Title: Dipolar spin ice states with fast monopole hopping rate in the spinels CdEr2X4 (X=Se,S)

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Abstract: CdEr2Se4, a spinel, was shown to be the first spin ice in a crystal structure other than the rare earth pyrochlore [1]. Although it has the correct entropy, the exact nature of the spin ice state therein, especially the form of the spin correlation function was not further established. A further particularity was the spin relaxation time, which, at low temperature, was found to display a similar activation energy to that of a canonical spin ice, yet the dynamics are three orders of magnitude faster. Using diffuse neutron scattering, we established that the spin correlations in both CdEr2Se4 and CdEr2S4 are well modeled by the dipolar spin ice Hamiltonian, and used this to parameterize the magnetic Coulomb gas existing in each compound. Both are dilute and non-interacting, as in canonical spin ices, so the monopole population alone cannot account for the enhanced dynamics. By a combination of conventional and high frequency susceptibility measurements, and neutron spin echo spectroscopy, we examine the full temperature dependence of the relaxation time, locating the previously known low temperature thermally activated regime [1], and the uncharacterized intermediate plateau and high temperature thermally activated regime, all as in a canonical spin ice but with much faster timescales. Following the approach of Tomasello et al.[2], we find that the crystal field Hamiltonian of CdEr2X4, as parameterized by our inelastic neutron scattering experiments, supports the faster monopole dynamics primarily through increased susceptibility to transverse fields. Ultimately CdEr2X4 are dipolar spin ices with dilute magnetic Coulomb gases, in which fast monopole dynamics are produced by an increased hopping rate.

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Dipolar spin ice states with fast monopole hopping in the spinels $CdEr_2X_4$ (X=Se, S)

International Workshop on Quantum Spin Ice, June 2017

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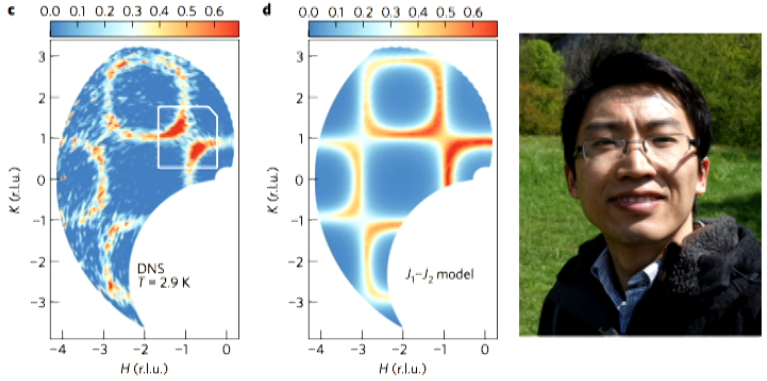
- Neutron Scattering: B Fåk, A Wildes, M Koza, C Ritter, P Fouquet (ILL); L Keller, E Canevet (PSI)
- Heat capacity: S Vrtnik, J Luzar (Josef Stefan Institute)



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FNSNF
Swiss National Science Foundation

$MnSc_2S_4$: SG *et al.*, Nat. Phys. (2016)

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PHYSICAL REVIEW B **72**, 054411 (2005)

Geometrical magnetic frustration in rare-earth chalcogenide spinels

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$CdEr_2Se_4$: A New Erbium Spin Ice System in a Spinel Structure

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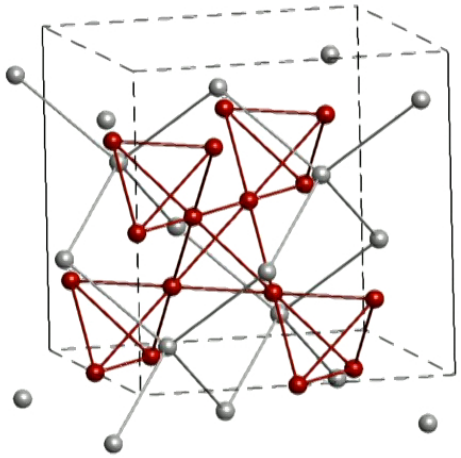
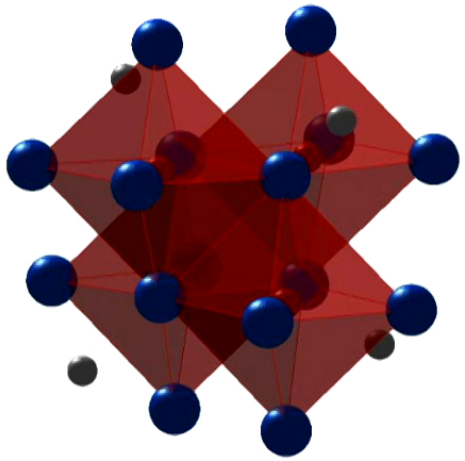
Spinel structure of $CdEr_2X_4$

TABLE I. Structural parameters for $CdEr_2S_4$ at room temperature. Space group: $Fd\text{-}3m$ (#227); lattice constant $a = 11.1178(4)$ Å. $\chi^2 = 1.23$; $R_{wp} = 11.25$; $R_p = 8.61$.

Atom	Position	x	y	z	Occ.	B_{eq}
Cd	8*b*	1/8	1/8	1/8	1	1.40(5)
Er	16*c*	1/2	1/2	1/2	1	1.14(4)
S	32*e*	0.2541(2)	0.2541(2)	0.2541(2)	1	1.50(6)





Lau *et al.*, PRB (2005)

Why an erbium spin ice in the spinel structure?

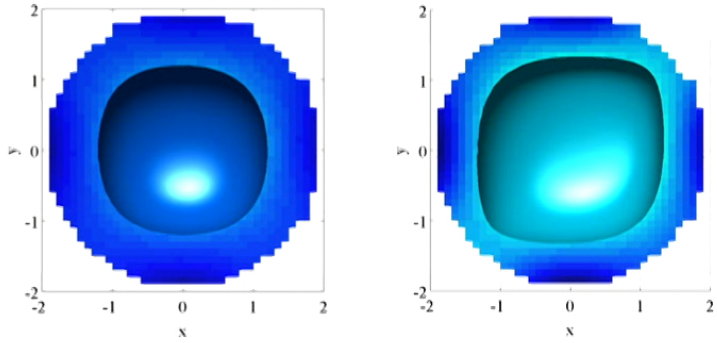
$$Er^{3+}:\ {}^4I_{15/2}$$

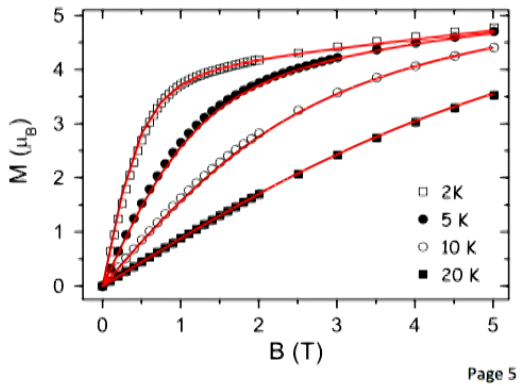
$$\mathcal{H} = B_0^4 C_0^4 + B_4^4 C_4^4 + B_0^6 C_0^6 + B_4^6 C_4^6$$

$$\mathcal{H} = B_0^2 C_0^2 + B_0^4 C_0^4 + B_3^4 (C_3^4 - C_{-3}^4) + B_0^6 C_0^6 + B_3^6 (C_3^6 - C_{-3}^6) + B_6^6 (C_6^6 + C_{-6}^6)$$

2 doublets + 3 quartets $\rightarrow$ 8 doublets

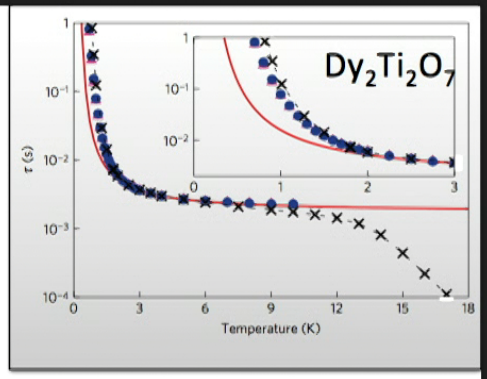
Γ_4 $(g_{\parallel} \neq 0, g_{\perp} \neq 0)$, Γ_{56} $(g_{\parallel} \neq 0, g_{\perp} = 0)$

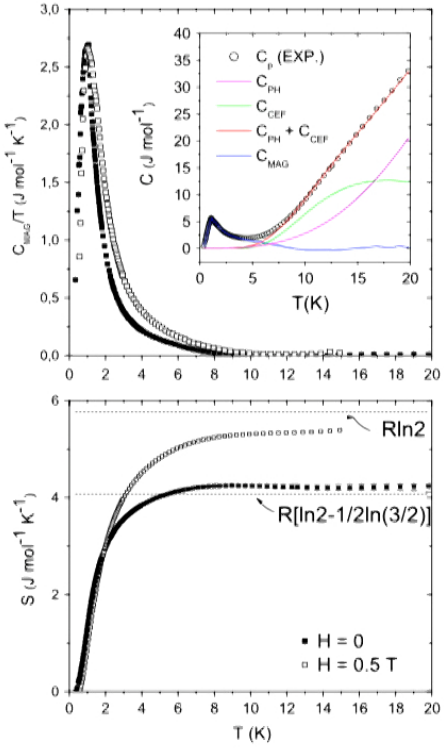


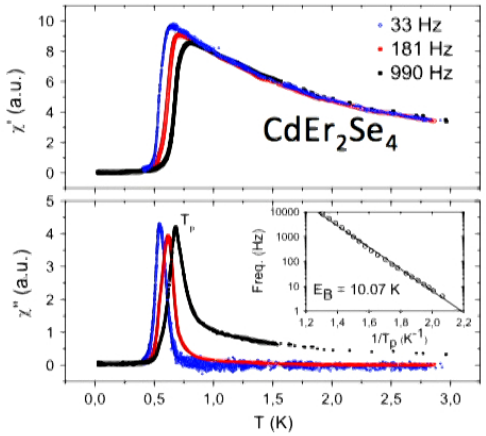


Lago *et al.*, PRL 2010; Malkin *et al.*, PRB (2004)

Spin ice in $CdEr_2Se_4$







- What is the nature of the spin ice state in $CdEr_2Se_4$? How about $CdEr_2S_4$?
- What are the origins and consequences of the much faster dynamics?

Lago et al., PRL (2010); Jaubert & Holdsworth N. Phys (2009); Snyder et al., PRB (2004)

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Sample

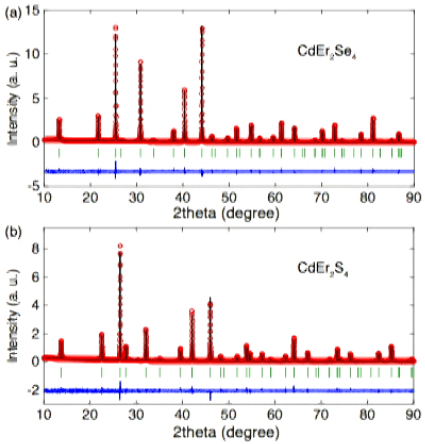
Big absorbers are very useful in neutron shielding design

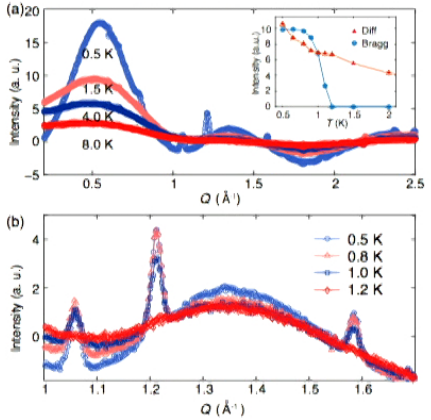
e.g. How much Cd is needed to absorb 99.9% of thermal neutrons?

$n_s = 0.046$ (barn.cm)$^{-1}$, $\sigma_{abs} = 2520$ b

$t = -\ln(T) / n_s\sigma_{abs} = -\ln(0.001) / 115.9 =$ 0.6 mm !

- But: σ_{abs} ^{113}Cd = 20600 b, σ_{abs} ^{114}Cd = 0.34 b





Stewart, Practical Neutron Scattering

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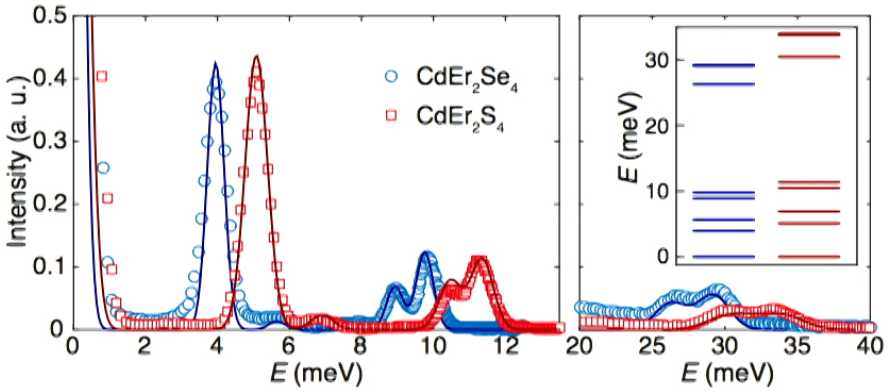
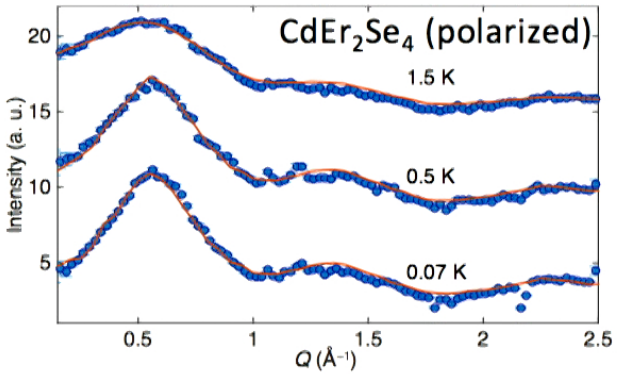
Crystal field parameters

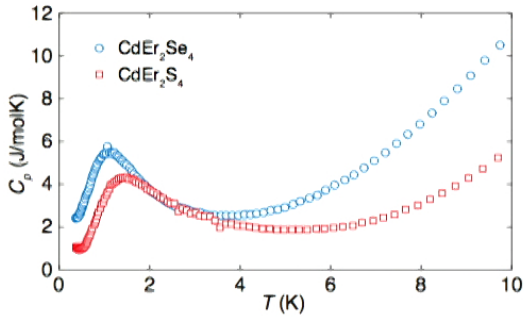


	B_2^0	B_4^0	B_4^3	B_6^0	B_6^3	B_6^6
$CdEr_2Se_4$	−25.70	−107.73	−97.74	25.31	−19.06	9.51
$CdEr_2S_4$	−29.18	−122.72	−113.66	25.97	−21.89	14.41
J_z	$\pm 15/2$	$\pm 9/2$	$\pm 3/2$	$\mp 3/2$	$\mp 9/2$	
$CdEr_2Se_4$	± 0.906	0.386	± 0.159	−0.073	± 0.004	
$CdEr_2S_4$	± 0.904	0.391	± 0.145	−0.094	± 0.006	

$$\mathcal{H} = B_0^2 C_0^2 + B_0^4 C_0^4 + B_3^4 (C_3^4 - C_{-3}^4) + B_0^6 C_0^6 + B_3^6 (C_3^6 - C_{-3}^6) + B_6^6 (C_6^6 + C_{-6}^6)$$

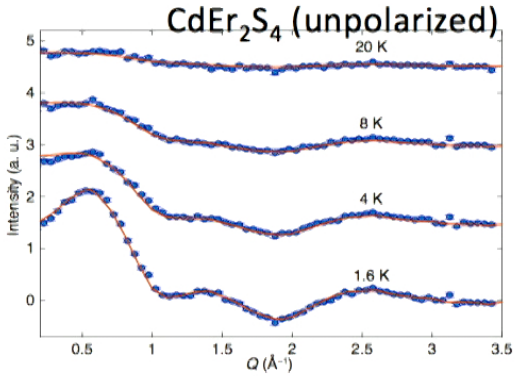
Diffuse neutron scattering and DSM (I)





$$\mathcal{H} = J_1 \sum_{\langle ij \rangle} \sigma_i \sigma_j + J_2 \sum_{\langle\langle ij \rangle\rangle} \sigma_i \sigma_j$$

$$+ D r_0^3 \sum_{ij} \left[\frac{\vec{n}_i \cdot \vec{n}_j}{|r_{ij}|^3} - \frac{3(\vec{n}_i \cdot \vec{r}_{ij})(\vec{n}_j \cdot \vec{r}_{ij})}{|r_{ij}|^5} \right] \sigma_i \sigma_j$$



$$D = \mu_0 (\langle J_z \rangle g \mu_B)^2 / (4\pi r_0^3) = 0.616 \text{ K}$$

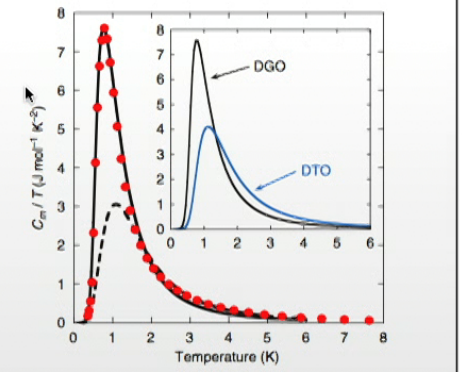
$$J_{\text{eff}} = J_1 + 5D/3 = 1 \text{ K}, \; J_1 = -0.027 \text{ K}$$

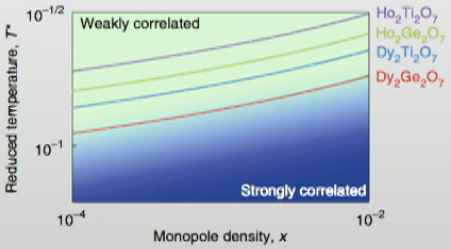
$$J_2 = 0.042 \text{ K}$$

Coulomb gas parameterization

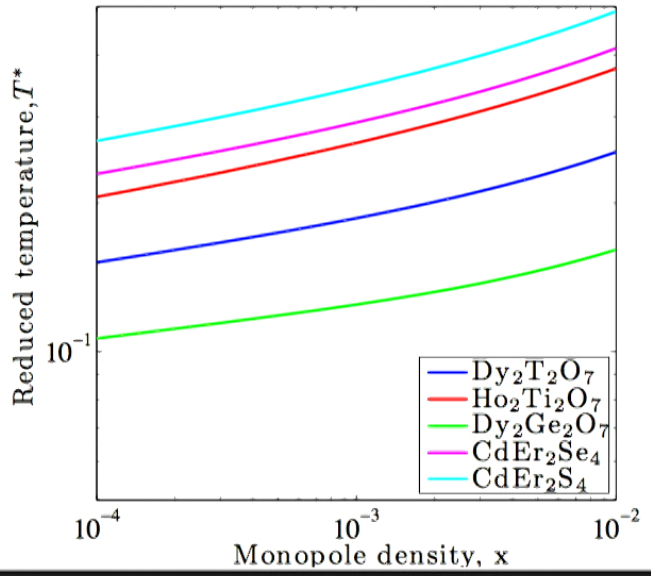
$$Q_m = 2\langle J_z \rangle g \mu_B \sqrt{3/2} r_0 \qquad v_0 = 2J_1 + (8/3)(1+\sqrt{2/3})D$$

	Q_m (μ_B/Å)	a (Å)	v_0 (K)
$CdEr_2Se_4$	3.28	5.03	2.93
$CdEr_2S_4$	3.42	4.83	3.84
$Dy_2Ti_2O_7$	4.53	4.38	4.35
$Dy_2Ge_2O_7$	4.6	4.3	3.35





Zhou *et al.*, Nat. Comm. (2011)



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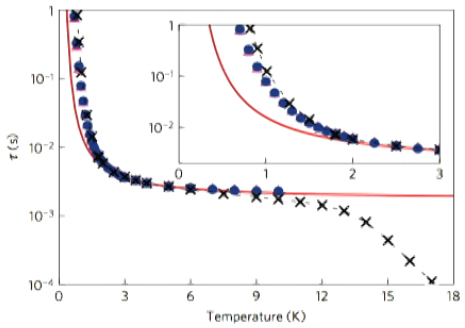
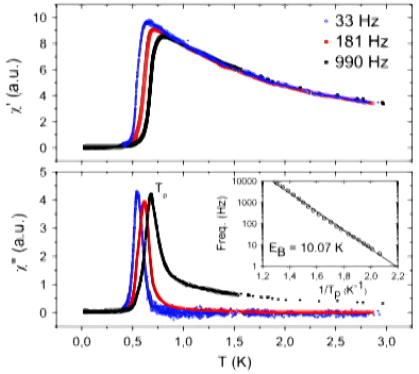
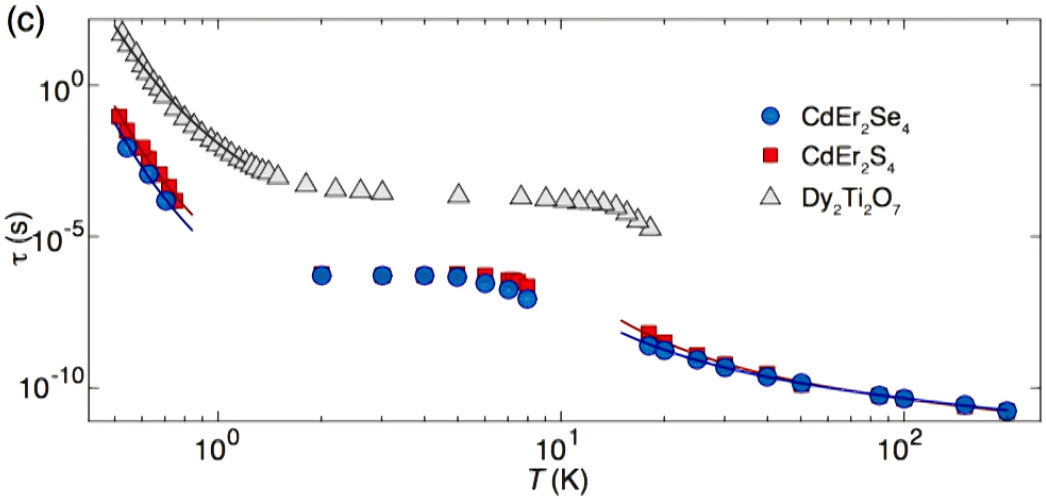
Overview of dynamics

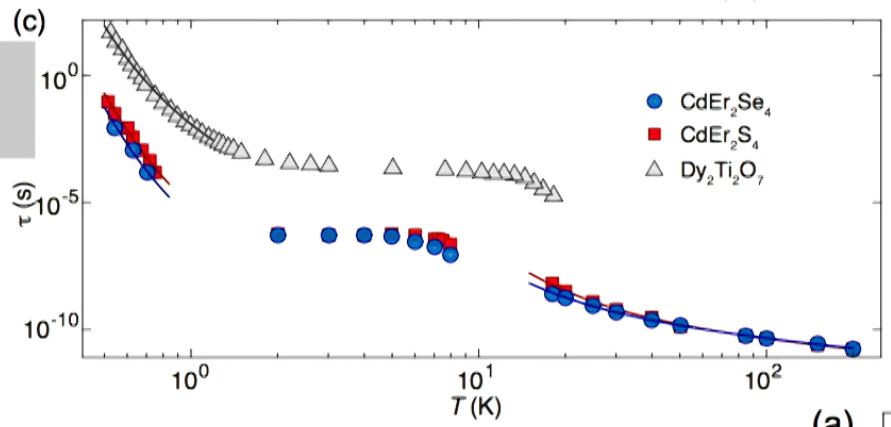




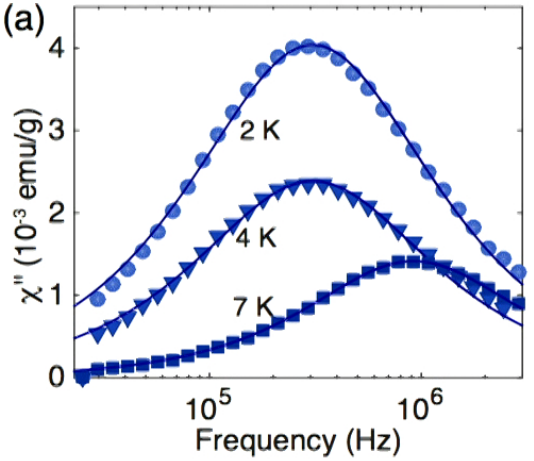


Lago et al., PRL (2010); Jaubert & Holdsworth N. Phys (2009); Snyder et al., PRB (2004)

Dynamical processes: moderate-low temperature



	τ_0 (s)	Δ (K)
$CdEr_2Se_4$	6.37×10^{-10}	10.07
$CdEr_2S_4$	1.84×10^{-9}	10.17
$Dy_2Ti_2O_7$	1.93×10^{-6}	9.93



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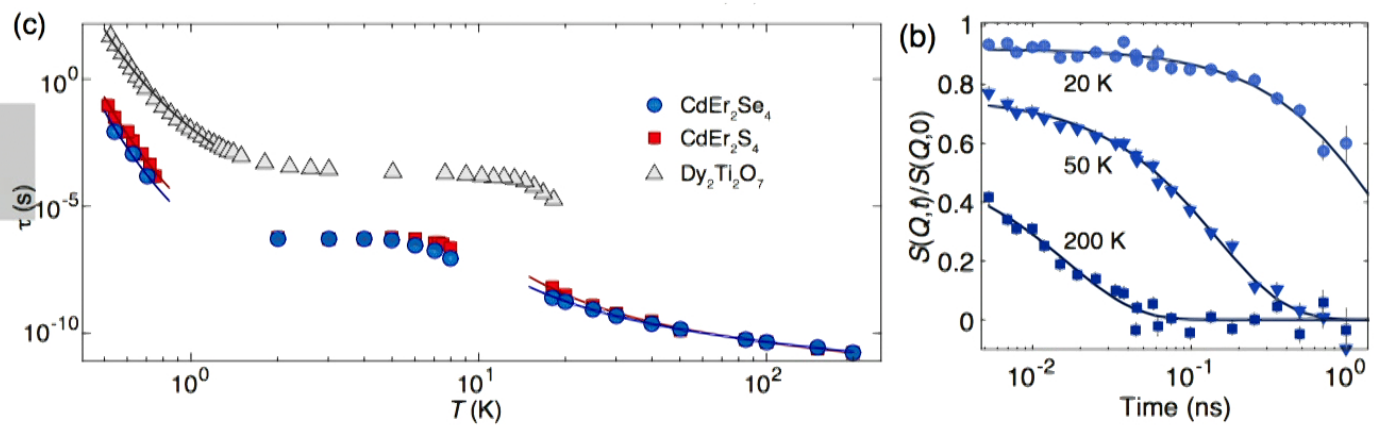
Dynamical processes: high temperature



$$\Gamma(T) = \sum_j B_j n_j$$

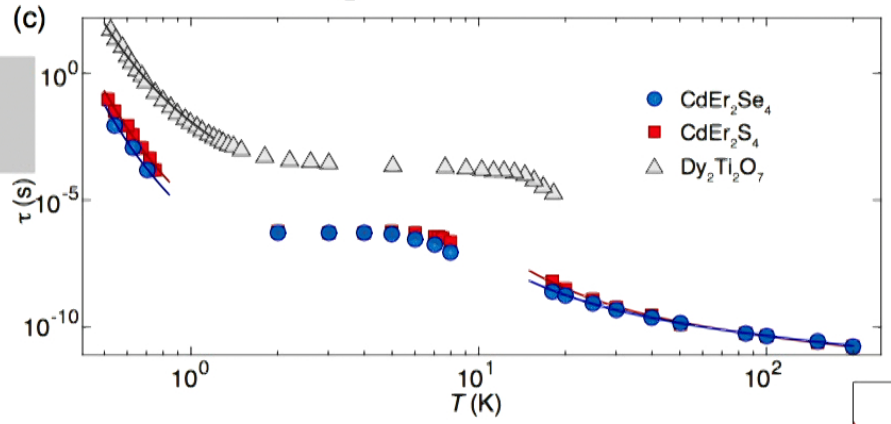
$$n = 1/(\exp(\Delta/k_B T) - 1)$$

$$B_j \propto \zeta_{\mu(u,v)} Z_{\mu(\Delta_j)}$$

$$\Delta_{\mathrm{Se}} = 75.3 \text{ K}; \ \Delta_{\mathrm{S}} = 96.3 \text{ K}$$

Ruminy, TF et al., PRB (2017)

Dynamical processes: moderate-low temperature



In Debye-Hückel theory $f \propto u\rho$

	τ_0 (s)	Δ (K)
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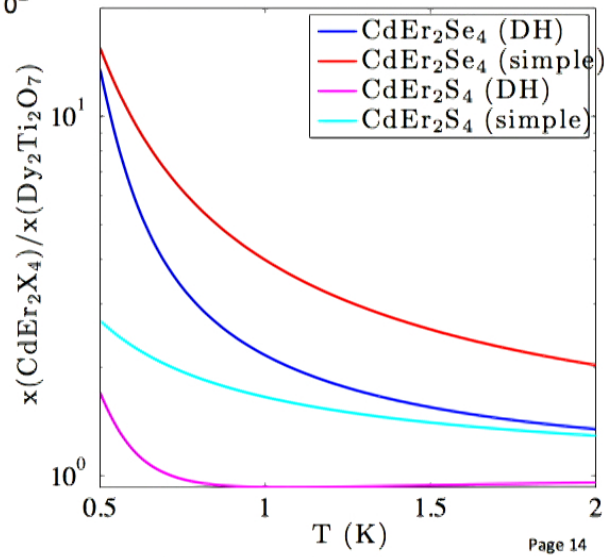
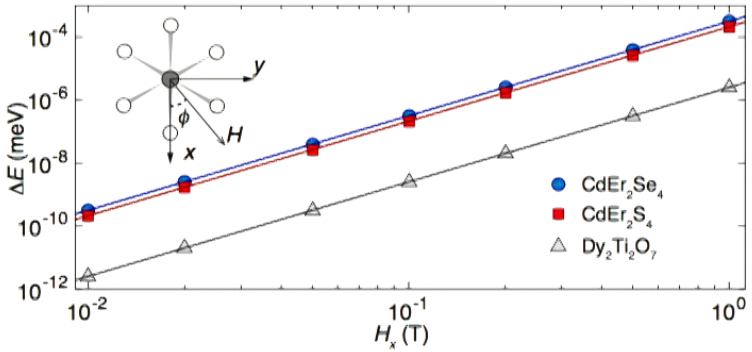


Origin of the fast dynamics

In Debye-Hückel theory $f \propto u\rho$



Tomasello et al., PRB (2015)

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Wir schaffen Wissen – heute für morgen

$CdEr_2X_4$ (X=Se,S) are dipolar spin ices with fast dynamics.

... The parameters of the dipolar spin ice states point to weakly correlated Coulomb gases of classical monopoles.

... The fast dynamics are due to a much greater monopole hopping rate.

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