Monopolar and dipolar relaxation in spin ice $\text{Ho}_2\text{Ti}_2\text{O}_7$

**Scientific Achievement**

Two distinct magnetic relaxation processes were discovered in the spin-ice compound $\text{Ho}_2\text{Ti}_2\text{O}_7$. The cross-over in the relaxation dynamics is associated with spin fractionalization into monopoles.

**Significance and Impact**

While dipolar relaxation dominates at higher $T$, a unique low $T$ regime with exponentially activated Debye-like relaxation is associated with monopole motion through the spin-ice vacuum.

**Research Details**

- A time resolved neutron scattering technique was developed to probe slow magnetic dynamics (ms to hours) with atomic scale spatial resolution
- A new class of ultra-pure $\text{Ho}_2\text{Ti}_2\text{O}_7$ crystals grown by a travelling solvent floating zone method was needed to manifest individual monopole dynamics
- Observing this regime in $\text{Ho}_2\text{Ti}_2\text{O}_7$ is encouraging for the prospects of coherent quantum dynamics of monopoles in quantum siblings such as $\text{Ce}_2\text{Zr}_2\text{O}_7$

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Yishu Wang, T. Reeder, Y. Karaki, J. Kindervater, T. Halloran, N. Maliszewskiy, Yiming Qiu, J. A. Rodriguez, S. Gladchenko, S. M. Koohpayeh, S. Nakatsuji, C. Broholm, *Sci. Adv., 2021; 7: eabg0908 (2021).*
Spin ice and magnetic monopoles at zero magnetic field

- Spin ice: ferromagnetic Ising spins + pyrochlore lattice → 2-in-2-out
- Zero-field neutron scattering reveals absent diffraction at Brillouin zone centers and diffuse intensity at zone boundaries.
- Successive spin-flips via quantum tunneling should fractionalize a spin dipole into a pair of magnetic monopoles.
Time-resolved neutron scattering under a small field

- Spin ice ground state is unperturbed. \( \mu H \ll k_B T, J_{\text{eff}} \)
- Spins aligned along the field direction give rise to magnetic scattering at the zone centers.
- The magnetization process relies on the thermal excitation.

![Arrhenius form](image)

\[ \tau = \tau_0 \exp \left( \frac{\Delta}{T} \right) \]

\( \Delta = 18(2) \) K
\( J_{\text{eff}} \approx 1.8 \) K

![Diagram](image)
AC susceptibility in a broad range of $T$ and frequency

$$\chi(\omega = 2\pi f) = \chi' - i\chi''$$

![Graph showing AC susceptibility with varying temperatures and frequencies.](image)
Thermal crossover revealed by AC susceptibility

\[ \chi(\omega = 2\pi f) = \chi_l + \chi_h = \frac{\chi_{0l}}{(1 + i\omega\tau_l)^{\beta_l}} + \frac{\chi_{0h}}{(1 + i\omega\tau_h)^{\beta_h}} \]

- \( \beta_l \approx 0.8 \): close to a single time scale.
- \( \beta_h \approx 0.5 \): a broad distribution of time scale.

\( \chi_0 \) — static susceptibility
\( \tau \) — time scale
\( 0 < \beta < 1 \) — distribution of time scale

Extend \( \tau-T \) to high \( T \) limit:
\( \tau_l \approx 10^{-7} \text{s}, \tau_h \approx 10^{-9} \text{s} \)

T(K)

\(|\chi_o(\text{emu Oe}^{-1}\text{cm}^{-3})|

0 0.25 0.5

0 0.6 1.0 1.4 1.8 T(K)

low T coherent, slow
magnetic monopoles

high T incoherent, fast
spin dipoles
Sensitivity to disorder

- Reduced disorder slows dynamics, making two time-scales experimentally distinguishable.
- Debye relaxation is approached at low-$T$ in the pure crystal.

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Current work compared to previous studies:

- Quilliam et al., Phys. Rev. B (2011)
- Eyvazov et al., Phys. Rev. B (2018)