# Dynamics of quasiparticle excitations in spin ice materials

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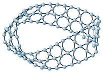


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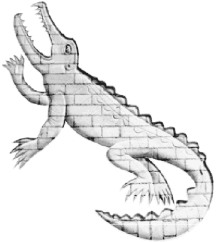

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International Conference on Highly Frustrated Magnetism
University of California Davis
Davis CA, USA

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# Dynamics in spin liquids

**e.g., Knolle and Moessner, arXiv:1804.02037**

- fundamental interest in response and relaxation properties, and motion of fractionalised excitations
- detection and diagnostics, e.g., the dynamical structure factor
- preparation of topological states
- operation on states $\rightarrow$ information processing and quantum computing
- connections with kinetically constrained and glassy systems

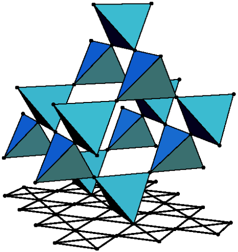
origin of the dynamics: thermal vs quantum fluctuations, interactions, crystal symmetries and environment

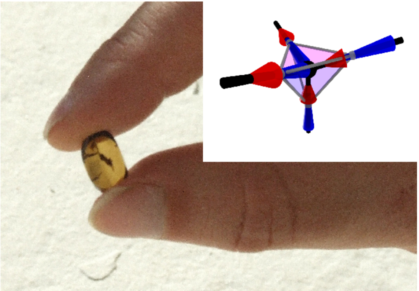
# Outline: Spin correlation dependent local dynamics

- a case in point: spin ice materials
  - dynamics due to internal fields
  - interactions + lattice geometry → strength of kinetic terms depends on local correlations
  - separation of effective 'time scales'
- new paradigm for stochastic dynamics in classical spin ice (?)
- quantum spin ice: fractal wave functions, (near) localisation, and 'monopole conductivity'
- conclusions

# Spin Ice ($RE_2TM_2O_7$)

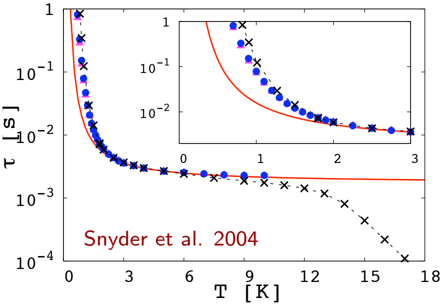
- large spins (Dy: 15/2; Ho: 8)
(small exchange $\sim 1$ K)
- local [111] crystal field $\gtrsim 100$ K
$\Rightarrow$ large gap above GS doublet
- large magnetic moment $3 - 10\,\mu_B$
$\Rightarrow$ long range dipolar interactions



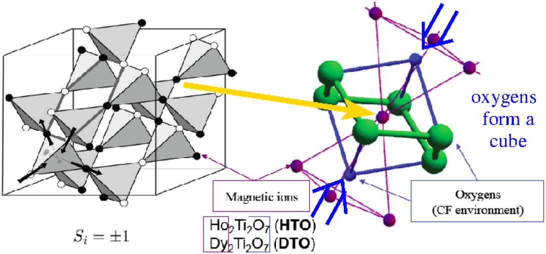


- extensively degenerate GS
- emergent Gauge symmetry
- elementary excitations fractionalise dipoles into monopoles
$+$ long-range Coulomb interactions

# Spin dynamics

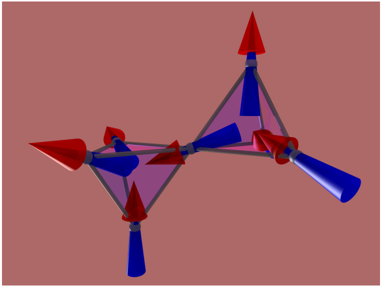



AC susceptibility suggests quantum mechanical magnetic relaxation at low $T$



# Quantum fluctuations from 'transverse' interactions

- CEF anisotropy projects spins along local [111] axis ('z')
- dipolar and exchange interactions act with longitudinal and transverse components



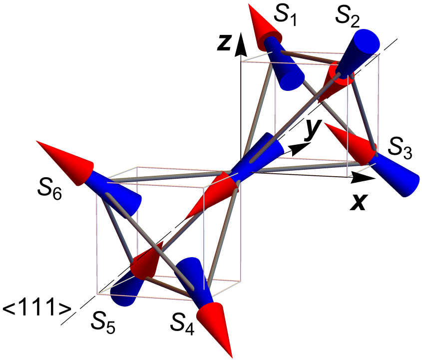
$$H = H_{\mathrm{exch}} + D r_{\mathrm{nn}}^3 \sum_{i,j} \left[ \frac{\vec{S}_i \cdot \vec{S}_j}{|\vec{r}_{ij}|^3} - \frac{3\left(\vec{S}_i \cdot \vec{r}_{ij}\right)\left(\vec{S}_j \cdot \vec{r}_{ij}\right)}{|\vec{r}_{ij}|^5} \right]$$

lowest energy state: two-in, two-out configuration on each tetrahedron

# Toy model: nearest-neighbour symmetric exchange

$$H_{\mathrm{nn}} = -J \sum_{\langle i,j \rangle} \vec{S}_i \cdot \vec{S}_j = -J \sum_i \vec{S}_i \cdot \vec{h}_{\mathrm{nn}}^{(i)}$$

$$\vec{h}_{\mathrm{nn}}^{(i)} = -J \left[ \left( \vec{S}_1 + \vec{S}_4 \right) + \left( \vec{S}_2 + \vec{S}_5 \right) + \left( \vec{S}_3 + \vec{S}_6 \right) \right]$$



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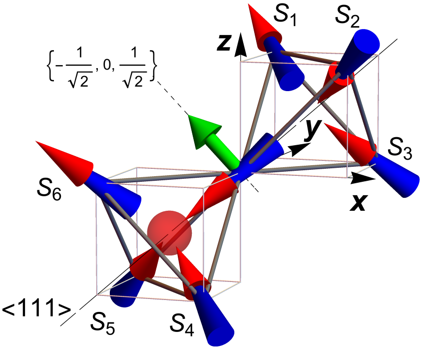

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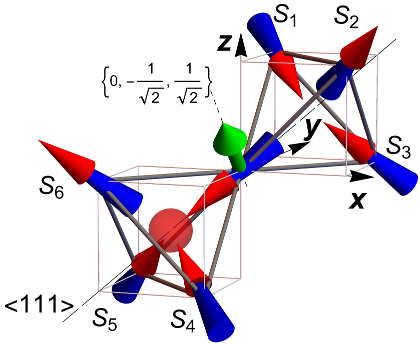

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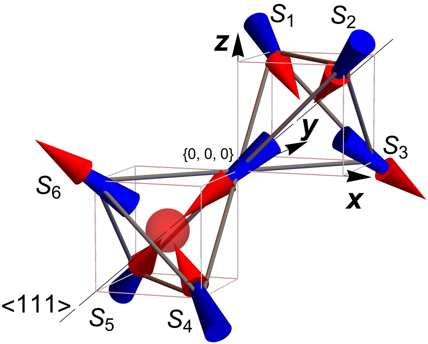
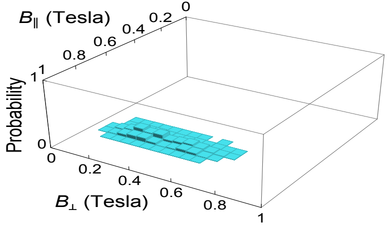


# Dipolar fields

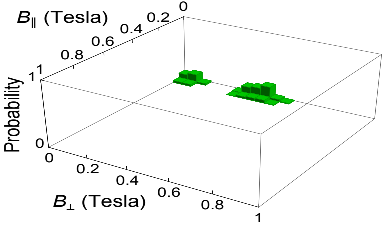
**Tomasello, et al. PRB '15 and in preparation**

internal dipolar fields at spin sites (DTO parameters; similarly for HTO)

**away from monopoles**




**next to a monopole**




- qualitatively similar to the toy model
- same 2:1 ratio (small but non-vanishing transverse term)

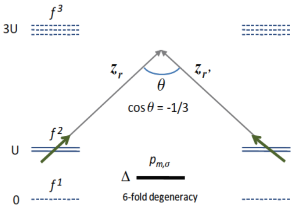
# Realistic exchange

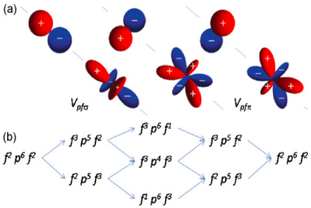
Tomasello, et al. in preparation

many electron extension of the calculation by Onoda and Tanaka (PRB 2011)

$$\hat{H}_{ff} = \frac{2}{(nU-\Delta)^2} \sum_{\langle \mathbf{r},\mathbf{r}' \rangle} \sum_{\substack{m_1,m_2,m_1',m_2'=0,\pm 1 \\ \sigma_1,\sigma_2,\sigma_1',\sigma_2'=\pm}} V_{m_1} V_{m_1'} V_{m_2} V_{m_2'} \times \hat{f}^{\dagger}_{\mathbf{r},m_1,\sigma_1} \hat{f}_{\mathbf{r},m_2,\sigma_2} \hat{f}^{\dagger}_{\mathbf{r}',m_1',\sigma_1'} \hat{f}_{\mathbf{r}',m_2',\sigma_2'}$$

$$\times \left[ -\frac{1}{nU-\Delta} \delta_{\substack{m_1,m_2 \\ \sigma_1,\sigma_2}} \delta_{\substack{m_1',m_2' \\ \sigma_1',\sigma_2'}} + \left( \frac{1}{nU-\Delta} + \frac{1}{U} \right) (\mathcal{R}^{\dagger}_{\mathbf{r}} \mathcal{R}_{\mathbf{r}'})_{\substack{m_1,m_2' \\ \sigma_1,\sigma_2'}} (\mathcal{R}^{\dagger}_{\mathbf{r}'} \mathcal{R}_{\mathbf{r}})_{\substack{m_1',m_2 \\ \sigma_1',\sigma_2}} \right],$$

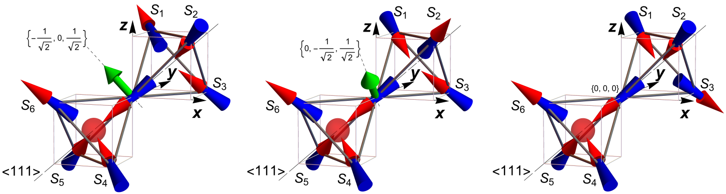


# Realistic exchange

**Tomasello, et al. in preparation**

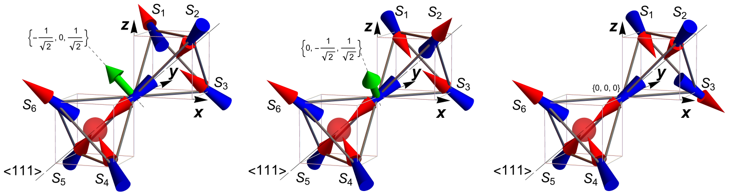
- from electron interactions $H_{ff}$ to $J$, $M_J$ total angular momentum basis
- assemble the exchange Hamiltonian operator $H^{(i)}_{\mathrm{exch}} = \sum_{j=1}^{6} H_{\mathrm{exch}}(i,j)$
- project onto the 'ground states' of the neighbouring spins $|S_j\rangle$: $\overline{H}^{(i)}_{\mathrm{exch}} = \sum_{j=1}^{6} \langle S_j | H_{\mathrm{exch}}(i,j) | S_j \rangle$



# Realistic exchange

Tomasello, et al. in preparation

- $\overline{H}^{(i)}_{\text{exch}}$ (now a single ion operator) has no transverse terms by symmetry in 1 out of 3 cases (as before)
- equivalently, $\overline{H}^{(i)}_{\text{exch}}$ splits the single ion GS CEF doublet only in 2 of the three surrounding spin configurations
- same 2:1 dependence on local spin correlations as for toy exchange and dipolar interactions



# Sanity check: time scales

Tomasello, et al. PRB '15 and in preparation

adding all terms together acting on the central spin $i$: CEF + dipolar + exchange $\rightarrow$ precession dynamics:

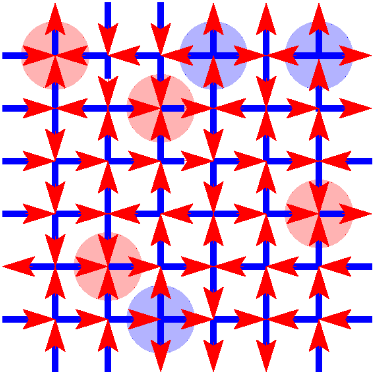
- time scales for DTO and HTO (classical spin ices):
  $\tau^{\text{fast}} \sim 1\ \mu$s vs. $\tau^{\text{slow}} \sim 1$ ms
- time scales for PSO and PZO (quantum spin ices):
  $\tau^{\text{fast}} \sim 10$ ps vs. $\tau^{\text{slow}} \sim 0.1$ ms

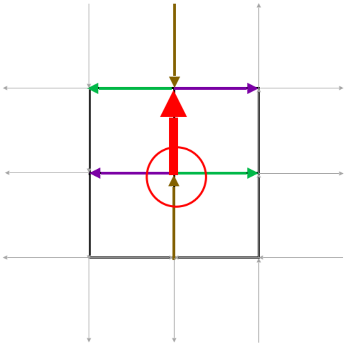
• new paradigm for stochastic dynamics in classical spin ice: two time scales dependent on local spin correlations

• extreme separation of time scales (equiv.: transverse fields / tunnelling amplitudes) in quantum spin ice candidates, with possible signatures in dynamical response functions

# Quantum dynamics of square ice

Gopalakrishnan et al. (in prep.)





(slow)

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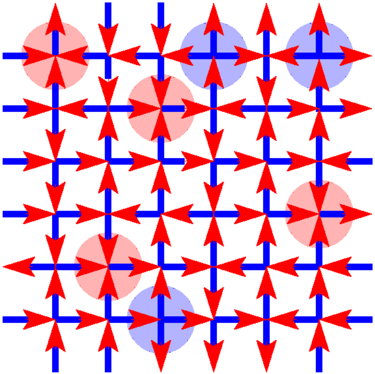

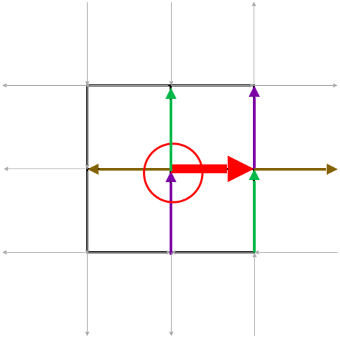

(fast)

# Quantum dynamics of square ice

Gopalakrishnan et al. (in prep.)

- low monopole density limit (isolated monopole motion)
- ignore ring exchange: the underlying spin configuration is static unless traversed by a monopole

  $\rightarrow$ stochastic ensemble of paths where monopoles hop with amplitudes $t_{\mathrm{slow}}$ and $t_{\mathrm{fast}}$
- quantum spin ice: $t_{\mathrm{slow}} \ll t_{\mathrm{fast}}$ (Born-Oppenheimer approx.)
- (possible comparison to quantum percolation on trees)

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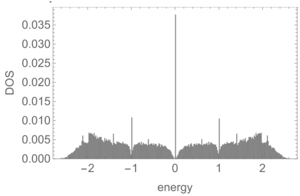
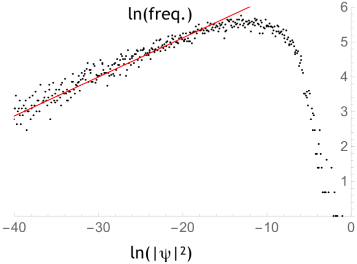
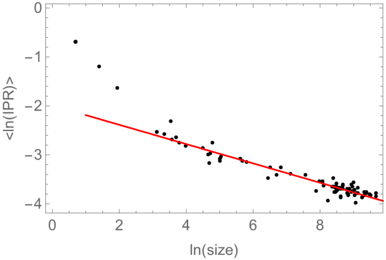



- sharp peaks in dos due to finite size clusters
- fractal, non-ergodic behaviour of wavefunctions
- ‘monopole localisation’



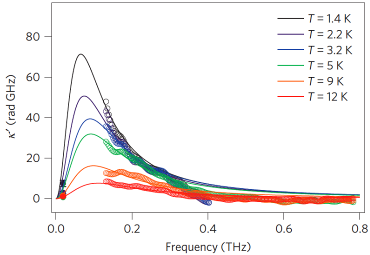




$P(|\psi|) \sim |\psi|^{-4/5}$ Inv. Particip. Ratio $\sim N^{-1/5}$

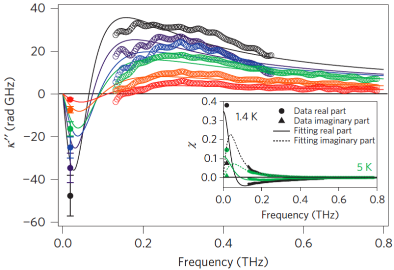
# Quantum dynamics of square ice

Gopalakrishnan et al. (in prep.)

- monopole hopping ⇔ spin flip ⇔ magnetisation dynamics
- 'monopole conductivity' can be probed via (ultrafast) magnetisation measurements (e.g., Pan et al. 2015 on $Yb_2Ti_2O_7$, Armitage 2018)




real (dissipative) part

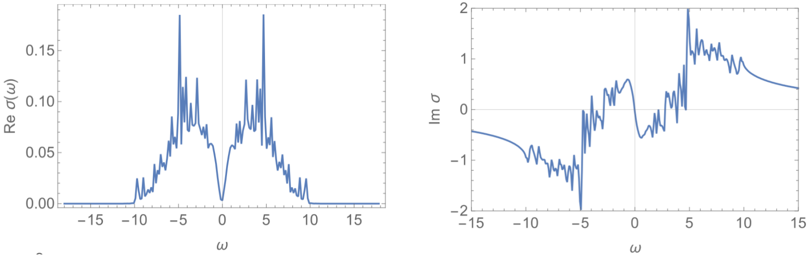



imaginary (reactive) part

# Quantum dynamics of square ice

**Gopalakrishnan et al. (in prep.)**

- compute current matrix elements in *real* space
- obtain real space conductivity (monopole position $\leftrightarrow$ 'dipole moment')




low frequency structure is a diagnostic of (near) localisation
(increasing $t_{\rm slow}/t_{\rm fast}$ ratio $\to$ ergodic wavefunctions)

# Conclusions

microscopic details of magnetic degrees of freedom in spin liquids can have important consequences on their dynamics

a case in point: spin ice materials

- dynamics due to internal fields strongly depends on local spin correlations $\rightarrow$ bimodal distribution of tunnelling amplitudes
- new paradigm for stochastic dynamics in classical spin ice?
- fractal wavefunctions and (near) localisation of monopoles in quantum spin ice
- monopole conductivity can be directly probed in (ultrafast) magnetisation dynamics experiments

# Quantum dynamics of square ice

Gopalakrishnan et al. (in prep.)