Decoherence in Quantum Magnets: Theory and Experiment on τ_ϕ

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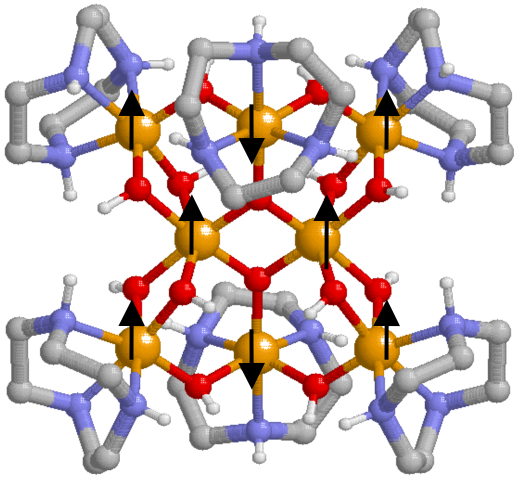
Individual properties of molecular magnets are controlled by chemistry rather than by nanoengineering and are highly tunable. This makes them very good candidates for solid-state qubits. However their advantages cannot be exploited until decoherence is understood and suppressed. Here we study decoherence in crystals of Fe_8 molecules.

Each molecule behaves as a Giant Spin **S=10** (T < 10 K):

$$H_S^{(Fe)} = -DS_z^2 + ES_x^2 + K_4^{\perp}(S_+^4 + S_-^4) - g_e\mu_B\vec{H}\vec{S}$$

Fe_8:S=10

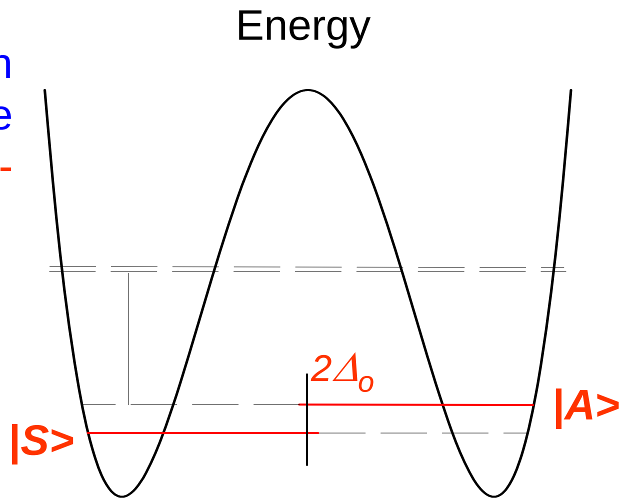


At low-T, only two state of each molecule are occupied: the molecules then behave as two-level systems, i.e. as QUBITs.

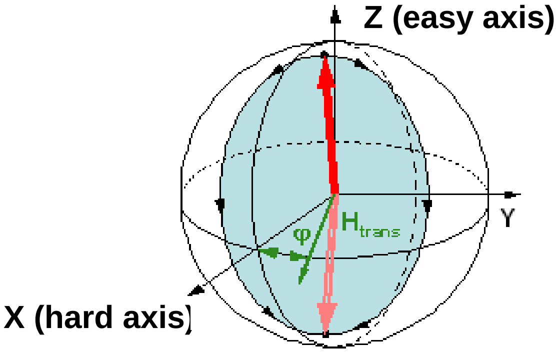
$$H_{TLS} = -\Delta_o\hat{\tau}^x - \xi\hat{\tau}^z$$

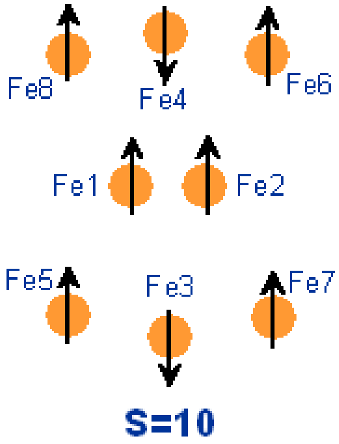
$2\Delta_o$ - tunneling gap, ξ - bias

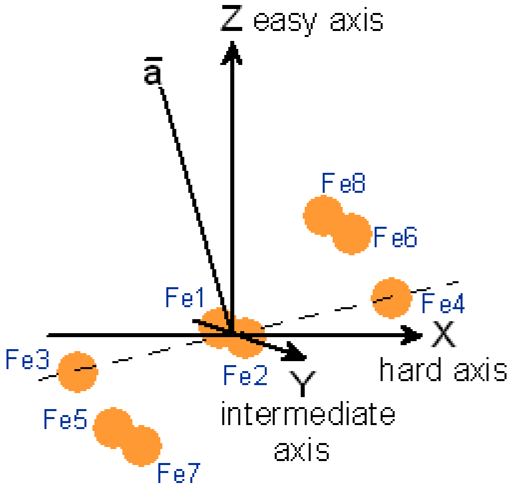
|S> and |A> - lowest symmetric and antisymmetric states of H_S



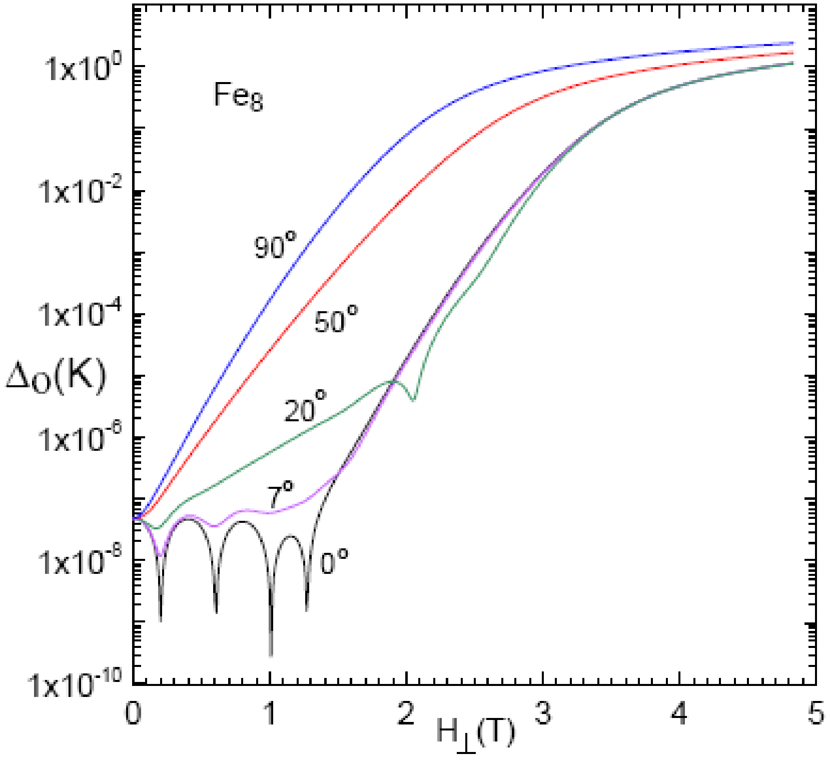
Parameters of QUBIT can be controlled by applying transverse (to the molecular easy axis) magnetic field $H_{\perp}$.



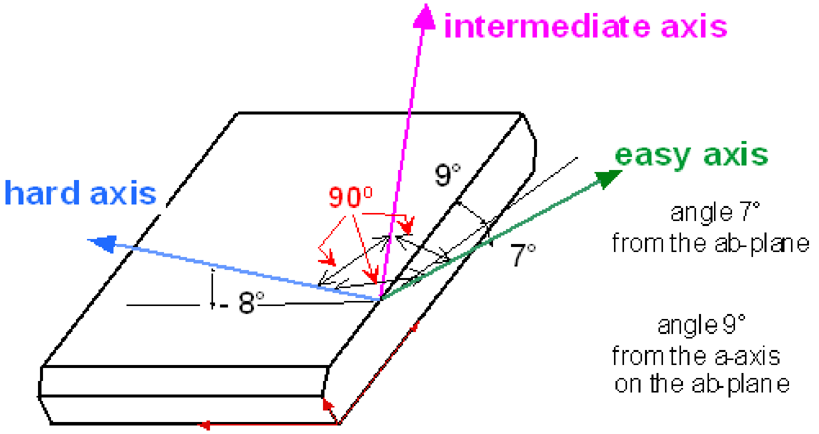


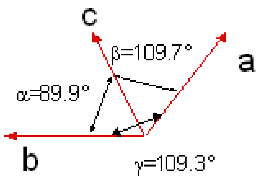


Tunneling matrix element in Fe_8 molecule



A typical Fe-8 sample





a = 10.5 Å
b = 14.1 Å
c = 15.0 Å

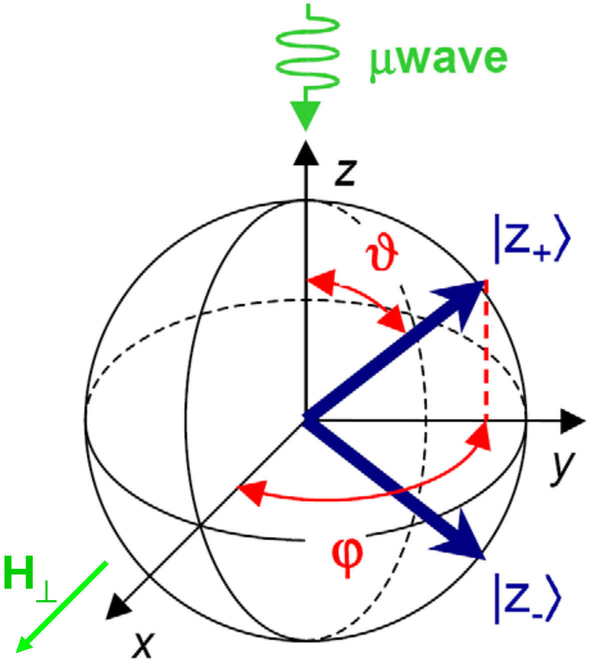
triclinic lattice

A. Barra et al, Chem. Eur. J. 6, 1608, (2000)

Experimental approach to measure decoherence time τ_ϕ in crystals of magnetic molecules

By applying transverse magnetic field one creates symmetric |S> and antisymmetric |A> states separated by the gap $2\Delta_o(H_\perp)$. By applying then micro-wave pulse one can mix up |S> and |A> states and create the one-well states $|Z_\pm> = (|S> \pm |A>)/2^{1/2}$, initiating oscillations between them. How long oscillations can last? ⇒ T_2 (τ_ϕ)

S. Takahashi, J. van Tol, C.C. Beedle, D.N. Hendrickson, PRL 102, (2009)



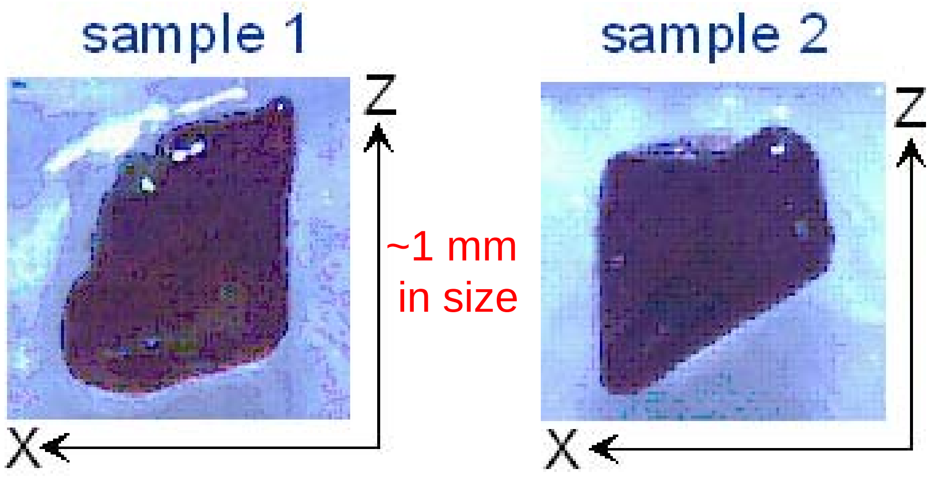
240 GHz pulsed EPR spectrometer at the National High Magnetic Laboratory in Tallahassee:

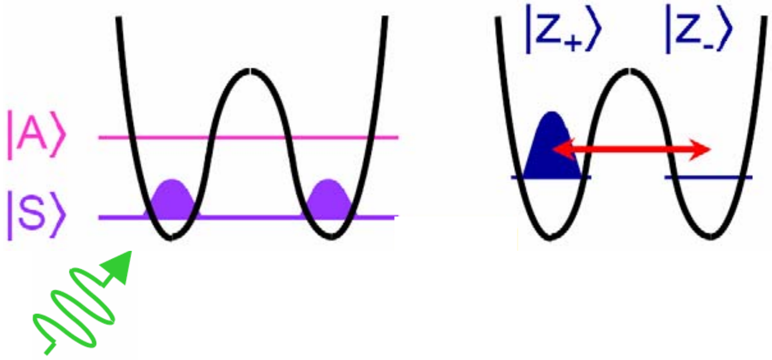
Hahn echo sequence: $\pi/2-\tau-\pi-\tau-$echo

Field $H_\perp$ along the hard axis X (10.5-12 T)
and intermediate axis Y (9.5 -11 T)

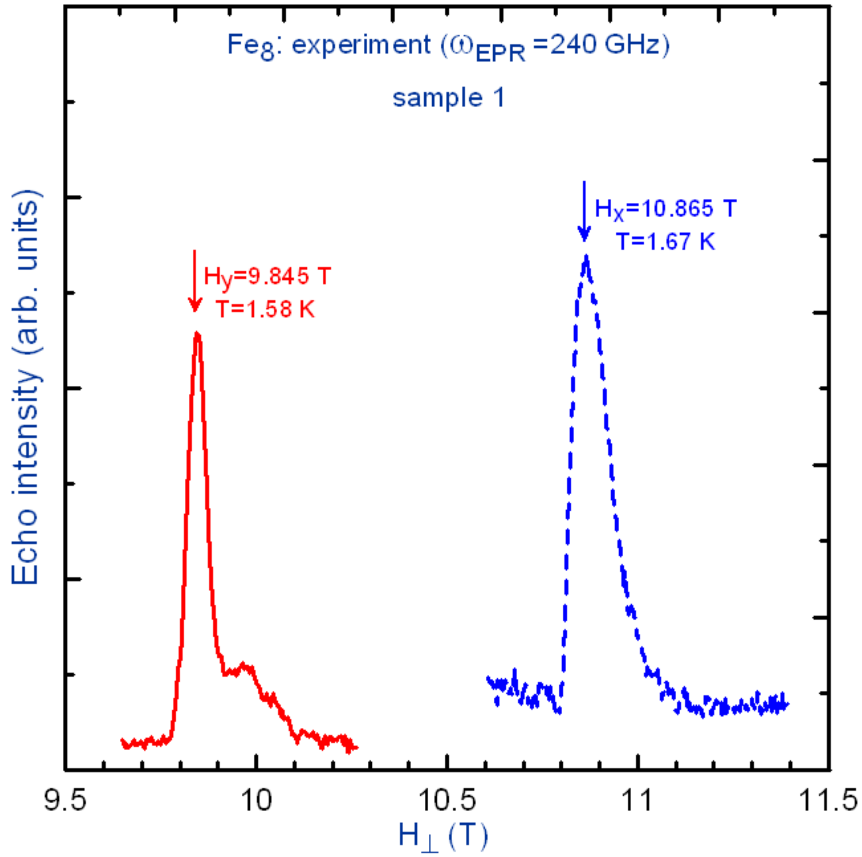
<u>Single exponential fit of data to obtain T_2</u>

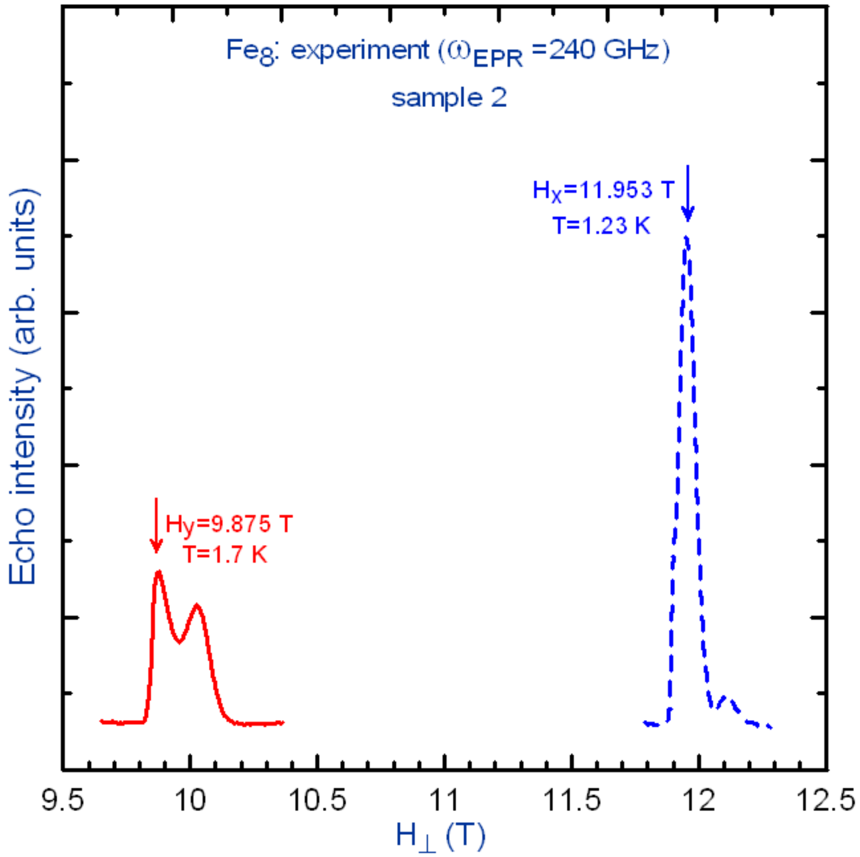
$1.2\ K < T < 2\ K$





Echo experiments in two samples:





The accuracy of detection of the anisotropy axes directions is ~15^o which allows to explain positions of the first three peaks. To explain the position of the last peak (H_x=11.953 T) one needs to assume either the existence of higher order anisotropy terms or the wider distribution of the anisotropy parameter D (D-strains; $\Delta D/D$ ~0.01: *K. Park, M.A. Novotny, N.S. Dalal, S. Hill, and P.A. Rikvold, Phys. Rev. B 65, 014426 (2002)*).

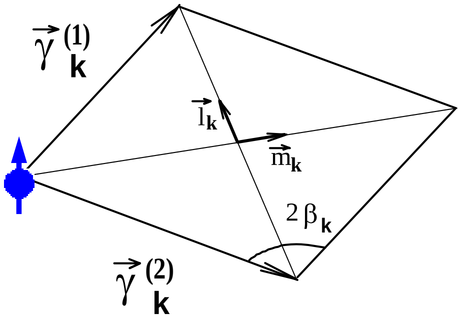
However, the exact positions of peaks are unimportant – they correspond to ω=240 GHz.

Theoretical approach to calculate decoherence time τ_ϕ in crystals of magnetic molecules

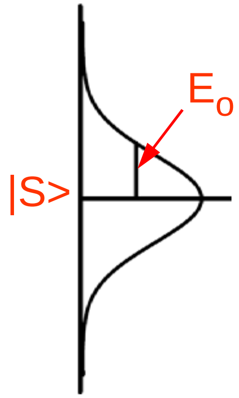
P.C.E. Stamp and I.S. Tupitsyn, PRB 69 (2004)

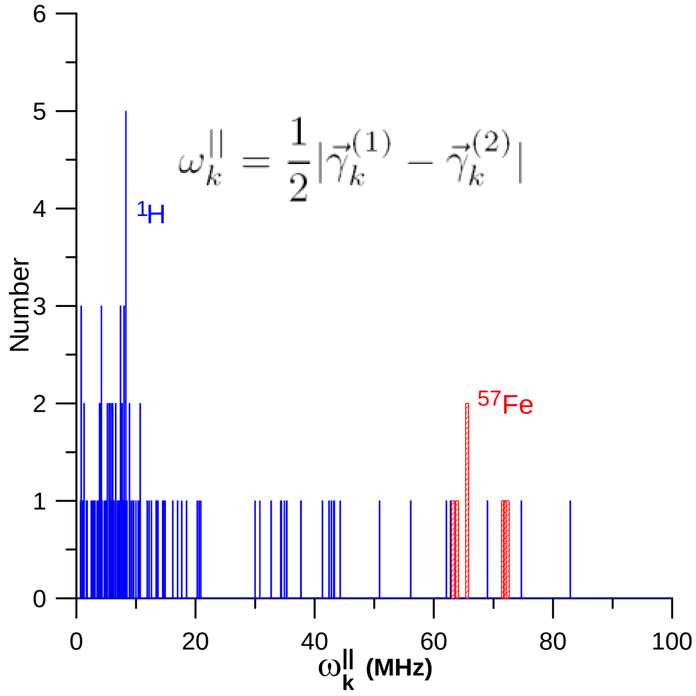
$$H_{env} = H_{nuc} + H_{sp\text{-}ph} + H_{exch} + H_{d\text{-}d}$$

Nuclear spin-bath:

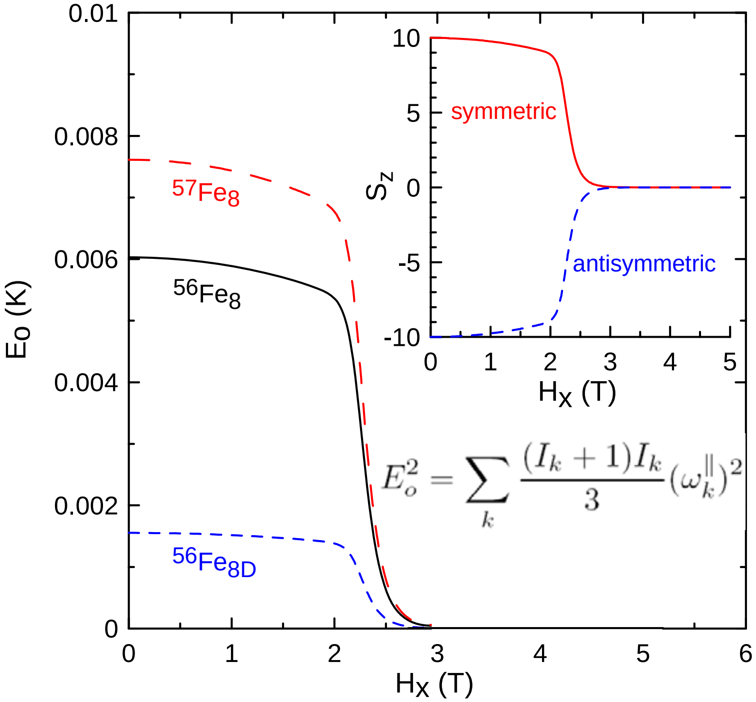


$$H_{\text{nuc}} = \sum_{k=1}^{N} \vec{\gamma}_k \cdot \mathbf{I}_k$$





$$\omega_k^{||} = \frac{1}{2}|\vec{\gamma}_k^{(1)} - \vec{\gamma}_k^{(2)}|$$



$$E_o^2 = \sum_k \frac{(I_k+1)I_k}{3}(\omega_k^{||})^2$$

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P.C.E. Stamp and I.S. Tupitsyn, PRB 69 (2004)

$H_{env} = H_{nuc} + H_{sp\text{-}ph} + H_{exch} + H_{d\text{-}d}$

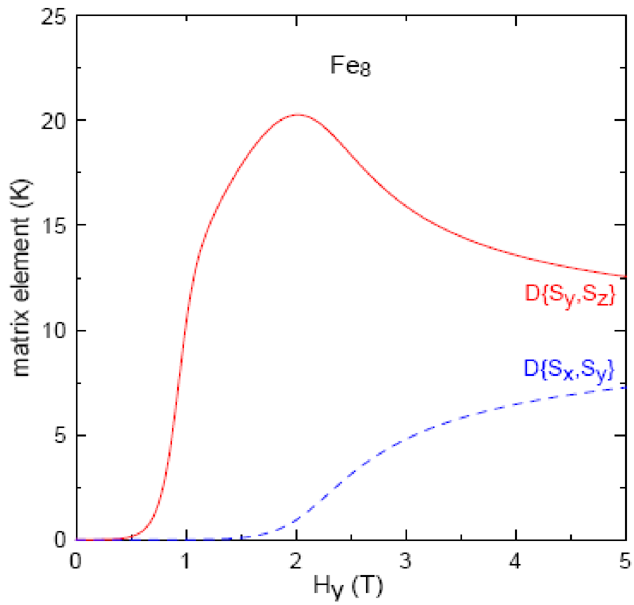
Phonons:

$$H_{\rm sp-ph} = \sum_t \eta_t \hat{O}_t^P \hat{O}_t^S$$

Symmetries: Triclinic & D_2

$\Downarrow$

$(\eta_1^{Fe}\epsilon_{yz} + \eta_2^{Fe}\omega_{yz})(S^y S^z + S^z S^y)$, etc

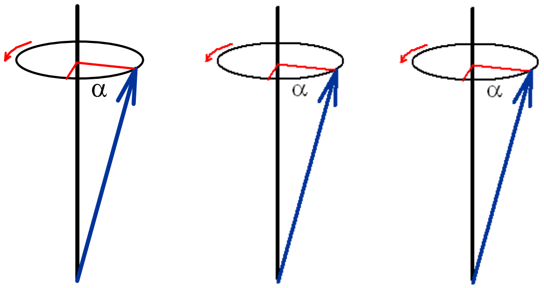


Theoretical approach to calculate decoherence time τ_ϕ in crystals of magnetic molecules

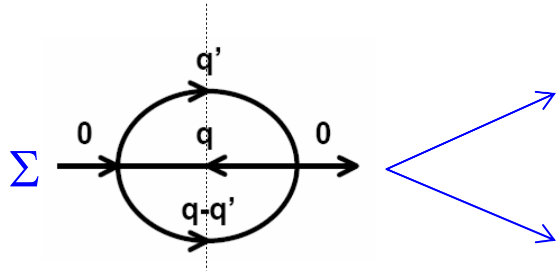
A. Morello, P.C.E. Stamp and I.S. Tupitsyn, PRL 97, (2006)

$$H_{env} = H_{nuc} + H_{sp\text{-}ph} + H_{exch} + H_{d\text{-}d}$$

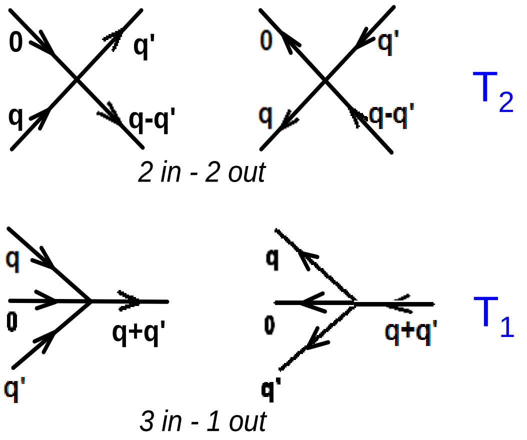
Magnons:



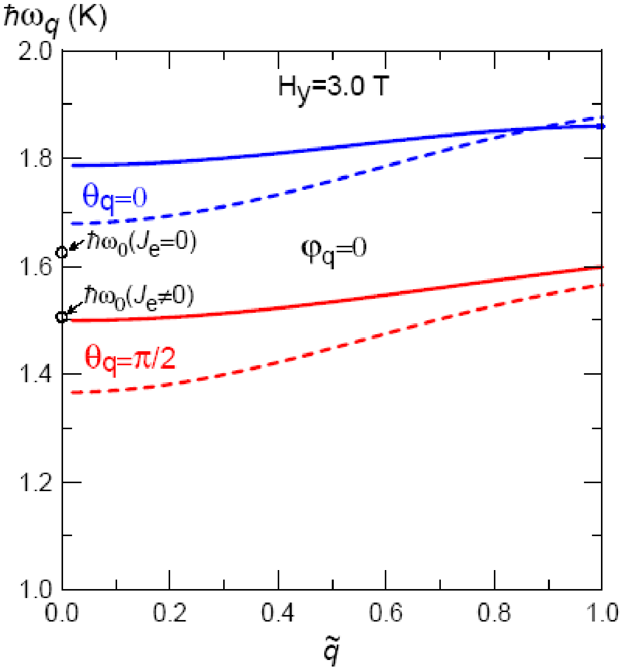
Uniform precession --> q=0 magnon



$$1/\tau_\phi = -\mathrm{Im}\Sigma$$



In a transverse magnetic field the oscillations between two states are equivalent to a uniform spin precession along the field directions, i.e., to a *q=0* magnon. Scattering of the *q=0* mode off thermal magnons leads to a decay of oscillations.



The spectrum is gapped, $\Delta_o/U_d >> 1$, and the only allowed leading process is *2 in – 2 out*. This process contributes to T_2.

Theoretical approach to calculate decoherence time τ_ϕ in crystals of magnetic molecules

A. Morello, P.C.E. Stamp and I.S. Tupitsyn, PRL 97, (2006)

Dimensionless decoherence rate: $\gamma_\phi = \hbar/(\Delta_o\tau_\phi)$ ~ Q^{-1}

Nuclear:

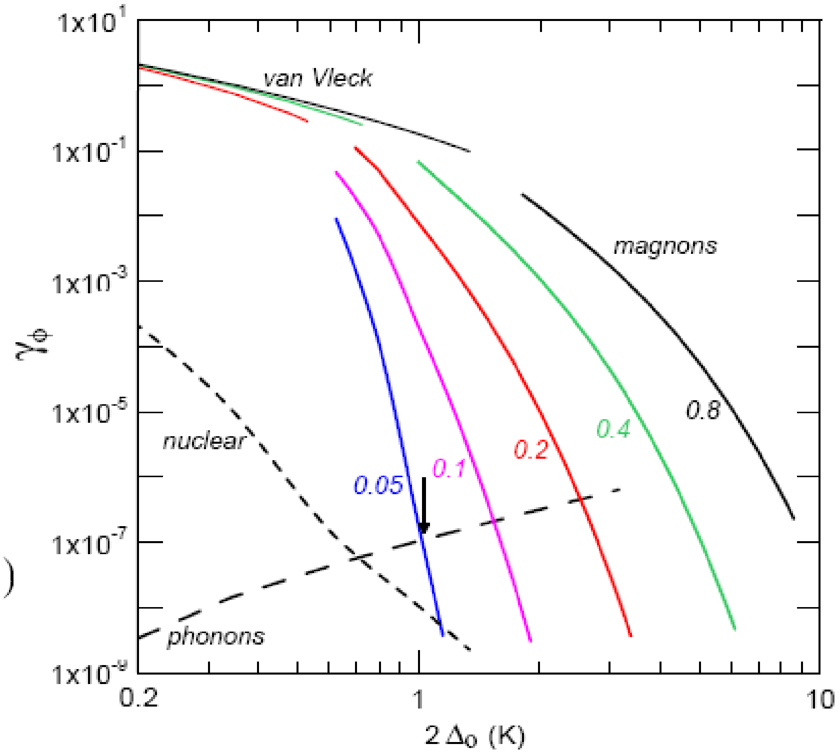
$$\gamma_\phi^{NS} = E_0^2/2\Delta_0^2$$

Phonon:

$$\gamma_\phi^{ph} = \frac{\mathcal{M}^2_{\mathcal{AS}}\Delta_0^2}{\pi\rho c_s^5\hbar^3}\coth\left(\frac{\Delta_0}{k_BT}\right)$$

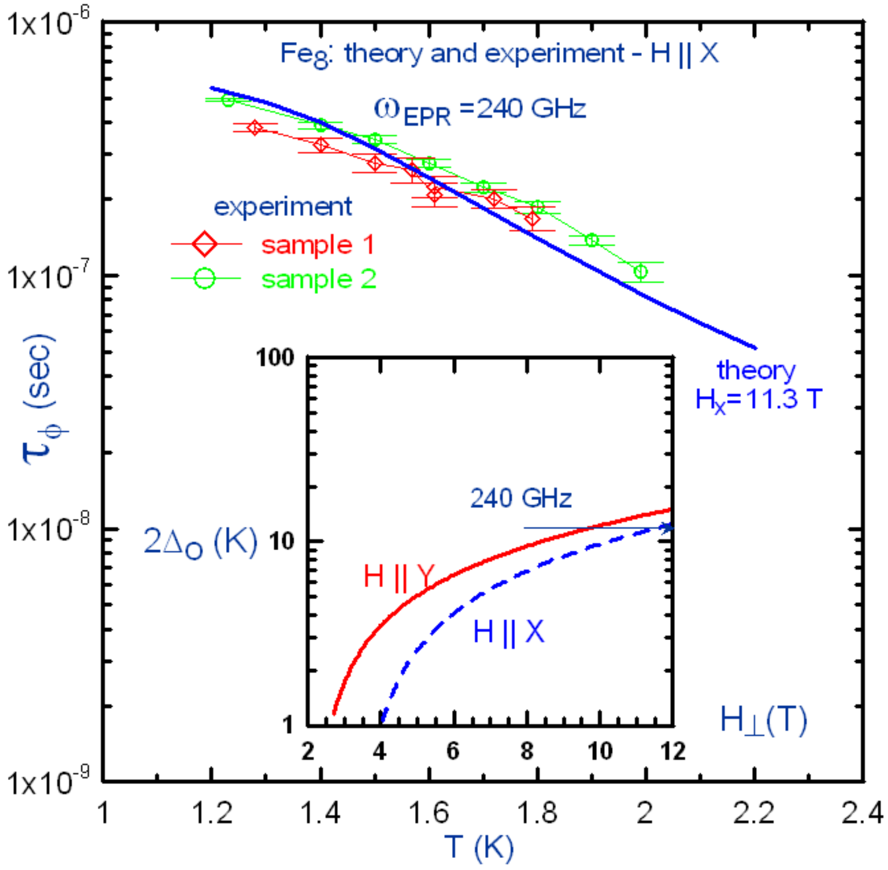
Magnon:

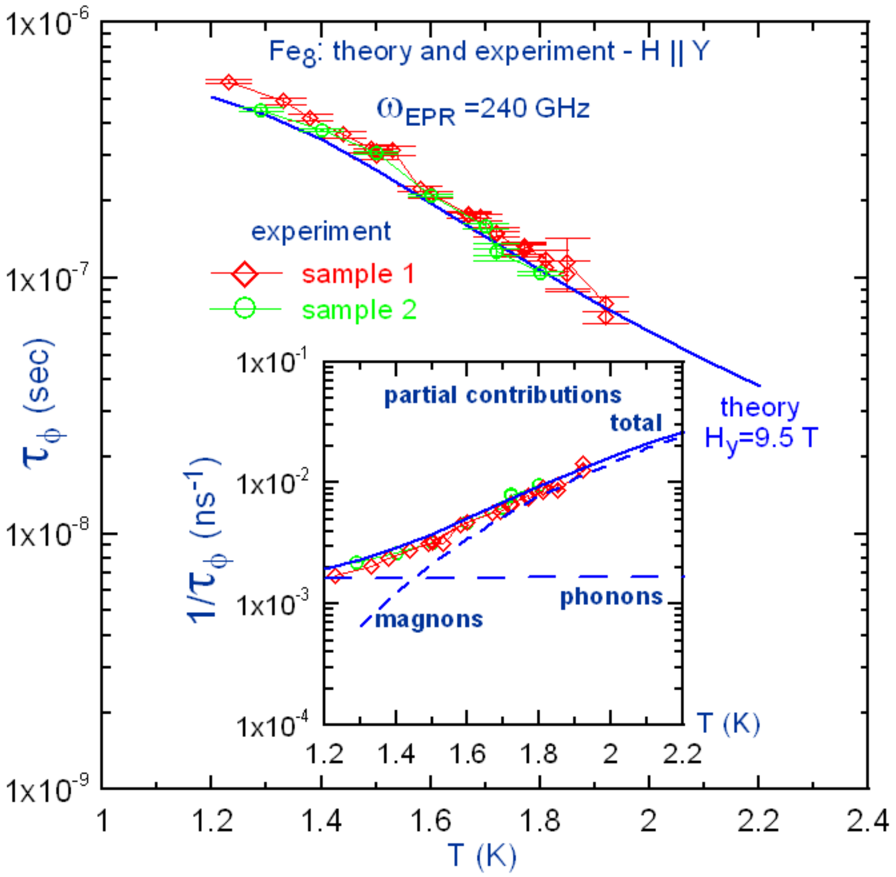
$$\gamma_\phi^{m} = \frac{2\pi}{\hbar\Delta_0}\sum_{\mathbf{qq'}}|\Gamma^{(4)}_{\mathbf{qq'}}|^2\mathcal{F}[\bar{n}_\mathbf{q}]\delta(\omega_0+\omega_\mathbf{q}-\omega_\mathbf{q'}-\omega_\mathbf{q-q'})$$



EPR frequency 240 GHz corresponds to ≈11.5 K. Theoretically the tunneling gap $2\Delta_o(H\perp) \approx$ 11.5 K in Fe_8 opens up at fields $H_x \approx$ 11.3 T and $H_y \approx$ 9.5 T. The contribution from the nuclear-spin bath in Fe_8 decays rapidly with the external transverse field. At given experimental fields the dominant contributions come from phonons and magnons.

Decoherence time τ_ϕ: Comparison of theory and experiment





In the high-field limit at higher temperatures the pair-wise (magnon) decoherence dominates over the phonon (as well as nuclear) one while at lower temperatures the phonon mechanism starts to play a leading role.

To our knowledge this is the first time when experimental and theoretical τ_ϕ in magnetic insulators are in agreement with each other.