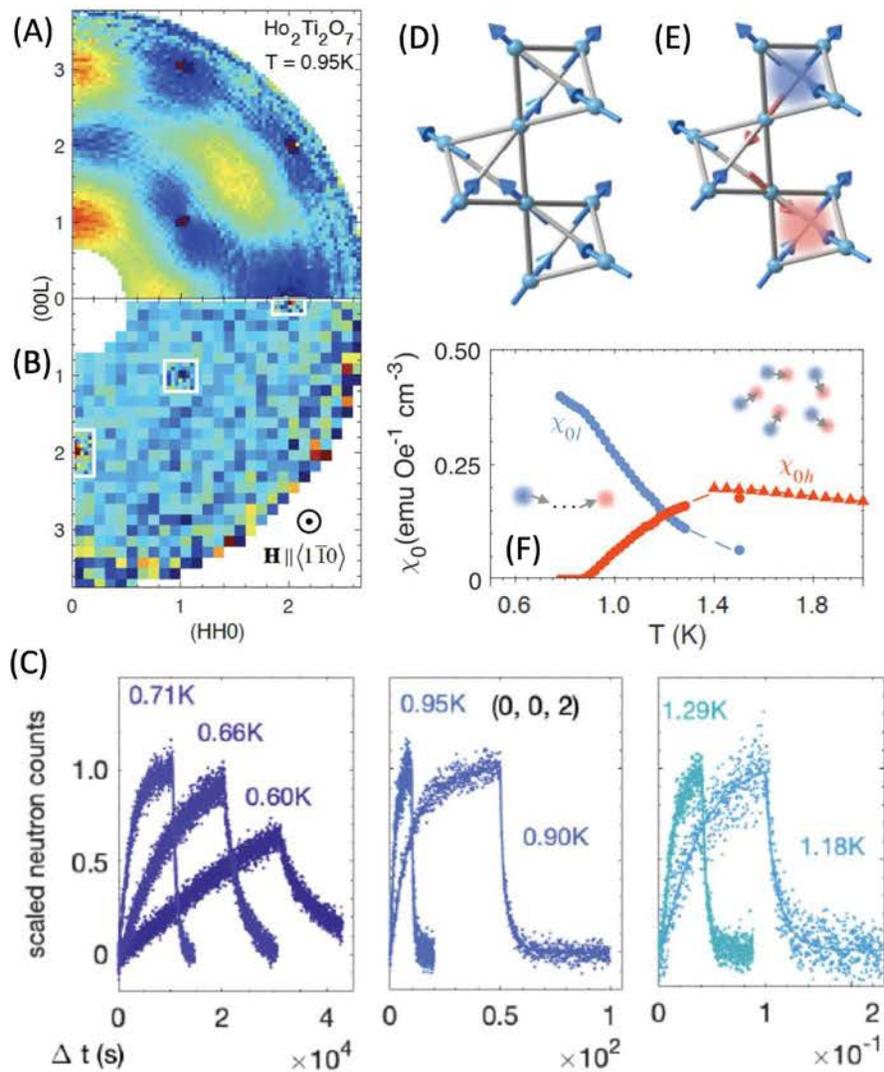


Monopolar and dipolar relaxation in spin ice $\text{Ho}_2\text{Ti}_2\text{O}_7$



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.

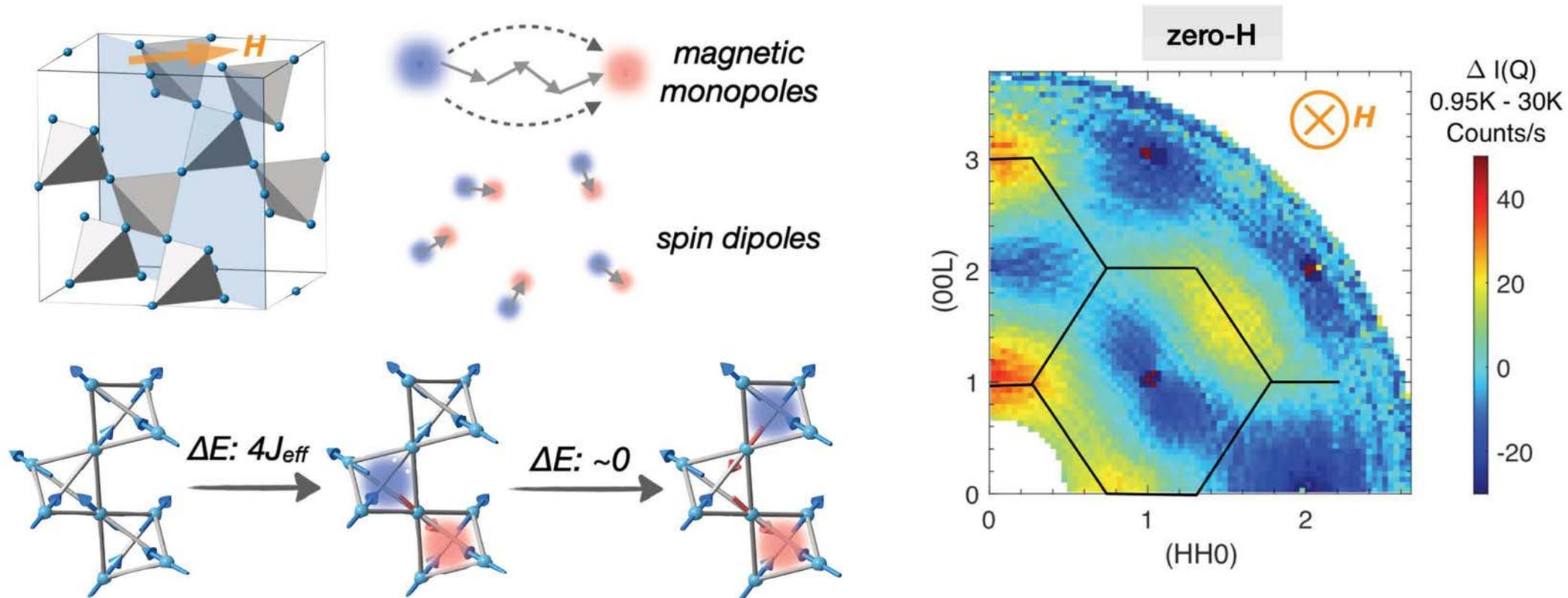
Research Details

- 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 $\text{Ce}_2\text{Zr}_2\text{O}_7$

Yishu Wang, T. Reeder, Y. Karaki, J. Kindervater, T. Halloran, N. Maliszewskij, Yiming Qiu, J. A. Rodriguez, S. Gladchenko, S. M. Koohpayeh, S. Nakatsuji, C. Broholm, [Sci. Adv. 2021; 7: eabg0908 \(2021\)](https://doi.org/10.1126/sciadv.2021.7.eabg0908).

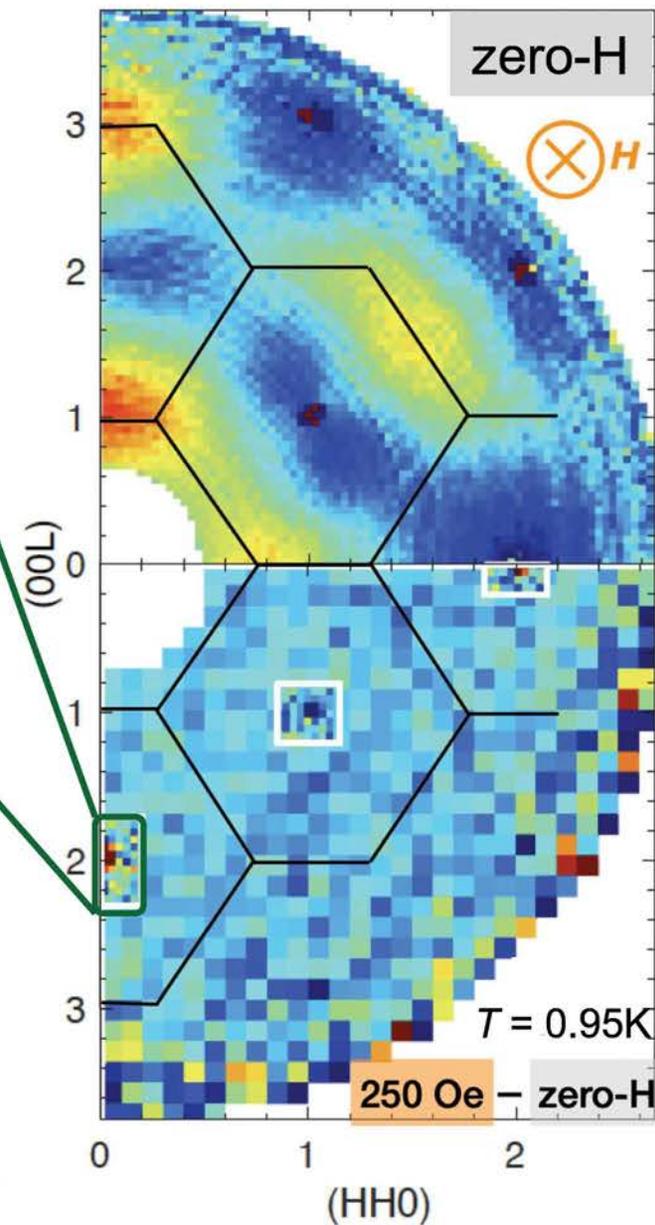
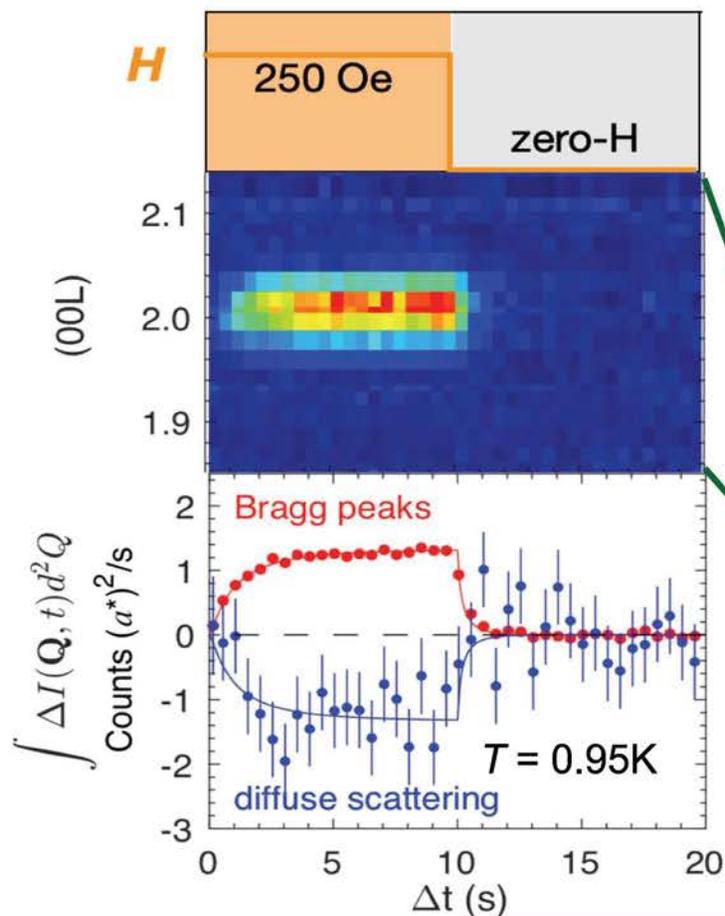
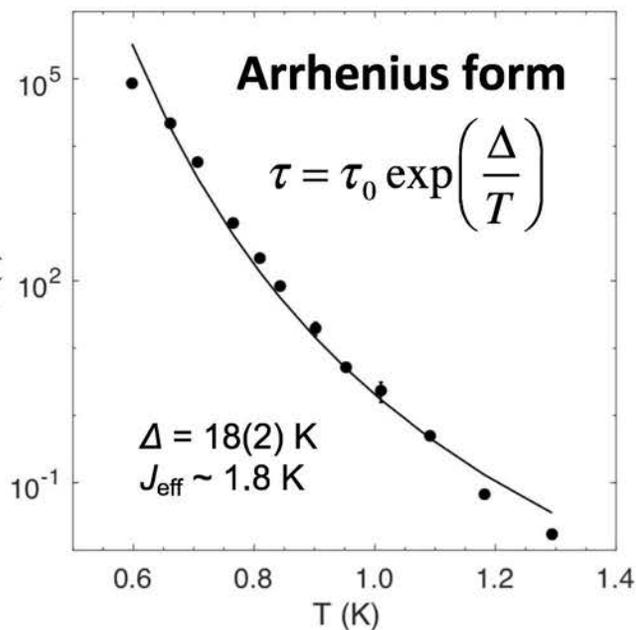
Spin ice and magnetic monopoles at zero magnetic field

- Spin ice: ferromagnetic Ising spins + pyrochlore lattice \rightarrow 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.



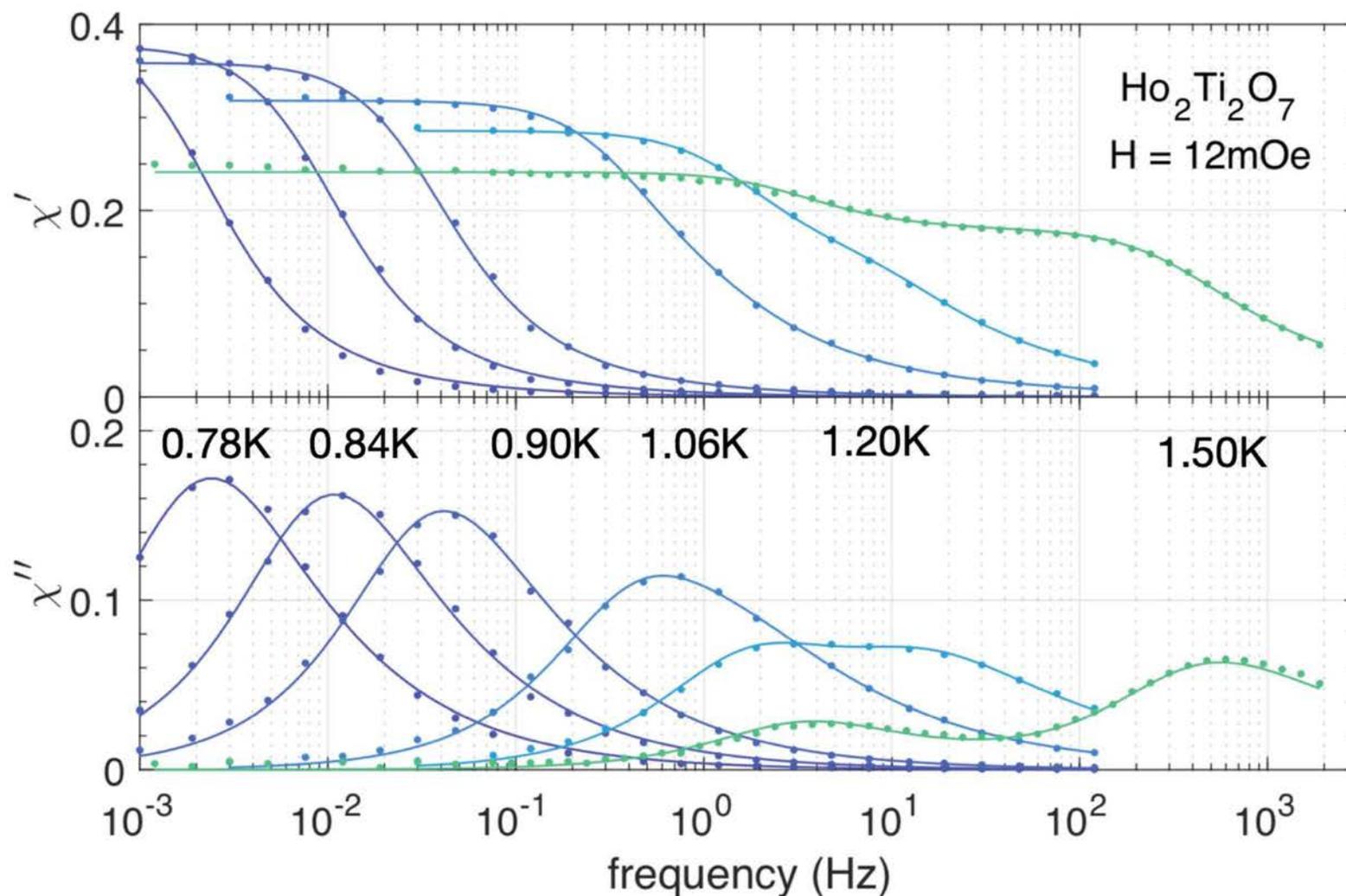
Time-resolved neutron scattering under a small field

- 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.



AC susceptibility in a broad range of T and frequency

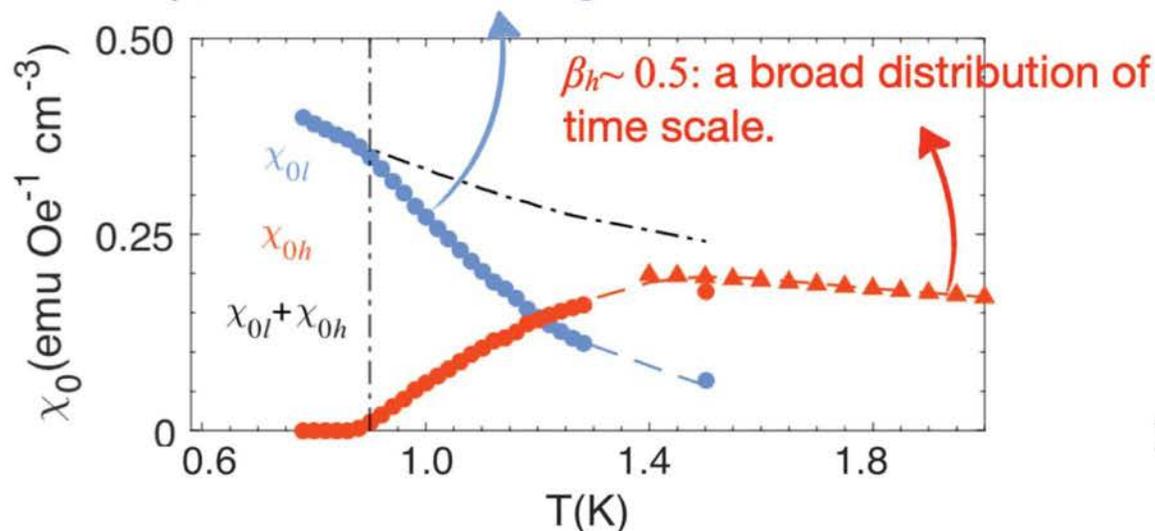
$$\chi(\omega = 2\pi f) = \chi' - i\chi''$$



Thermal crossover revealed by AC susceptibility

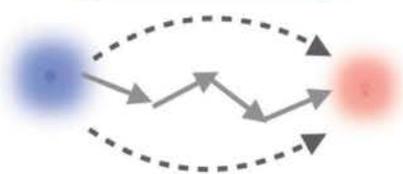
$$\chi(\omega = 2\pi f) = \chi_l + \chi_h = \frac{\chi_{0l}}{(1+i\omega\tau_l)^{\beta_l}} + \frac{\chi_{0h}}{(1+i\omega\tau_h)^{\beta_h}}$$

$\beta_l \sim 0.8$: close to a single time scale.



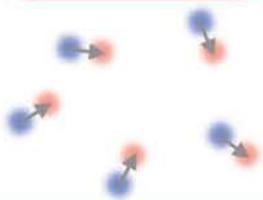
low T
coherent, slow

magnetic
monopoles



high T
incoherent, fast

spin dipoles

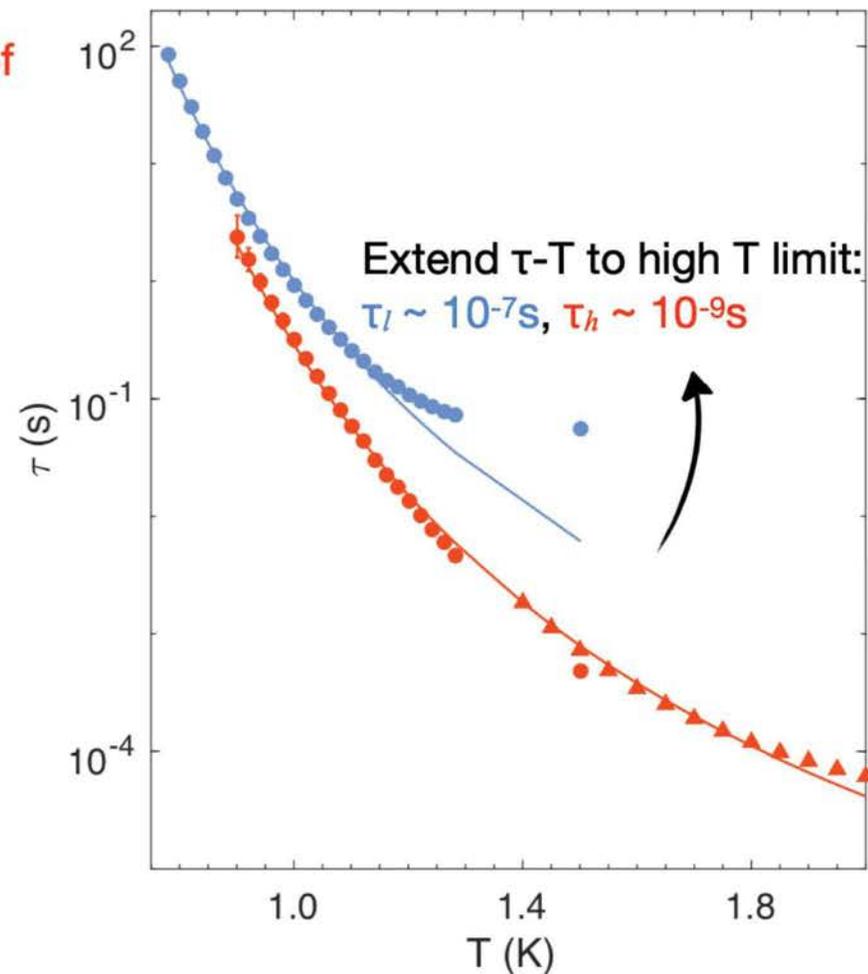


Cole-Davidson form

χ_0 — static susceptibility

τ — time scale

$0 < \beta < 1$ — distribution of time scale



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Sensitivity to disorder

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

