Correlated Quantum Tunnelling of Monopoles in Spin Ice

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The spin ice materials Ho2Ti2O7 and Dy2Ti2O7 are by now perhaps the best-studied classical frustrated magnets. A crucial step towards the understanding of their low temperature behaviour – both regarding their unusual dynamical properties and the possibility of observing their quantum coherent time evolution – is a quantitative understanding of the spin-flip processes which underpin the hopping of magnetic monopoles. We attack this problem in the framework of a quantum treatment of a single-ion subject to the crystal, exchange and dipolar fields from neighbouring ions. By studying the fundamental quantum mechanical mechanisms, we discover a bimodal distribution of hopping rates which depends on the local spin configuration, in broad agreement with rates extracted from experiment. Applying the same analysis to Pr2Sn2O7 and Pr2Zr2O7, we find an even more pronounced separation of timescales signalling the likelihood of coherent many-body dynamics.

Introduction — Some of the most exciting discoveries in strongly correlated systems in recent years are related to topological phases of matter. These are novel types of vacua hosting quasiparticle excitations charged under an emergent gauge field. In contrast to Fermi [1, 2] and Luttinger liquids [3], where quasiparticles provide a complete description of all low energy states, here one needs to keep track of the joint time evolution of quasiparticles and gauge fields. Doing this in full generality is a highly non-trivial task [4], and remains largely unexplored, despite the huge interest in the context of, e.g., parton theories of correlated quantum matter [5, 6]. In practice, one can instead resort to largely uncontrolled approximation schemes such as mean-field treatments in which the particle moves in an averaged background gauge field.

The spin ice compounds Ho2Ti2O7 (HTO) and Dy2Ti2O7 (DTO) [7] are generally believed to host a topological Coulomb spin liquid [8]. Here, the emergent gauge field is particularly simple to visualise, as its gauge flux is encoded in the spins themselves. The motion of a quasiparticle amounts to spin flip processes, which are subject to the energetics and quantum dynamics of the underlying many-body Hamiltonian. Despite the elegance of this physical scenario, a comprehensive understanding of the large variety of experimental timescales [9–16] is still lacking. Puzzling timescales are not uncommon in geometrically frustrated magnets [17].

In this paper, we report an analysis of the elementary building block of quasiparticle motion, with the detailed microscopic knowledge on spin ice available in the literature [7, 8] as foundation. We study the local dynamics of emergent monopole excitations, which has a quantum mechanical (tunnelling) origin, rooted in the transverse terms of the dipolar and exchange interactions between rare-earth (RE) ions [18, 19]. We focus on elastic processes (monopole hopping), since inelastic ones (monopole creation/annihilation) are suppressed at low temperatures. The key question is: how do the predominantly off-diagonal terms (‘transverse fields’) necessary to induce monopole hopping arise in a material
whose statistics are excellently described by a classical Ising model? Our central result is that there is a fundamental feedback mechanism between spin dynamics, monopole quasiparticles, and the local spin environment. Whereas the vast majority of spins in the sample experience longitudinal fields, which justify a classical description, some of the spins adjacent to a monopole experience predominantly transverse fields. As illustrated in Fig. 1, a monopole has 3 available lattice bonds to hop across, and, statistically, we find a bimodal distribution of transverse fields, and hence of quasiparticle hopping rates, in ratio 2:1 (fast:slow). We posit that these $\tau^{\text{fast}}$ and $\tau^{\text{slow}}$ are the fundamental (tunnelling) timescales underlying a broad range of dynamic phenomena in spin ices [9–16] and find they are consistent with experimental timescales (Table I). We extend our calculations to ‘quantum spin ice’s’ Pr$_2$Sn$_2$O$_7$ (PSO) and Pr$_2$Zr$_2$O$_7$ (PZO) and find, as expected, much faster timescales and also, more surprisingly, much greater separation between $\tau^{\text{fast}}$ and $\tau^{\text{slow}}$. Finally we argue that decoherence may play an essential role in the emergence of slow, classical spin flips out of fast, quantum tunnelling and we provide a simple model based on the Zeno effect. Interestingly the large separation of timescales in PSO and PZO implies a coexistence of coherent and incoherent processes.

Model — Spin ices are magnets where anisotropic Ising-like spins reside on a pyrochlore lattice of corner-sharing tetrahedra. The exchange and dipolar interactions between the spins are largely frustrated, and at low temperatures the ground state is described by an extensively degenerate manifold of configurations obeying the so-called ‘ice rules’ (in each tetrahedron, 2 spins point ‘in’, towards its centre, and 2 point ‘out’) [7]. The lowest excitations above such ground state are effective magnetic monopoles with Coulomb interactions [8].

The local Ising anisotropies originate from the strong crystalline-electric-fields (CEF) acting on the J-manifold of the RE$^{3+}$ ions (for Ho$^{3+}$, Dy$^{3+}$ and Pr$^{3+}$ ions, the total angular momentum quantum number is, respectively, $J = 8, 15/2$ and 2) [19, 20]. Low energy dynamics between the single-ion states of the ground-state doublet, $|\pm\rangle_i$ and $|\mp\rangle_i$ (labelled by $S_i = -1, 1$), necessarily involve transitions via the CEF excited states, with energies $\Delta E \gg 10^2$ K [21, 22]. In the temperature range where the monopole description is valid ($T \lesssim 1$ K), thermal activation of CEF excited states is negligible so that quantum tunnelling must underpin the spin dynamics [9]. This provides a mechanism for the flipping of the minority of spins that are not frozen by a local (longitudinal) combined dipolar and effective exchange field. These are of course the flippable spins next to a monopole.

We focus on a given spin, say at $i = 0$, to study the single spin-flip dynamics which amounts to the hopping of a monopole. Our Hamiltonian,

$$\hat{H}(0) = \hat{H}_{\text{CEF}} + \hat{H}_{\text{dip}}(0) + \hat{H}_{\text{exc}}(0),$$

(1)
describes a RE-ion at site 0 of an N-site pyrochlore system. $\hat{H}$ acts on the Hilbert space of the RE$^{3+}$ of interest with total angular momentum quantum number $J$. (We work in the $2J + 1$ dimensional $|M\rangle \equiv |J,M\rangle$ eigenbasis of $\hat{J}_z$ for the local quantisation axis $\hat{z}_0 \propto \langle 111 \rangle$.) $\hat{H}_{\text{CEF}}$ is the crystal-field Hamiltonian [19], and $\hat{H}_{\text{dip}}(0)$ and $\hat{H}_{\text{exc}}(0)$ describe, respectively, dipolar and exchange interactions with other RE$^{3+}$ ions.

Our main approximation for Eq. (1) is that each of the other $N - 1$ spins in the system is projected onto one of its own CEF ground-state doublet states, $|\pm\rangle_j$ (i.e. $S_j = \pm 1$ for $j \neq 0$). We thus ignore joint dynamical correlations that may develop in the simultaneous motion of all flippable spins (e.g., spins $S_0, S_1, S_5$ in Fig. 1) we use to be studied together, as potentially entangled spins [23]). Notice however that the simultaneous re-orientation of two or three of the spins $S_0, S_4, S_5$ in Fig. 1 produces higher excited states; and that the re-orientation of one of them generates a longitudinal field pinning the other two in their initial state. This observation supports the validity of our approximation.

Dipolar interactions — Let us consider the simplest pyrochlore system of interest for the hopping of a monopole: two adjacent tetrahedra, where only one tetrahedron hosts a monopole and flipping the central spin $S_0$ allows the monopole to hop to the other tetrahedron. There are only 3 symmetry-inequivalent such spin configurations, illustrated in Fig. 1.

Using the conventional dipolar interaction [7], it is straightforward to show that the 6 neighbouring spins exert a field on the central site given by

$$B_{\text{dip}}^{(6)}(0) = \frac{\mu_0 |m|}{4\pi r_{3\text{nn}}} \sum_{j=1,2,3} (\hat{z}_j + \sqrt{6}\hat{r}_j) (S_j + S_{j+3}).$$

(2)

As represented in Figs. 1a-1c, spin pairs ($S_j, S_{j+3}$) with $j = 1, 2, 3$, have the same anisotropies $\hat{z}_j = \hat{z}_{j+3}$ and opposite n.n. positions $\hat{r}_j \equiv \hat{r}_{0j} = r_{\text{nn}} \hat{r}_j = -r_{0j+3}$, with $|\hat{z}_j| = |\hat{r}_j| = 1$.

Using Eq. (2), we find that all 2-tetrahedron configurations hosting one monopole next to a flippable spin (Fig. 1) have vanishing longitudinal component, $B_{\text{dip}}^{(6)}(0) \cdot \hat{z}_0 = 0$, as expected for flippable spins. Remarkably, such spin configurations do not all give the same (transverse) fields: in 2/3 of the cases, Figs. 1a-b, $B_{\text{dip}}^{(6)}(0)$ is finite and points along one of the high-symmetry CEF angles $\phi_n$ of Ref. [19]; whereas in the remaining 1/3, Fig. 1c, $B_{\text{dip}}^{(6)}(0) = 0$ identically, since $r_0$ is a centre of inversion [24].

Because of corrections to the projective equivalence [25], dipolar-fields from farther spins ($N - 1 > 6$) can alter our conclusions from the 2-tetrahedron system. To check this, in Fig. 1d we compare the field-distribution of $B_{\text{dip}}^{(6)}$ (left panel) and $B_{\text{dip}}^{(24)}$ (right panel) compatible with the monopole constraint in Figs. 1a-1c ($B_{\text{dip}}^{(24)}$ includes the farther 18 spins belonging to...}
the next 6 tetrahedra adjacent to the 2 tetrahedra at \( r_1, \ldots, r_6 \). The histograms in Fig. 1d show that the bimodal field-distribution is qualitatively unchanged: the two values for \( B_{\text{exc}}^{(6)} \) evolve into two well separated distributions for \( B_{\text{exc}}^{(24)} \approx 0.45 \) Tesla in 2/3 of the cases, and \( B_{\text{exc}}^{(24)} \approx 0.33 \) Tesla in 1/3. Note that the differences in values between \( B_{\text{dip}}^{(6)} \) and \( B_{\text{dip}}^{(24)} \), and in particular the non-zero spreads, are due to quadrupolar corrections, which are well-captured by the 24 spin calculation. Indeed, the histograms in Fig. 1d agree quantitatively with Monte Carlo simulations on larger systems [26].

These results imply that the associated spin dynamics is remarkably correlated to the local environment. For a flippable spin next to a monopole, two very distinct flipping rates, \( \tau_{\text{fast}} \) and \( \tau_{\text{slow}} \), appear, with a 2:1 ratio. In the following we show that the same comes to pass for the full fledged form of the exchange interactions.

**Exchange interactions** — To achieve a realistic model of exchange couplings in RE\(^{3+}\) pyrochlores, we first write

\[
\hat{H}_{ff}(r,r') = \sum_{m_1, m_2} \frac{\delta m_1 m_2}{\mu_0} \sum_{\sigma_1, \sigma_2} \langle \hat{F}_{m_1, \sigma_1} \hat{F}_{m_2, \sigma_2} \rangle |x^m_1| + |x^m_2| + \frac{1}{2} \left[ a \delta m_1 m_2 \delta m'_1 m'_2 + \langle R_{\sigma_1} R_{\sigma_2} \rangle m_1 m'_2 \langle R_{\sigma_1} R_{\sigma_2} \rangle m'_1 m_2 \right],
\]

(3)

the Hamiltonian for the oxygen-mediated super-exchange of \( f \)-electrons between two n.n. RE\(^{3+}\) ions at \( r \) and \( r' \). Eq. (3) generalises Eq. (18) in Ref. [27] — originally written for Pr\(^{3+}\) pyrochlores — to any RE\(^{3+}\) pyrochlore (details in Ref. [28]). The operator \( \hat{F}_{m, \sigma} \) (\( \hat{F}_{m, \sigma} \)) creates (annihilates) at \( r \) an \( f \)-electron with orbital and spin magnetic quantum numbers \( m = 0, \pm 1 \) and \( m_\sigma = \sigma/2 \), respectively (\( \sigma = \pm 1 \)). \( R_{\sigma_1} R_{\sigma_2} \) matches the local systems of coordinates at \( r \) and \( r' \). The parameters \( \varepsilon_{\text{exc}} = 2 \left( \frac{V_{\text{exc}}}{U_{\text{exc}}} \right)^2 \left( \frac{1}{U_{\text{exc}}} - \frac{1}{U} \right) \), \( a = U/(\Delta - U(n + 1)) \), and \( x = V_{\text{exc}}/V_{\text{exc}} \) contain the complex relationships between the \( n \) electrons in the \( f \)-shell, their (repulsive) Coulomb energy \( U \), the change in energy \( \Delta \) for the removal of an electron from the oxygen, and, most importantly, the Slater-Koster hybridisation parameters \( V_{m = \pm 1} = V_{\text{exc}} \) and \( V_{m = 0} = V_{\text{exc}} \).

We then project each n.n. ion, as we did for the case of dipolar interactions, to obtain the exchange Hamiltonian

\[
\hat{H}_{\text{exc}}(0) = \sum_{j=1}^{6} \left( \langle \pm | \hat{H}_{ff}(r_0, r_j) | \pm \rangle \right),
\]

(4)

which operates in the 2\(J + 1\) dimensional Hilbert space of the central ‘single-ion’. This is a highly anisotropic Hamiltonian that cannot be easily interpreted as a field distribution \( \hat{H}_{\text{exc}} \neq -g_{\mu_B} J \cdot B_{\text{exc}}^{(6)} \). We study it by considering \( \langle \hat{J} \rangle = \langle \psi | \hat{J} | \psi \rangle \), where \( | \psi \rangle \) is the ground state of \( \hat{H}_{\text{exc}}(0) = \hat{H}_{\text{CEF}} + \hat{H}_{\text{exc}}(0) \). Once again, the spin configurations in Fig. 1 exhibit a bimodal behaviour: in 2/3 of the cases, \( \langle \hat{J} \rangle = 0 \) (e.g., Fig. 1e, 1f, 1g). As a matter of fact, for this inversion-symmetric case, \( \hat{H}_{\text{exc}}(0) \) is diagonal in the (\( | M \rangle \) basis, and symmetric under \( M \leftrightarrow -M \).

The above behaviour holds for any \( E_{\text{exc}} \), \( a \) and an, as long as \( \hat{H}_{\text{exc}} \) is a small perturbation to \( \hat{H}_{\text{CEF}} \). Notwithstanding, we summarise here how we set these parameters to obtain quantitative results (further details can be found in the Supplemental Material [29]). Firstly, a relationship between \( a \) and \( x \) is found by requiring the diagonal part of \( \hat{H}_{ff}(r, r') \), projected onto the ground state CEF doublet manifold of each of the two spins involved, to be proportional to \( \sigma_x \otimes \sigma_x \), which is a central feature in both classical and quantum (n.n.) spin-ice Hamiltonians [7, 27]. Then the behaviour of \( \hat{H}_{\text{exc}}(0) \) projected on the GS doublet of the central spin is compared with \( \hat{H}_{\text{exc}} = J_{\text{min}} \sum_{j=1}^{6} \sigma_{\varphi_1} \otimes \sigma_{\varphi_2} \) for given configurations of the 6 outer spins. The value of \( J_{\text{min}} \) obtained experimentally, sets therefore \( \varepsilon_{\text{exc}} \) as a function of \( x \).

We are finally left with only one parameter, \( x \), which was argued in Ref. [30] to vary in the range \((-1, 0)\). We study its effect by looking at the behaviour of the central spin under the single-ion Hamiltonian \( \hat{H}_{\text{CEF}} + \hat{H}_{\text{exc}}(0) \) derived from a 2-tetrahedron system where the 6 outer spins are projected on different CEF GS configurations. We observe no appreciable change in HTO and DTO: the results are consistent throughout the range with the known sign of the interaction and a nearly fully polarised GS dipole moment. The same is not true for PSO and PZO, and in particular the correct sign of the exchange interactions (opposite to HTO and DTO) occurs only for \( x \in (-1, -0.3) \) in PSO and for \( x \in (-1, -0.6) \) in PZO. Beyond these values we observe a reversal of the GS dipole moment, with corresponding closing of the gap, signalling a change between ferromagnetic and antiferromagnetic nature of the exchange interactions. Such phenomena are worthy of further investigation in their own right, but are beyond the scope of the present work. Here, we find it sufficient to set \( x = -1 \) for all systems.

**Spin dynamics and timescales** — We study the unitary spin dynamics of the central spin under the influence of both dipolar and exchange interactions by means of Eq. (1). In Fig. 2 the magnetic moment \( \langle \hat{J} \rangle = \langle \hat{J}_z \rangle \), initialised in \(-1\), completely reverses direction (\(+1\)) with a precession timescale \( \tau = \hbar/(\Delta E_{J}) \) (\( \tau_{\text{fast}} \) and \( \tau_{\text{slow}} \) correspond to the cases illustrated in Fig. 1a-1b and Fig. 1c, respectively). For HTO and DTO, Figs. 2a-2b, it is worth noting that, in spite of having weak transverse terms [30], exchange does make a quantitative difference (e.g., in DTO dipolar fields alone give \( \tau_{\text{fast}} \approx 53 \) ms, in contrast to \( 1.5 \) ms for the full Hamiltonian). In PSO and PZO...
Figure 2. Time-evolution of the probability density $P_M(t) = |\langle M|\psi(t)\rangle|^2$ (black regions, $P_M(t) \approx 1$) as a function of $M = -J, \ldots, J-1, J$ and $t$ (sec), as discussed in the main text. On each density plot is overlaid the curve $\langle J_z(t) \rangle$.

(Figs. 2c-2d), $\tau_{fast}$ is several orders of magnitude shorter than for HTO and DTO, consistent with the expectation of much stronger quantum fluctuations.

| $|x| = 1$ | HTO | DTO | PSO | PZO |
|----------|------|------|-----|-----|
| $\alpha$ | -0.17 | -0.17 | -0.20 | -0.19 |
| $\varepsilon_{\text{exc}}$ | 0.15 | 0.22 | -32 | -87 |
| $\tau_{fast}$ | $7.4 \times 10^{-6}$ | $1.5 \times 10^{-6}$ | $5.4 \times 10^{-10}$ | $7.9 \times 10^{-12}$ |
| $\tau_{slow}$ | $2.0 \times 10^{-4}$ | $4.4 \times 10^{-2}$ | $1.0 \times 10^{-4}$ | $1.0 \times 10^{-4}$ |
| $\tau_{\text{exp}}$ | $10^{-5}$ [22] | $10^{-5}$ [31] | $10^{-11}$ [32] | $10^{-12}$ [34] |

Table I. Key-quantities in different compounds. Rows 2-3: estimations of $\alpha$ (dimensionless) and $\varepsilon_{\text{exc}}$ (meV) in Eq. (3). Rows 4-5: corresponding fast and slow tunnelling timescales – unitary evolution under Eq. (1) with experimental parameters from Refs. [7, 20, 33, 34]. Row 6: experimental timescales from the literature. All timescales $\tau$ are expressed in seconds.

**Experimental timescales (HTO, DTO)** — It is worth contrasting our timescales in Tab. I with the experimental ones in the literature. On the one hand, $\mu$SR experiments report $\mu$s timescales due to persistent spin dynamics [35–37]. On the other, AC-susceptibility measurements report ms timescales for relaxation in DTO [13, 14]. Suggestively, such two timescales have the same order of magnitude as $\tau_{fast}$ and $\tau_{slow}$, respectively, and it is tempting to look for a direct correspondence. However, we hitherto neglected any source of decoherence. Even the fast timescales for HTO and DTO in our work are relatively ‘slow’ in that respect. Experiments on molecular spin-qubits with Ho$^{3+}$ ions in the same point symmetry $D_{3d}$ yield decoherence times up to 50 $\mu$s at best [38]. In conventional spin ice experimental settings, we therefore expect decoherence timescales faster than spin ice dynamics, by orders of magnitude. The effective spin-flip time can then be much longer than the precession time, due to the environment projecting the time-evolved state back to its initial one – a phenomenon called quantum Zeno effect [39, 40]. The effect can be illustrated by considering a toy model – details can be found in the Supplemental Materials [29] – of a precessing pseudospin-1/2 degree of freedom (with precession time $\tau$ caused by a transverse field), coupled to an effective bath at exponentially-distributed random times (with an ‘observation’ time constant $\tau_o$). The bath takes the form of projections onto the states $|\pm\rangle$. A fast-decoherence regime $\tau_o \ll \tau$ results in a large effective spin-flip timescale $\langle \Delta t \rangle \sim \tau^2/\tau_o$. In HTO and DTO, this could reconcile the AC experimental timescales ($\sim$ ms) with $\tau_{fast}$ ($\sim \mu$s). These values require spin-decoherence timescales of order $0.1 - 1$ ns, which are plausible. Notice that, under this assumption, $\tau_{slow}$ becomes of the order of 1 s.

**Conclusions** — Our work highlights an intriguing and hitherto poorly understood correlation in the dynamics of spin ice models and materials, whereby a monopole alters locally the spin background, and the latter (pre)determines whether and how fast the former can hop. Specifically, the arrival of a monopole in a tetrahedron quenches the longitudinal fields acting on its spins, inducing a bimodal distribution of temperature-independent spin-tunnelling timescales dictated solely by the CEF, dipolar and exchange transverse terms.

We believe that this is crucial for understanding the mechanism of hopping of a monopole [41] and, more in general, of the dynamics below the ‘quantum-classical’ crossover at $T \approx 13$ K [14, 22, 31]. While at higher temperatures, monopole diffusion is well-understood in terms of thermal population of CEF levels [22], the predictions of that theory differ considerably from the behaviour observed at lower temperatures. In contrast, the broad agreement of the experimental low-temperature timescales with our predictions, and their temperature independence, provide evidence in favour of our theory.

In classical spin ice (HTO and DTO) our finding of $\tau_{fast}$ and $\tau_{slow}$ may have important implications on the response and equilibration properties. For instance, Monte Carlo simulations used to model AC susceptibility [41–43] ought to be modified to account for the two timescales discussed in our work, which can lead to measurable effects [44]. Ascertainment the extent to which the modified scenario improves agreement with experiments deserves dedicated studies, beyond the scope of this paper. Furthermore, that $\tau_{fast}$ and $\tau_{slow}$ turn out to be longer than the expected decoherence times in HTO and DTO is one of our principal conclusions. It implies that any theory of the long experimental timescales and other puzzling dynamical phenomena (e.g., the falling out of
equilibrium at 600 mK) must take decoherence into account. Our simple one, based on the Zeno effect, shows how this might work in terms of one adjustable parameter τ₀. A predictive theory of τ₀ will probably need to include degrees of freedom ‘extrinsic’ to the spins (which thus become an open quantum system decohering via quantum dissipation induced by their environment [45]). This remains an outstanding challenge in the field.

In quantum spin ice (PSO and PZO) the separation in timescales becomes remarkably large, with \( \tau \) fast potentially shorter than the decoherence time scale, consistently with expectations. This is in agreement, for example, with Ref. [32], where timescales for PSO – extracted from quasi-elastic neutron scattering experiments – were contrasted to the ones for DTO and HTO. The fast timescales in PSO and PZO suggest the need for a few-body dynamical description incorporating coherent longer-range hopping processes for the monopoles (e.g., flips of \( S_0 \) and \( S_3 \) in Fig. 1a) which cannot be described within our current approximations and is thus a further challenge for future work. Our results also imply that the dynamics corresponding to the slow time scale will not occur quantum-coherently. It will be interesting to determine which observable consequences this may have.

In particular, it provides us with the intriguing possibility of an approximate modelling of these systems that, à la Born-Oppenheimer, focuses on a quantum mechanical description of the fast processes, and a classical stochastic description of the slow ones, thus substantially reducing the complexity of a three dimensional strongly correlated quantum system [46].

Finally, we note that since a monopole breaks the inversion symmetry of the tetrahedron, it is expected to generate an electric dipole moment [47]. This may alter the crystal electric fields and split the g.s. doublet in non-Kramers systems, potentially affecting the hopping timescales in HTO, PSO and PZO. In contrast, DTO and other Kramers system should remain unperturbed.

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References:
ter would require coupling to other degrees of freedom which are not present in our model, e.g., lattice distortions or spins tilting away from the easy axes.

[26] G. Sala, (private communication).
[29] See Supplemental Material (SM) at [URL will be inserted by publisher] for more details.
[44] Thus far, experimental AC susceptibility is only in coarse agreement with Monte Carlo simulations.
[46] On average, there are two fast spins per tetrahedron, and the corresponding quantum many body system is a network of coordination 2 (1D on average) with occasional 3-way branching and end points.