to find an unmistakable signature of the pinch points. Unlike prior studies, Fennell et al. performed a polarized neutron-scattering experiment where the scattering signal is separated in two components. The pinch points are clearly revealed in the component where the neutron spin is flipped, confirming the theoretical prediction. The pinch points are obscured in the more intense "non-spin flip" signal, which helps to explain why previous studies were inconclusive.

In their analysis, Fennell et al. introduced a parameter that cuts off the pinch-point singularities in reciprocal space, which they associate with the typical length of the Dirac strings. Whereas their experiment was performed in zero magnetic field and did not directly probe those strings, Morris et al. applied a magnetic field B along the [100] crystal direction to induce a magnetically polarized state where the ice rule and the minimum magnetic field energy, or Zeeman energy, are satisfied simultaneously (see the figure, panel C). The magnetic field strength can be tuned near a transition where thermally excited monopole-antimonopole pairs start to proliferate. The resulting flipped spins of the Dirac string are then oriented against the magnetic field direction, with the strings causing cone-like features observable in the scattering intensity pattern. The conic features transform in inclined sheets of scattering when the field direction is tilted away from the [100] direction, in close concordance with the calculations of Morris *et al.* for this state. The specific heat in zero magnetic field can also be described well in terms of a dilute gas of thermally excited monopoles (4).

The demonstration that dissociated monopole-like excitations in spin ices can be observed and manipulated may help guide future studies of similar topological excitations in other exotic condensed matter systems. Of particular interest is the exploration of geometrically frustrated magnetic systems with large quantum mechanical zero-point fluctuations of the magnetic moments away from the classical Ising spin directions (*15*, *16*) enforced in the Ho<sub>2</sub>Ti<sub>2</sub>O<sub>7</sub> and Dy<sub>2</sub>Ti<sub>2</sub>O<sub>7</sub> spin ices considered in (*3*, *4*). Such quantum magnets could provide condensed matter physicists with systems that mimic the physics of quantum electrodynamics.

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