Magnetic Coulomb Fields of Monopoles in Spin Ice and Their Signatures in the Internal Field Distribution

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Fractionalization—the breaking up of an apparently indivisible microscopic degree of freedom—is one of the most counterintuitive phenomena in many-body physics. Here we study its most fundamental manifestation in spin ice, the only known fractionalized magnetic compound in 3D: we directly visualize the $1/r^2$ magnetic Coulomb field of monopoles that emerge as the atomic magnetic dipoles fractionalize. We analyze the internal magnetic field distribution, relevant for local experimental probes. In particular, we present new zero-field NMR measurements that exhibit excellent agreement with the calculated line shapes, noting that this experimental technique can in principle measure directly the monopole density in spin ice. The distribution of field strengths is captured by a simple analytical form that exhibits a low density of low-field sites—in apparent disagreement with reported muon spin rotation results. Counterintuitively, the density of low-field locations decreases as the local ferromagnetic correlations imposed by the ice rules weaken.

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Introduction.—The magnetic field set up by a spin configuration is the most direct manifestation of the underlying magnetic moments. The discovery of a new spin state thus holds the promise of generating—and revealing its existence in—novel properties of the field it sets up.

A case in point is spin ice [1], which, uniquely among magnetic materials in three dimensions, exhibits an emergent gauge field and magnetic monopole excitations [2] that have analogies in magnetic nanoarrays [3–6]. The spin ice state has the great advantage of exhibiting phenomena of fundamental conceptual importance in a setting, which as we describe below, is simple enough to be easily and intuitively visualized: an order of topological nature manifests itself in the fractionalization of the microscopic dipole degrees of freedom, leading to the deconfined magnetic monopoles [7].

Neutron scattering experiments, which provide magnetic field correlations in reciprocal space, have produced some of the strongest evidence so far for the gauge structure [8,9] and "Dirac strings" [10] that emerge at low temperatures. Another probe that has been prominently employed is muon spin rotation [11–13] (μSR), which like NMR, is sensitive to the local fields in real space. For such local probes, studies of the level of detail characteristic of the neutron analysis are still lacking.

We remedy this situation by computing the spatially resolved distribution of internal fields in spin ice. Most fundamentally, the internal fields in spin ice contain a contribution from the underlying magnetic monopoles [2]. Thus, isolating and identifying this contribution is of great conceptual importance in corroborating the peculiar nature of these unique elementary excitations.

Here we show how to visualize the monopole contribution: by measuring the field strength at the considerably sized magnetic voids of the lattice, we find a radially symmetric signal (Fig. 4) that is well described by Coulomb's law, $\propto 1/r^2$, with a coefficient that is in good agreement with the theoretical prediction [2]. Even if measuring the field strength deep inside the sample may be beyond the reach of current experiments, the Coulomb field of a magnetic monopole near the sample surface could be accessible to a sufficiently spatially resolved measurement.

To make contact with μSR and NMR experiments, we compute the full field distribution in the unit cell (Fig. 1). This provides detailed predictions for NMR experiments, with the line shape (Fig. 2) in excellent agreement with the first zero-field NMR measurements, the results of which we report here.

Our analysis places strong constraints on the μSR signatures of the spin ice state. In particular, it seems highly unlikely that the signal detected in Ref. [12] is due to muons implanted in pristine bulk spin ice. (Our results are consistent with earlier estimates of the internal field strength at the muon site [11,13,14].)

In finer detail, we find that the internal field distribution fits well to a simple functional form at all temperatures (Fig. 3). Counterintuitively, we find an "enhancement" of the weak

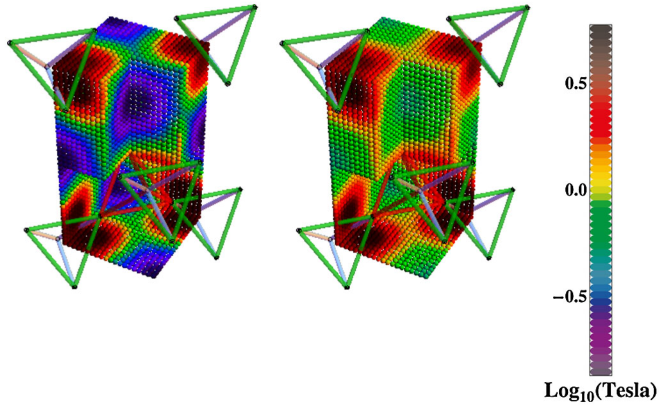

FIG. 1 (color online). Cross sections of the primitive unit cell. Left: average field strength in 2in–2out spin ice configurations. The logarithmic color scale ranges from dark blue (0.137 Tesla, the smallest average field strength recorded) to deep red (6 Tesla). Right: average field strength in completely disordered Ising spin configurations.

field sites as the temperature is lowered. This is surprising, as spin ice is a ferromagnet—as defined by the sign of its Weiss temperature—and one might expect enhanced internal fields to appear as spins align. We interpret this unusual behavior as a result of the interplay of the nanoscopic lattice structure of spin ice and the slow decay of monopolar fields.

Overall, our analysis plugs two gaps: firstly, the conceptual one between the effective long-wavelength emergent gauge theory [2] and the nanoscale physics of the lattice; and secondly, the practical one between theory and real-space experimental probes.

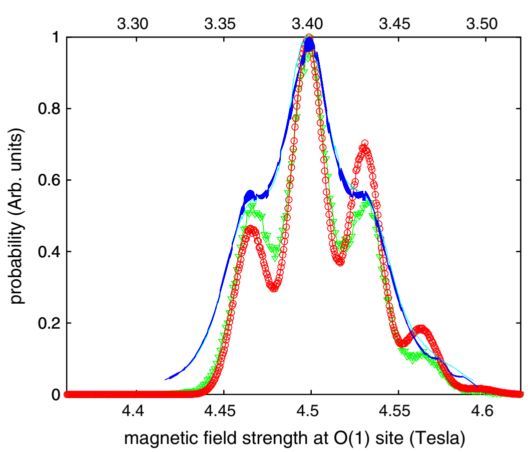

FIG. 2 (color online). Histograms of the magnetic field strength at the $O(1)$ sites obtained from: experimental NMR data nominally at $T = 0.1$ K (blue) and at $T = 0.4$ K (cyan, almost overlapped with the blue line)—top axis; Monte Carlo simulations in equilibrium at $T = 0.6$ K (green triangles) and equally weighted ensemble of 2in–2out spin ice configurations (red circles)—bottom axis. The experimental curves have been shifted so as to match the main peak position from numerics (see the Methods Section in the Supplemental Material [15]); the vertical axis is chosen to set the maxima equal to unity.

Distribution of internal field strengths.—In spin ice, the magnetic dipoles reside on the sites of the pyrochlore lattice, which consists of corner-sharing tetrahedra. We provide details of this structure as Supplemental Material [15] but all that is necessary for digesting the following is (i) in any of the exponentially numerous spin ice configurations, two spins point into each tetrahedron and two point out and (ii) an (anti)monopole corresponds to a tetrahedron with three spins pointing in (out).

In Fig. 3, we show histograms of the internal field distribution $P(h)$ collected across the primitive unit cell for three different classes of spin configurations (see the Methods Section in the Supplemental Material [15]). We consider the cases of monopole-free states (red line) and of configurations containing two maximally separated monopoles, evaluating the field in a primitive cell containing a monopole (blue dots) or halfway between the pair (magenta crosses). This is compared to a random configuration of Ising spins with local [111] easy axes, corresponding to an infinite temperature state (green).

In all cases, at small fields, $P(h) \propto h^2$, while at large fields, $P(h) \propto h^{-2}$. The latter reflects the geometric probability of probing the $1/r^3$ divergence of h close to a spin. The former is a nontrivial result that will play an important role in the interpretation of μSR experiments further below; it implies that a site with a vanishing field is not ''special'' in the sense that even an entirely flat probability distribution for each of the three components of the field vector would yield this functional form for $P(h)$.

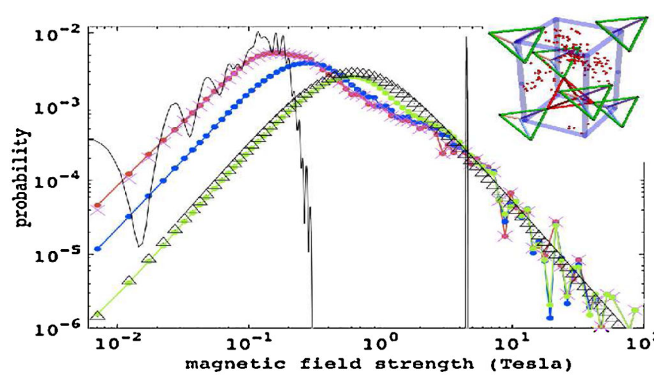

FIG. 3 (color online). Histograms of the field strengths across a uniform cubic grid spanning the primitive unit cell in a system of size $L = 4$ containing $16L^3 = 1024$ spins. Red (leftmost) dotted line: without monopoles. Magenta crosses and blue (intermediate) dotted line: with two monopoles (see text). Green (rightmost) dotted line: random spin ice configuration (i.e., T much larger than any interaction energy scale). Thin black lines: centers of the supertetrahedra and of the rare earth tetrahedra (low and high field curves, respectively). Black triangles: $P(h) \sim h^2/(h^2 + H_0^2)^2$ fit to the random spin ice behavior. Inset: spatial distribution of locations of field strength smaller than 10 mTesla in at least one of the 10 000 statistically independent configurations sampled. The dimensionless volume fraction of such sites is approximately 5×10^{-5}.

The presence of a monopole is only weakly visible far away from it but nearby its effect is felt strongly—statistical weight is shifted from low to higher fields, whereas the overall shape of the distribution does not appreciably vary. This is highly counterintuitive, if one considers that spin ice 2in–2out tetrahedra are "ferromagnetically ordered": all the spins point in the same direction, to the extent allowed by the local easy axes. For a ferromagnet, one would naively expect that the internal fields are larger in the 2in–2out arrangement than they are in presence of a monopole or otherwise disordered spins.

Nonetheless, there are two reasons why this happens. Firstly, the characteristic dipolar correlations between 2in–2out tetrahedra lead to an unusually large cancellation between fields from different tetrahedra. The spins form "flux loops" where the sum of their dipole moments vanishes. These flux loops get broken down as monopoles appear, whose field decreases with distance more slowly than that of any dipole. This effect is captured by the dumbbell model [2], which accounts well for the long-wavelength aspect of the field distribution. (A more detailed explanation can be found in a dedicated section in the Supplemental Material [15] for this Letter.)

However, to reveal the second reason, such a picture needs to be supplemented to account for the detailed structure of the field distribution on the lattice scale. This exhibits considerable local structure, as shown in Fig. 1: near the spins and at the centers of tetrahedra there are large fields in excess of 4 Tesla. By contrast, in the voids between the spins, the fields average much lower. Most saliently, at the centers of supertetrahedra (Fig. 4, inset), the probability of finding a low-field site is greatly enhanced (Fig. 3, black dots). Indeed, the oscillations in this latter curve provide a crucial pointer: at these locations, aided by symmetry, the fields of nearby spins can cancel locally, leaving a lower characteristic field scale, and hence, enhanced low-field probability.

This shows up in the field distribution averaged across a unit cell near a monopole (Fig. 3, magenta line), which follows the form $P(h) \sim h^2/(h^2 + H_0^2)^2$ derived for the distribution of fields due to randomly located and oriented spins [16]. Crucially, the value of H_0 is "reduced" compared to that of a defect-free configuration (red line). This picture is backed up by the good fit of the above equation to a high-temperature state corresponding to a collection of randomly oriented [111]-easy-axis dipoles (green line), and hence a high density of randomly distributed monopoles.

Finally, Fig. 1 directly demonstrates that, along with the breaking of ice rules, the spontaneous spatial organization of fields strengths into high- and low-field locations within the unit cell is suppressed.

Average field due to magnetic monopoles.—Having analyzed the spatial distribution of fields inside the unit cell, we next turn to visualizing the field set up by a monopole. Recall that magnetic monopoles experience a relative force of Coulombic nature. We ask: can one also measure the corresponding magnetic field $(\mu_0/4\pi)(q/r^2)$? This is difficult for two reasons. Firstly, the internal field away from the lattice sites varies tremendously between configurations. Secondly, the Dirac string [2] emanating from the monopole carries a magnetization, $\vec{M}$, which cancels off the field, $\vec{H}$, to give a net $\vec{\nabla} \cdot \vec{B} = \mu_0(\vec{\nabla} \cdot \vec{H} + \vec{\nabla} \cdot \vec{M}) = 0$. These two issues can be taken care of by (i) averaging over many configurations (or, in experiment, over time while keeping the observed monopole position fixed) and (ii) measuring the field at points as far away as possible from any lattice sites. (This also minimizes the strong near field of the spins.)

In Fig. 4, we display the direction of the average fields at the centers of the supertetrahedra set up by two stationary monopoles, which visually reproduce the expected hedgehog-like monopolar field pattern. We have verified that by subtracting the analytical Ewald-summed field for two point monopoles with charge given in Ref. [2], the residual fields appear randomly oriented with average strength tenfold suppressed.

To be more quantitative, in Fig. 4 we show the average field (over 10^6 configurations) evaluated along a line

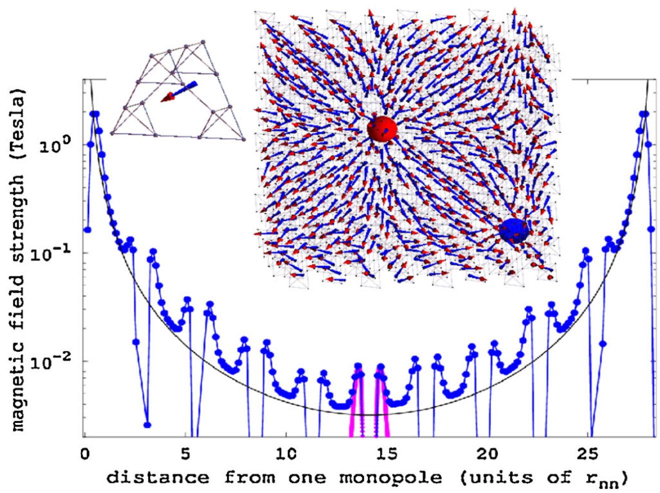

FIG. 4 (color online). Top: illustration of the magnetic field due to the monopoles (red and blue spheres) at the centers of the supertetrahedra of the pyrochlore lattice, visualized by unit vectors in the local field direction (red and blue arrows), for 1024 spins with periodic boundary conditions, averaged over 10 000 independent configurations. As shown, each supertetrahedron is formed by four regular tetrahedra in the lattice. The supertetrahedra centers are (locally) the farthest points from any spin on a pyrochlore lattice site. Bottom: averaged fields along the [001] line joining two monopoles (connected blue dots). The leading behavior is captured, to within 20% error, by the Ewald-summed field from two point magnetic charges at the locations of the monopoles, with charge from the theoretical prediction [2], $2\mu/a_d \simeq 4.6\mu_B/\text{Å}$ (thin black line). The periodic deviations from the Coulomb form are due to spins that sit very close to the line—this contribution is explicitly shown by a thick magenta line for the spin at the midpoint between the monopoles.

joining the two monopoles halfway across a system of 128,000 spins ($L = 20$), along the [001] direction. The Coulomb field predictions are borne out, but masked in part by the line passing very close to spins, which contribute a strong and periodic deviation (magenta line) from the theoretical curve at positions $2\sqrt{2}n$, $n = 0, \ldots, 10$ (in units of $r_{\rm nn}$).

It is tempting to speculate that, if a quantum spin ice material were to be discovered where monopole motion can be made slower than quantum dynamics of spin rearrangements preserving the ice rules, the internal fields would be due almost entirely to the emergent magnetic monopoles.

Experiment I: zero-field NMR at the O(1) sites.—Given the ferromagnetic interactions, the centers of the rare earth tetrahedra experience a large internal field of several Tesla (from numerical simulations using Ewald-summed interactions and fields, see the Methods Section in the Supplemental Material [15] for further details), which as we show next can be used to distinguish between tetrahedra that host monopoles and those that do not.

The centers of the rare earth tetrahedra in the pyrochlore lattice are occupied by oxygen ions [customarily referred to as $O(1)$ oxygens]. The ^{17}O isotopes are NMR active and can be used to detect the internal fields in a zero-field NMR measurement. Indeed, two of the present co-authors have carried out the first NMR experiments on the "monopole-free" line as shown in Fig. 2. Also shown is a comparison to Monte Carlo simulations in thermal equilibrium. The agreement of the line shape is remarkably good. The experimental spectrum is actually peaked at 3.4 Tesla, i.e., about 25% below the calculated peak. This shift is most likely due to spatial distribution of Dy-4f electrons causing deviations from a point dipole approximation (multipolar effects) and/or effects of Dy-O chemical bonding.

A promising aspect of these NMR measurements lies in the possibility of the direct detection of monopoles. Indeed, when a tetrahedron hosts a monopole, the field at its $O(1)$ site drops by approximately 13%. This effect is 3 times larger than the line width due to variations in the field at the $O(1)$ site because of neighboring monopoles or more distant spins. Therefore, the relative intensity of the NMR signal at this pair of field values provides a quantitative measure of the density of monopoles. In practice, small densities may be hard to detect above the background. Also, monopoles must not move over the timescale of NMR spin echo experiments (a few tens of microseconds) to be detected as a distinct resonance line. Given the insights from modeling ac susceptibility results, which suggest a hopping rate in the range of milliseconds [17,18], this condition seems comfortably achievable.

Experiment II: muon spin rotation.—The other major probe of the local magnetic fields is μSR. Indeed, Ref. [12] has reported using μSR experiments to measure the monopole charge via an ingenious analogy to the Wien effect familiar from electrolytes. This is currently a contested experiment [13] and we would like to make a few observations on it from the perspective of the current Letter.

First, we note that one important feature of the data presented in Ref. [12] is that their signal was extracted from sites where the muons experience very low fields, of order a few milliTesla. This follows from the observation that doubling the strength of the applied transverse field from 1 to 2 mTesla results in doubling the $\mu_{\rm SR}$ precession frequency. From our work, it is clear that there is a very low density of such sites (Fig. 3) located preferentially near the centers of supertetrahedra (Fig. 1). Thus, it seems very unlikely that the substantial muon signal emanates from sites in the pristine bulk.

Second, the analysis of Ref. [12] invokes two distinct phenomena: first, the applied magnetic field induces an increase in the monopole density; and second, this increase leads to an enhanced depolarization rate. The first feature is indeed an expected phenomenon for the steady state of electrolytes in a field, but for spin ice, it can only occur as a transient. Since monopole motion magnetizes the sample, there can be no steady state with a nonzero monopole current. Moreover, in thermal equilibrium, the density of monopoles is believed to decrease in an applied field. About this, our present work has nothing to say.

However, regarding the second step, our results indicate that a field-induced regime with heightened monopole density would be accompanied by a depletion of low-field sites. At least this aspect is qualitatively in keeping with the finding in Ref. [12] that the rate of decay of the μSR asymmetry gets larger in the Wien setting as the applied field is increased.

Third, combining the above observations leads us to consider the interesting possibility that the signal arises from the action of an enhanced monopole density on muons implanted outside the sample. The idea here is that outside the sample the monopole fields would dominate over the much smaller fields present in their absence—given that the field of a monopole decays less slowly than that of an isolated spin. (Interestingly, it was very recently suggested in Ref. [13] that the signal from muons inside the sample is lost altogether because of large [compared to 1 mTesla] and fast magnetic fluctuations and that the only measurable signal due to spin ice comes from muons implanted outside, sensitive to stray fields, which are analyzed in Ref. [19].)

A very rough estimate based on a monopole liquid subject to Debye screening [20] suggests that in the temperature range $T \sim 0.2$–0.5 K, the magnetic field set up by a monopole measured a distance roughly 100 ± 50 Å from the sample surface both lies in the range relevant for μSR (between 0.1 and 1 mTesla) and dominates that set up by an individual spin in the sample. Note that, unlike stray fields set up by the magnetization induced by a uniform external field, the monopole density grows with temperature, thus providing a qualitative discriminant between the two. Therefore, a combined study of temperature-dependent (uniform) susceptibility and μSR experiments looks like the most prominent direction to make progress on this issue.

All such considerations point to the need for more detailed studies of what happens near the surface of a sample [21], e.g., what happens to the surface monopole density (as a function of time and field)? Moreover, at present, little is known about the surface of spin ice samples, e.g., what the nature of the local crystal fields is or how the magnetic lattice terminates. It is also worth bearing in mind that equilibration in spin ice may be incomplete at low temperatures, as the equilibrium monopole density vanishes exponentially. At 70 mK, the lowest temperature accessed so far in μSR measurements, their numberdensity is estimated to be such that there is only one pair for a ‘‘macroscopic’’ volume of a quarter of a cubic meter.

Finally, we would be remiss if we did not note that our considerations here have everything to do with spatially fluctuating static fields and their dephasing of muon precession, whereas Ref. [13] proposes that large dynamical fluctuations are present. The tension between their proposal and the magnetic susceptibility data that are consistent with much slower dynamics for the spins will need to be resolved before a consistent account of spin dynamics in spin ice can be given.

Conclusions.—We have provided a way of visualizing the magnetic field of a magnetic monopole inside spin ice, yielding the first nanoscopic real space picture of this fractionalized excitation. Our work attests to the reality of the monopolar magnetic field not only at long wavelengths but also on the lattice scale.

As experimental proofs of the existence of monopoles move in the direction from thermodynamics towards increasingly microscopic ‘‘single-monopole’’ detection, we hope that this work will lay the theoretical groundwork for future searches, such as the ones using NMR or local field probes outside the sample that we have outlined above.

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