NMR relaxation in spin ice due to diffusing emergent monopoles I

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At low temperatures, spin dynamics in ideal spin ice is due mainly to dilute, thermally excited magnetic monopole excitations. I consider how these will affect the dynamics of a nuclear spin (the same theory applies to muon spin resonance if implanted muons do not diffuse). Up to the time scale for nearby monopoles to be rearranged, a stretched-exponential form of the relaxation functions is expected. I work out the expected exponent in that exponential and the formulas for the $T_1$ (longitudinal) and $T_2$ (dephasing) relaxations, as a function of the monopole density (and implicitly the temperature).

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Introduction —

The dipolar spin ice state [1–5] occurs at sufficiently low temperatures in certain highly frustrated pyrochlore structure magnets such as Dy$_2$Ti$_2$O$_7$. The magnetic ions (here Dy$^{3+}$) sit on a “pyrochlore” magnetic lattice consisting of corner-sharing tetrahedra, and have a strong uniaxial anisotropy along their local (111) symmetry axis. The practically degenerate lowest energy states consist of the (exponentially many) configurations in which every tetrahedron has two spins pointing inwards and two spins pointing outwards. The elementary excitations in such a material are defect tetrahedra with three spins in and one out, or the reverse, which were shown to behave as (emergent) magnetic monopole [3]. The low-temperature thermodynamics and dynamics of dipolar spin ice depend on the density and mobility of the monopoles.

Measurements of the specific heat [6] and (static) spin correlations in the presence of a large field [7], corroborate the monopole picture. An analog of the Wien effect in electrolytes was invoked to infer the effective monopole charge from muon spin relaxation [8] but this has been called into question [9, 10]. The dynamics in magnetic relaxation experiments [11] shows an activated dynamics, but not the activation energy expected from the basic monopole model [12]. Low-temperature nuclear magnetic resonance (NMR) [13, 14] or muon spin resonance experiments [15, 16] may aid in disentangling this confusion, as they probe local spin dynamics rather than the uniform bulk magnetization.

In this Letter, I will first review the essentials of spin ice and its emergent monopoles, and then set up the simplest possible model for NMR relaxation functions, both $T_1$ (longitudinal) and $T_2$ (dephasing) kind. In the presence of disorder that is fixed for the duration of the measurement — which includes monopoles if they do not diffuse too fast — the NMR signal is a superposition of relaxation functions due to the inequivalent environments of many probe spins, giving a stretched exponential form. In the monopole case, the form of the $T_1$ relaxation function will be shown to have the same stretched-exponential form as the case of fixed magnetic impurities; but the $T_2$ relaxation function is novel, in that a power of $t^{1/2}$ is replaced by $t^{3/2}$ in the exponential. Experiment does find stretched exponentials, but I will show their parameters are incompatible with a monopole origin. It is proposed that interstitial “stuffed” moments (additional to the spin ice Hamiltonian) are responsible for the NMR signal at very low temperatures.

Dipolar spin ice and dumbbell model —

The experiments in Ref. [13] are on Dy$_2$Ti$_2$O$_7$ in which the magnetic ions are rare earth Dy$^{3+}$ (Ho$_2$Ti$_2$O$_7$ is similar). Each Dy spin has a strong Ising-like anisotropy with easy axis aligned along the (111) direction of its local 3-fold symmetry. The pyrochlore lattice nearest-neighbor distance is $a_p = 1/\sqrt{8}$ times the cubic lattice constant. The tetrahedron centers, separated by $a_d = \sqrt{3}/4$ times the cubic lattice constant, constitute the vertices of a diamond lattice, of which the magnetic (pyrochlore) sites are the bond midpoints. The dipole interaction of spins at $r$ and $r'$ is

$$D[1 - 3 \cos \psi \cos \psi']/|r - r'|^3$$

with $D \equiv \mu_0 \mu^2/4\pi a_p^3$, where $\mu$ is the moment of one spin, and distances are measured in units of $a_p$. Also, $\cos \psi$ and $\cos \psi'$ are the angles made by their respective easy axes with the vector $(r - r')$. The effective nearest-neighbor Ising spin interaction is $J_{\text{eff}} = (J + 5D)/3 \approx 1.1K$, with a net ferromagnetic sign that is frustrated since the 3-fold axes differ by the tetrahedral angle 109°, enforcing the well-known two-in/two-out spin structure in every tetrahedron.

I adopt the “dumbbell” approximation for the spin Hamiltonian of the model system; each point dipole is represented by a pair of opposite magnetic charges on neighboring tetrahedron centers. Then, in any two-in/two-out state, these magnetic charges cancel at every diamond lattice site so that (within this approximation, and nearly so in dipolar spin ice [1]) all such states are exactly degenerate.

The elementary excitation is a tetrahedron having 3 in and 1 out-pointing spins, or the reverse, which is the “monopole” defect (located at the tetrahedron center). Each monopole contributes a far field of Coulomb form $|B| = Q/4\pi |R|$, where $Q = \pm 2\mu/a_d$ is its (conserved) magnetic charge, so they are indeed emergent magnetic monopoles [3]. Monopole pairs interact with the corresponding Coulomb-like potential, and even outside the dumbbell approximation each microstate’s total energy depends almost entirely on the monopole positions and very little on the configuration of spins in 2-in/2-out tetrahedra apart from the monopoles. Where $\mu$ is the electron spin’s moment and $a_d$ is the separation of tetrahedron centers.
A single spin flip in the ground state produces a pair of oppositely charged monopoles, which after separating can diffuse independently over tetrahedron centers or ultimately recombine, much like electrons and holes in a semiconductor. We let $\Delta_0$ be the cost to create and pull apart a pair of monopoles. Within the dumbbell model, this is set by making the nearest-neighbor spin interaction to match $J_{dd}$, by setting $\Delta_0 = \frac{1}{2}\left[ J + 4(1 + \sqrt{2/3})D \right] \approx 6K$ [3]. If we ignore Debye screening, the thermally excited density of unbound monopoles of both signs is thus
\[ n_{\text{mono}} = \frac{(C_{\text{mono}}/v_d)e^{-\Delta_0/2T}}{2T} \] (2)
where $v_d = \sqrt{2\pi a_d^3}$ is the volume per diamond vertex, and Pauling’s approximation for the entropy gives $C_{\text{mono}} \approx 4/3$ for the prefactor.

There are two monopole-dominated temperature regimes. Most studies to date (including NMR [17]) focused on moderately low temperatures 1–5 K, at which thermally excited monopoles are common but dilute enough to be useful degrees of freedom, but still dense enough that Debye screening is crucial and reduces the effective $\Delta$ in (3). The present study focuses on the very low temperatures $T < T^* \approx \Delta_0/10 \approx 0.6K$, at which $n_{\text{mono}} < 0.01$ and screening is negligible. ($T^*$ also appears experimentally as the crossover between different behaviors [13].)

**Bound pairs** of opposite monopoles are possible with finite separations. The closest separation is nearest-neighbor tetrahedra (separation $a_d$), which is equivalent to a flip of just one spin (shared by the tetrahedra) relative to the 2-in/2-out background. This has a cost $\Delta_2$ implying a density of pairs $\approx \exp(-\Delta_2/T)$. Although the energy is reduced by the Coulomb attraction ($\Delta_2 < \Delta_0$), we lose the factor of 2 in the exponent of (2) (due to the entropy of deconfinement), so it depends on details whether bound or unbound monopoles are commoner. As will be shown below, only the unbound ones make a novel contribution to the $T_2$ relaxation. (Following a quench, “back-to-back” bound pairs of monopoles on adjacent tetrahedra may also persist out of equilibrium whenever the intervening spin has the minority in/out sense. [18])

Dy spins are assumed to flip at rate $\tau_0^{-1}$ if the energy $\Delta E$ is unchanged or decreases, otherwise $\tau_0^{-1}\exp(-\Delta E/T)$. The only spin flip with $\Delta E = 0$ is by one of the 3 spins in the majority (in or out) direction on a monopole tetrahedron; this transfers the monopole to the other tetrahedron sharing the flipped spin. [19] Thus the dynamics at low $T$ consists of random-walking monopoles which have a combined rate to hop of $3\tau_0^{-1}$, plus the occasional creation or annihilation of a monopole pair. Based on magnetic relaxation experiments [11], it is believed that $\tau_0 = 10^{-8}$ to $10^{-3}$ s and is temperature independent. (The $\tau_0$ value can be inferred from NMR, as explained further below.)

**NMR model**

To model the zero-field NMR, I assume a single probe moment located at the origin, which for simplicity is imagined to be spin 1/2. [20] To zero order, the probe sees a base field $h_0$ depending on the (frozen) nearby Dy spins, the direction of which defines the probe spin’s quantization axis. Specifically, I have in mind an $O^{17}$ nucleus situated at a tetrahedron center: there, $I_0$ is due mainly to the four surrounding Dy spins (which satisfy the 2-in/2-out constraint) and $h_0$ points along a $\langle 100 \rangle$ axis. The base NMR precession frequency is $\omega_0 = \gamma h_0$ where $\gamma$ is the gyromagnetic ratio. (In the case of the $T_{17}$ nuclear moment [21], $\omega_0$ instead represents its quadrupolar frequency.) All nontrivial probe spin dynamics is a response to additional, time-varying perturbing fields $h(t)$ due to distant flipping spins (presumably Dy: interaction between nuclear moments will be neglected). We call these “flippers” for short: they might either be diffusing monopoles, or fixed spin impurities, and each flips randomly with a time constant $\tau_x$.

Within the material, the frozen local magnetic field has strong and complicated spatial modulations within each unit cell [22, 23]. However, only changes of $h(t)$ matter for the NMR response: $T_1$ relaxation is sensitive only to high frequencies — i.e. the flips themselves — while the $T_2$ relaxation is measured using the spin-echo technique which cancels the frozen non-uniformity of the field. Such changes are given, at separations $R > a_d$, by the “Coulomb” fields of the monopoles, that is $\pm Q/4\pi R$ in the radial direction. Actually, since a single monopole hop is the same as one spin reversal, the field change $\Delta h$ is just twice the dipole field due to that spin, which I describe by a field scale $h_D/R^3$, where the distance $R$ is measured in units of $a_d$. We can convert the field scale to a frequency using the gyromagnetic ratio, $\omega_\perp = \sqrt{2}\omega_d/R^3$. The factor $\sqrt{2}$ is the root-mean-square of the angular factor in (1).

I will compute both kinds of NMR relaxation functions $g_1(t)$ and $g_2(t)$. To start with, consider the relaxation functions $g_{01}(t)$ and $g_{02}(t)$ due to a single flipper at distance $|R|$ from the probe spin. To set up the longitudinal or $T_1$ relaxation, say the probe spin is initially aligned with its local axis. The base NMR precession frequency is $\omega_{\perp}^0 = \gamma h_0^0$; assuming this changes much faster than a precession period, the old state is projected onto $\perp$ (which satisfy the 2-in/2-out constraint) and $I_0$ points along a $\langle 100 \rangle$ axis. The base NMR precession frequency is $\omega_0 = \gamma h_0$ where $\gamma$ is the gyromagnetic ratio. (In the case of the $T_{17}$ nuclear moment [21], $\omega_0$ instead represents its quadrupolar frequency.) All nontrivial probe spin dynamics is a response to additional, time-varying perturbing fields $h(t)$ due to distant flipping spins (presumably Dy: interaction between nuclear moments will be neglected). We call these “flippers” for short: they might either be diffusing monopoles, or fixed spin impurities, and each flips randomly with a time constant $\tau_x$.

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\[ g_1(t) \equiv \left< (\cos \delta)^{N_f} \right> \approx \exp \left( -\frac{\Delta h_\perp^2 t}{4h_0^2/\tau_x} \right), \] (3)
where the $N_f$ is the number of flips in time $t$, its probability given by a Poisson distribution $(t/\tau_x)^N_f \exp(-t/\tau_x)/N_f!$.

The “$T_2^*$” relaxation represents dephasing due to the precession of the in-plane angle of transverse polarization due to the fluctuating part of the longitudinal field, $\omega(t) = \gamma(h_\parallel(t) - h_0)$, where $\gamma$ is the probe spin’s gyromagnetic ratio and $h_\parallel$ is field component along the probe spin’s local axis. The $T_2$ relaxation is measured using the spin-echo technique: the polarization is (effectively) flipped by a $\pi$-pulse at a time $t$.
and then evolved till time $2t$. The relaxation function is thus $g_2(2t) = \langle \cos(\Phi(2t)) \rangle$ where

$$\Phi(2t) \equiv \int_0^t \omega(t')dt' - \int_0^{2t} \omega(t')dt'$$

(4)

is the accumulated phase difference for precession around the quantization axis. Thus, the $T_1$ relaxation is dominated by the fluctuations at high frequency (compared with $\omega_0$), whereas the $T_2$ relaxation is dominated by the slower fluctuations at frequencies comparable to $1/t$.

Now consider specifically the dephasing relaxation function $g_{02}(\mathbf{R}; t)$ due to a single “flipper” at distance $|\mathbf{R}|$. The change in precession frequency at each flip is $\pm 2\omega_0$. If we know that exactly one spin flip occurred at a random time within the interval $[0, 2t]$, it can be worked out that $g(2t) = 1 - A(2t)$, where $A(2t) \equiv 1 - \sin(\omega_0 t)/(\omega_0 t) \approx (\omega_0 t)^2/6$. If the flips that are independent, we use the Poisson distribution to obtain

$$g_{02}(\mathbf{R}; t) = e^{-\frac{2\omega_0}{\tau_x}A(t)} \approx e^{-(t/T_{02})^3}$$

(5)

where $T_{02}^{-3} = 1.4x_2^{-2}$. The assumption of independent flips is valid for a monopole, [24]. This is the key difference between the spin flip statistics of a monopole and those of a fixed impurity: a conventional Ising impurity must alternate plus and minus flips since it has only two states.

In the case of an alternating flipper, if $\omega_0\tau_x \gg 1$ then even one flip by the flipper suffices to randomize the phase, so $g_{02}(2t) \approx e^{-2t/\tau_x}$. On the other hand, if $\omega_0\tau_x \ll 1$ and $t/\tau_x \gg 1$, then $\langle \omega(t)^2 \omega(t')^2 \rangle = \omega_0^2 e^{-2(t-t')^2/\tau_x}$ and $\Phi$ has a Gaussian distribution with $\langle \Phi(2t)^2 \rangle = (2t)\omega_0^2 \tau_x$, implying $g_{02}(2t) \approx e^{-2t/T_{02}}$ with $T_{02}^{-3} = \omega_0^2 \tau_x$.

We can summarize all cases of the single-flipper results by

$$g_{0i}(\mathbf{R}; t) \approx e^{-[\mathbf{R}/T_{0i}]^{3/2} \beta_i^0/|\mathbf{R}|}$$

(6)

where $\beta_{01} = 1$, while $\beta_{02} = 1$ for a fixed flipper at $t > \tau_x$, but $\beta_{02} = 3$ for short times or a diffusing flipper; the corresponding time constants are

$$T_{01}^{-1} = (\omega D^2/\omega_0)^2 \tau_x^{-1};$$

(7a)

$$T_{02}^{-1} = \omega D^2 \tau_x \quad \text{fixed, } t \gg \tau_x;$$

(7b)

$$T_{02}^{-3} = (\omega D^2)^3/\tau_x \quad \text{diffusing or } t \ll \tau_x.$$  

(7c)

and $\omega_0 \tau_x \sim \omega D T_{02}/|\mathbf{R}|^{3} \ll 1$ is assumed.

Random environments: stretching exponentials due to in-homogeneity

When different probe moments have different environments during the measurement time, the observed signal is an average of the relaxation function over these environments and is likely to acquire a stretched exponential form. This fact is familiar (since the 1960s) in the case that the field fluctuations are due to fixed paramagnetic impurities. The same thing happens if the fluctuations are due to diffusing monopoles whose displacement during the measurement time is small compared to their distance $|\mathbf{R}|$ from the probe spin. I next work through the universal form for the relaxation function, averaged over environments, valid for both the $T_1$ and $T_2$ relaxations. The key trick, allowing for the simple final result, is that (for either kind of relaxation) the combined relaxation function due to many flippers at sites $\{\mathbf{R}_i\}$ is simply a product $g_i^{\text{comb}}(t) = \prod_{\mathbf{R}_i} g_{0i}(\mathbf{R}_i; t)$. (This follows immediately from the fact that both kinds of relaxation functions are products of independent random variables depending on the respective flippers.) A (grand-canonical) ensemble is specified by setting the flipper occupation of each site to be $1$ with probability $n_\mathrm{x}$ or $0$ with probability $1 - n_\mathrm{x}$ [25]. Defining $g_i(t)$ to be the ensemble average of $g_i^{\text{comb}}(t)$, I use conditional probability to obtain:

$$g_i(t) = \prod_{\mathbf{R}} \left(1 - n_\mathrm{x}g_{0i}(\mathbf{R}; t)\right) \approx e^{-n_\mathrm{x}F_i(t)},$$

(8)

where, independent of $n_\mathrm{x}$

$$F_i(t) \equiv \sum_{\mathbf{R}} \left[1 - g_{0i}(\mathbf{R}; t)\right] \approx \sum_{\mathbf{R}} \left[1 - e^{-\left(\omega/\tau_{x}\right)^{\beta_i}|\mathbf{R}|^{-\beta_i}}\right]$$

$$\approx c_6(t/T_{0i})^{\beta_i/2}$$

(9)

where I converted the sum in (8) to an integral, using $\sum_{\mathbf{R}} \to d^3 \mathbf{R} / \sqrt{2}$ in units of $1$/Appl. Phys. 3, thus $c_6 = (4\pi/3) \int_{0}^{\infty} (1 - e^{-t^2})d(1/\xi) = (32\pi^3/9)^{1/2} \approx 10.5$.

We can qualitatively interpret the result (8) as follows. More distant flippers contribute smaller fields which take longer to decohere the probe spin; thus in (8), $g_{0i}(\mathbf{R}; t) \approx 1$ at short $|\mathbf{R}|$ or 1 for large $|\mathbf{R}|$. Hence the sum in (9) roughly counts how many are in the first category; if you write $g_{0i} \sim \exp(-R^3/|\mathbf{R}|^6)$, then $R^3$ is the effective radius within which we count the sites so $F_i(t) \propto R_i^3$.

Incidentally it can be seen that if $\tau_x$ is temperature-independent, then the only temperature dependence in (8) is via $n_\mathrm{x}$; thus plots of $\ln g_i(t)$, taken at different temperatures, ought to be identical, apart from an overall prefactor which gives the temperature dependence of $n_\mathrm{x}(T)$.

The final result in all cases is a stretched exponential

$$g_i(t) \propto \exp[-(t/T_i)^{\beta_i}].$$

(10)

where $\beta_i = \beta_{0i}/2$ and

$$T_i^{-\beta_i} = c_6 n_\mathrm{x} T_{0i}^{-\beta_i}. $$

(11)

If the “flipper” (such as monopoles) are thermally excited, then the temperature dependence of $T_1$ and $T_2$ follows from $T_i \propto (\tau_x/n_\mathrm{x})^{1/\beta_i}$. Thus if $\tau_x$ is temperature independent and $n_\mathrm{x}$ has an activation energy $\Delta$, the activation energy for $T_i$ is $\Delta/\beta_i$.

Experimental results and comparison to theory —

Unpublished work of Kitagawa, Takigawa, et al [13] found stretched exponential forms for both relaxation functions, of form (11). The time scales are $T_1 \sim 1$ s and $T_2 \sim 10^{-4}$ s at 0.5 K, growing with decreasing temperature. Eventually they saturate with $T_1 \sim 10^3$ s from 0.2 K downwards, while
$T_2 \sim 10^{-4}$ s from 0.4 K downwards. Both exponents $\beta_i$ decrease with temperature, starting with $\beta_i \approx 1$ (i.e. unstretched exponential) for $T > 1$ K, it would appear each exponent saturates to 1/2 at about the same temperature that the corresponding $T_i$ saturates, so $\beta_1 < \beta_2$ at intermediate temperatures. (As for the $^{17}$Ti NMR relaxations, the temperature was not taken far below $T^*$ so not much can be said about $T \to 0$ behavior, but at the temperatures investigated, both $T_1$ and $T_2$ were an order of magnitude longer than they were for $^{17}$O.)

The low-$T$ limiting behavior has two fundamental contradictions with any diffusing monopole theory. First is the temperature dependence of $T_2$. First, in Eq. (13) the only $T$ dependence comes from the density of flippers $n_x$ (presumed in this picture to be monopole density $n$) or conceivably the flip frequency $\tau_x^{-1}$. In (14) this implies $T_i$ must have an activated temperature dependence, which contradicts the observed saturation at low temperatures. (If $\tau_x$ were also activated, it would just add to the activated dependence.)

The second contradiction is that I found $\beta_2 = 3/2$ in the monopole diffusion regime, contradicting the experimental value $\beta_2 \approx 1/2$, and indicating that $\beta_0 = 1$. But that holds only when the flipper is fixed and flipping rapidly compared to the measurement time, $\tau_x \ll T_2 \sim 10^{-4}$s.

One is forced by the data to assume a density of “flippers” that has negligible temperature dependence. This can only be some kind of quenched disorder in the material, with a density perhaps $10^{-4} - 10^{-3}$ per tetrahedron so as to dominate the monopole density at temperatures below $T^*$, where the temperature dependence of $T_1$ and $T_2$ levels off. From here on, let us accept that the flippers are fixed, giving $\beta_1 = \beta_2 = 1/2$, and see what the experiment tells us about them.

From the two relaxation times $T_1$ and $T_2$ we can solve for the two unknown parameters $\tau_x$ and $n_x$. Eliminating from Eqs. (7), we find

\[
\omega_0 \tau_x \approx \sqrt{2}(T_1/T_2)^{1/2} \quad \text{(13a)}
\]
\[
n^{-1} \approx \left( \frac{\omega_0}{\omega_d} \right) \left( \frac{\omega_0 \sqrt{T_1T_2}}{\tau_0} \right)^{1/2} \quad \text{(13b)}
\]

For Dy$_2$Ti$_2$O$_7$, the two known parameters are $\omega_0/2\pi \approx 20$MHz, and $\omega_d/\omega_0 \approx 1/20$.

Insertion of these and the experimental $T_i$’s into Eqs. (13) yields $\tau_x \approx 3.5 \times 10^{-5}$ s, which is (barely) consistent with the condition $\tau_x \ll T_2$, and $n_x \approx 0.002$, still in units of $a_p^{-3}$.

What can the fixed flippers be? The out-of-equilibrium, back-to-back bound monopole pairs are ruled out: any fluctuations depend on a momentary energy increase, but the rate $\tau_x^{-1}$ would be thermally activated, contrary to observation. Diffusion by nonmagnetic sites produces a very similar situation: pairs of unbalanced tetrahedra, each of which is analogous to a midgap impurity in a semiconductor. The minimum energy state adjacent to the nonmagnetic site is a bound pair of half-monopoles, and the fluctuation rate is again activated.

The nearest out-of-plane spins are almost as close, but their easy axis is oriented exactly so that the two terms in the dipole interaction cancel.

My best guess is that extra magnetic (Dy) spins appear on the nonmagnetic (Ti) site, as in the “stuffed spin ice” [26, 27] but much more dilutely. (Note the Ti sites themselves form a pyrochlore lattice dual to the Dy pyrochlore lattice.) Indeed, it was proposed very recently [28] that in the related material Yb$_2$Ti$_2$O$_7$, around 5% of the Ti sites are occupied by the magnetic Yb ion.

In order for the “stuffed” spins to fluctuate at such with such a short time constant $\tau_0$, they must be practically decoupled from the nearly frozen lattice of regular Dy spins. This decoupling seems plausible when one considers the location of the Ti sites, at the centers of hexagons formed by Dy sites, and assumes the “stuffed” spin has an easy direction along its local three-fold axis. First, the local field at the Ti site due to its six nearest-neighbor pyrochlore spins cancels if they are all oriented in the same sense around the ring, which should happen $\sim 12\%$ of the time. Second, the dipole coupling to its second-nearest Dy spins happens to have an angular factor that exactly cancels. Indeed, it appears from Figure 1 of Ref. [22] that the typical local field at a Ti site is $\sim 0.2$ Tesla or about $\sim 1/20$ of the maximum local field, which occurs on the O(1) sites containing the probe nuclei. [29]

Conclusion —

In conclusion, I have rederived the stretched-exponential form of the NMR relaxation functions due to independent flipping spins at random, distant positions, which might be either fixed impurities (weakly coupled to any ordered or frozen spin background) or else the spin-flips which induce the hopping of emergent monopole defects in spin ice. In particular, monopole hopping implies a novel power of 3/2 in the stretched exponential for the $T_2$ relaxation, in contrast to 1/2 for a fixed magnetic impurity.

Experiment at the lowest temperatures shows, for both $T_1$ and $T_2$ relaxations, a power tending to 1/2 in the stretched exponential and relaxation times tending to a constant, both of which are incompatible with the monopole picture. An analysis was presented that allows extraction of $\tau_x$ and $n_x$ from $T_1$ and $T_2$ with no bias as to the cause (except it depends crucially on both kinds of relaxation being due to the same fluctuations). I suggested that dilute magnetic impurities “stuffed” on the Ti sites are responsible.

It would be interesting to see if NMR relaxation in the higher temperature regime around 0.5 K can be explained by monopoles. This may be more complex, though: there may be no temperature range in which thermally excited monopoles are dense enough to dominate over the “stuffed” impurities, and are at the same time dilute enough for the small density approximations used here.

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It must be checked whether the muons are freely diffusing: see P. Quémerais, P. McClarty, and R. Moessner, preprint [arXiv:1203.3039]. "Possible Quantum Diffusion of Polaronic Muons in Dy2Ti2O7 Spin Ice"

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[10] I have ignored a small anticorrelation between successive hops, due to the dependence of available hopping direction on spin configuration.

[11] The actual nuclei have larger moments but this does not affect the results.

[12] The behavior is practically the same in the case that ω0 is due to a uniaxial anisotropy acting on the nuclear spin (known as nuclear quadrupole resonance), as in the case of the 47Ti nucleus; in any case hω0 is the level splitting.

[13] Indeed, Ref. [22] addressed how the static local field variation due to nearby monopoles splits the NMR line i.e. the ω0 value; but in this paper, our concern is the dynamic effects of distant monopoles.

[14] In fact, it is easy to see (using the Coulomb formula for the monopole’s field) that the accumulated displacement vector. Thus the ΔH jumps can add up without bound.

[15] More exactly, since I assumed the diffusing monopoles are non-interacting, the occupation number has a Poisson distribution with mean nmono. If this is used, the result (9) becomes exact within that assumption.

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[19] Ref. [2] found the “supertetrahedron corners” to have the smallest typical field strengths (these are the centers of tetrahedra of Ti sites.)