PCE STAMP: LECTURE 2

A CASE STUDY
in
QUANTUM RELAXATION, COHERENCE, & DECOHERENCE

In this lecture we fill out some of the general theoretical discussion of the first lecture, by examining in some detail a particular physical system. This is the Fe-8 molecular magnetic system, which has the great advantages of (i) being well understood at the microscopic level, and (ii) well-characterised experimentally – it has been until recently a kind of 'Rosetta stone' for the testing of theory by experiment.

One can analyse this system both at the level of individual molecules, where we deal at low T with a prototype magnetic qubit coupled to its environment, or in crystalline arrays, where we deal with an interacting 'spin net' of qubits coupled to an environment.

In both cases there are various important parameter regimes of interest, involving the relative strengths of (i) the quantum energy Δ_o, driving the coherent spin dynamics; (ii) the decoherence energy ξ_o, characterising the coupling to the bath; and (iii) in the case of a spin net, the interqubit coupling V_o.

2.1: A CASE STUDY – the EFFECTIVE HAMILTONIAN for the Fe-8 MOLECULE

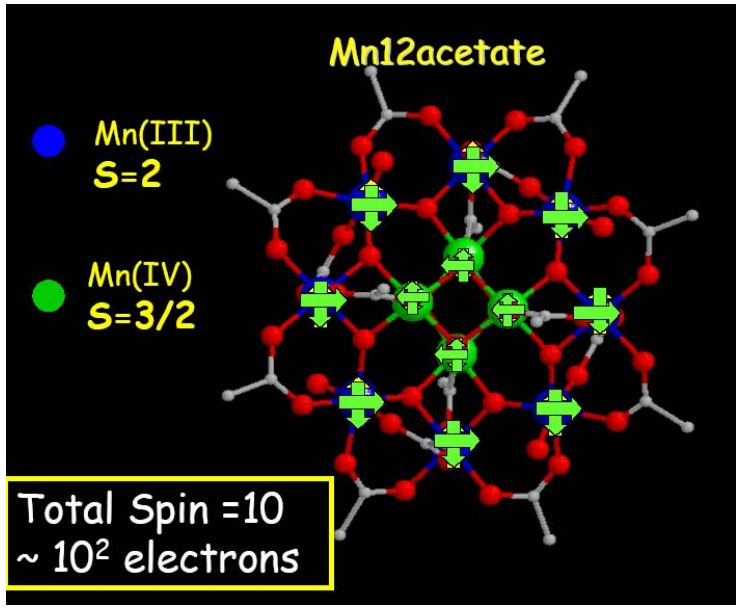
In order to focus the discussion, it is terribly important to see how theory gives a really detailed treatment of a real system, proceeding all the way from the abstract general formulation to detailed quantitative experimental predictions & comparison with experiment. In this lecture we concentrate on systems relevant to the construction of 'magnetic qubit' networks.

The Fe-8 molecule is ideal for this purpose, for 2 reasons. First: it can be analysed theoretically in very great detail, and essentially ALL relevant parameters can be determined quantitatively both theoretically and experimentally. Second: chemists have been able to prepare very well-characterised and pure samples of this system, so experiment and theory can be brought into intimate contact, & theory tested very thoroughly. This makes the Fe-8 system a kind of 'Rosetta stone' for the field (NB: recently very pure samples of the Mn-12 molecule have been prepared, without the usual Jahn-Teller distorted isomers – see lectures of Christou).

In what follows the detailed effective Hamiltonian for the Fe-8 system is derived quantitatively, including all relevant low-T excitations. We also make passing remarks about some of the other systems of interest, including the Mn-12 system and the $LiHo_xY_{1-x}F_4$.

Q. NANOMAGNETS: the GS Model

Chemists and physicists like to try and reduce the Hamiltonian of a nanomagnet to a simple 'Giant Spin' Model. This often works quite well, but only for certain systems (here we show the Mn-12 system):



See, eg. Tupitsyn, I.S., & Barbara, B. (2001) Drillon & J. Miller(Eds.). *Magnetoscience - from Molecules to Materials*(pp. 109-168). Weinheim: Wiley VCH Verlag Gmbh.

EXAMPLES of GIANT SPIN MODEL

(1) Transition-Metal based SMM's

$$H = -DS_z^2 - BS_z^4 - E(S_+^2 + S_-^2) - C(S_+^4 + S_-^4) + g\mu_B S_x H_x$$

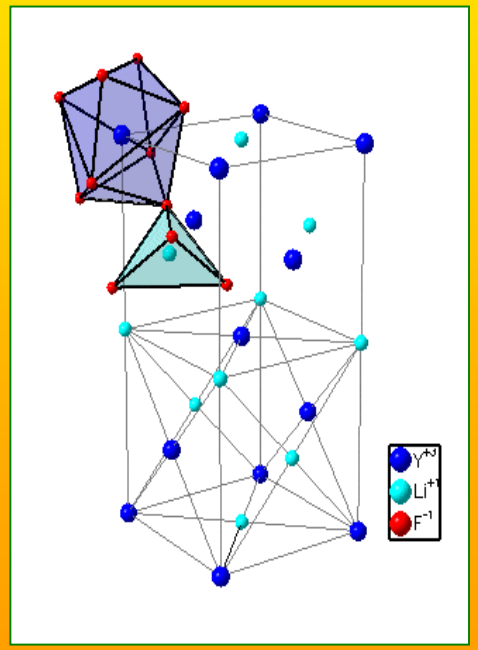
Mn_{12}-ac : D = 0.56 K, E = 5±2 mK, B = 1.18 mK, C = 3 10^{-5} K

Fe_8 : D = 0.23 K, E = - 47 mK, B = 0.03 mK,

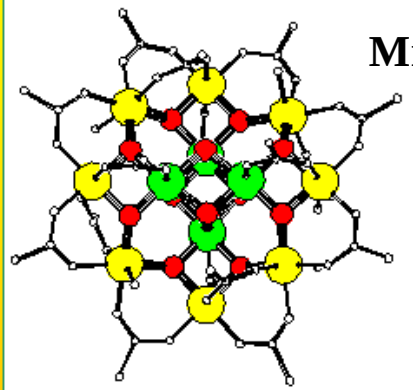
(2) Rare Earth Salts

$$H_{CF\text{-}Z} = -B_2^0 O_2^0 - B_4^0 O_4^0 - B_4^4 O_4^4 - B_6^0 O_6^0 - B_6^4 O_6^4 + g_J \mu_B \mathbf{JH}$$

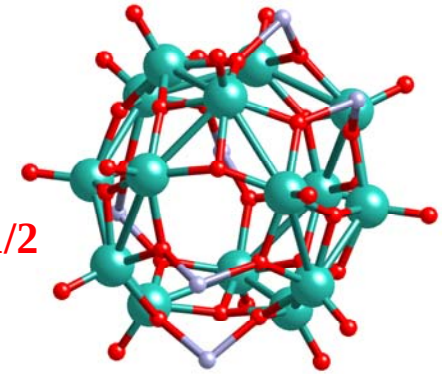
We are interested in systems which truncate to a low-E doublet



Ho ions in $LiYF_4$ host



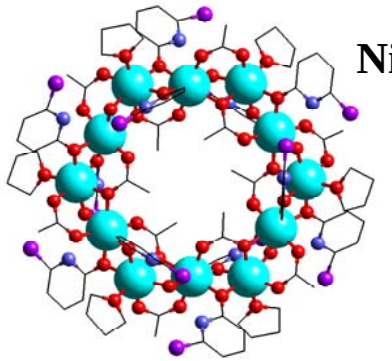
Mn_{12} S = 10



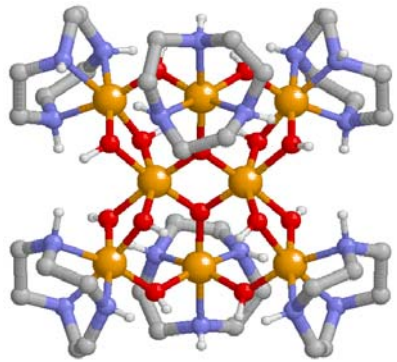
V_{15} S = 1/2

Single-molecule magnets (SMM)

Giant spins



Ni_{12} S = 12



Fe_8 S = 10

The TOPOLOGICAL SPIN PHASE

The Lagrangian for a constrained system like a spin is linear in the time derivative:

$$\mathcal{L} = \mathbf{A} \cdot \frac{\mathbf{dS}}{\mathbf{dt}} - \mathcal{H}_0(\mathbf{S})$$

Here A is the vector potential of a monopole at the centre of the unit spin sphere (Bloch sphere).

The monopole charge is quantized: $q = \hbar S$

This result has interesting consequences. One obvious one comes from considering the action:

$$\mathcal{A} = \int dt \mathcal{L}$$

When calculating the action, we typically integrate around a closed circuit on the Bloch sphere- then the integral over the 'kinetic' or 'topological' term gives a pure number:

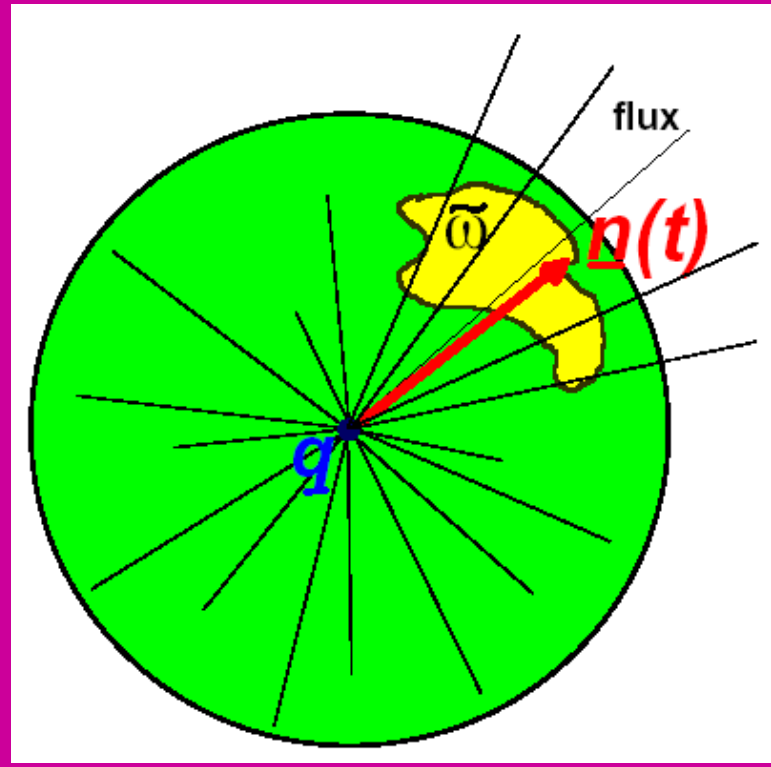
$$\phi_B = q/\hbar \int \mathcal{A} \cdot d\mathbf{n}$$

Here the unit vector $\hat{\mathbf{n}}(t) = \mathbf{S}(t)/S$ **just describes the spin dynamics on the Bloch sphere.**

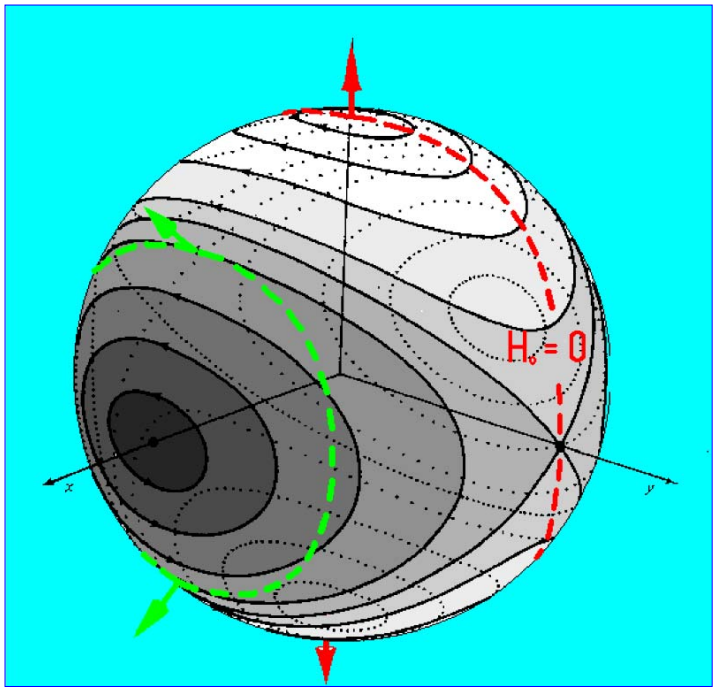
We immediately see that the total topological phase $\phi_B = \varpi S$ **is just proportional to the area swept out by the moving spin on the sphere. This is the famous Berry phase for a spin.**

Berry M V 1984 *Proc. R. Soc.* A **392** 45

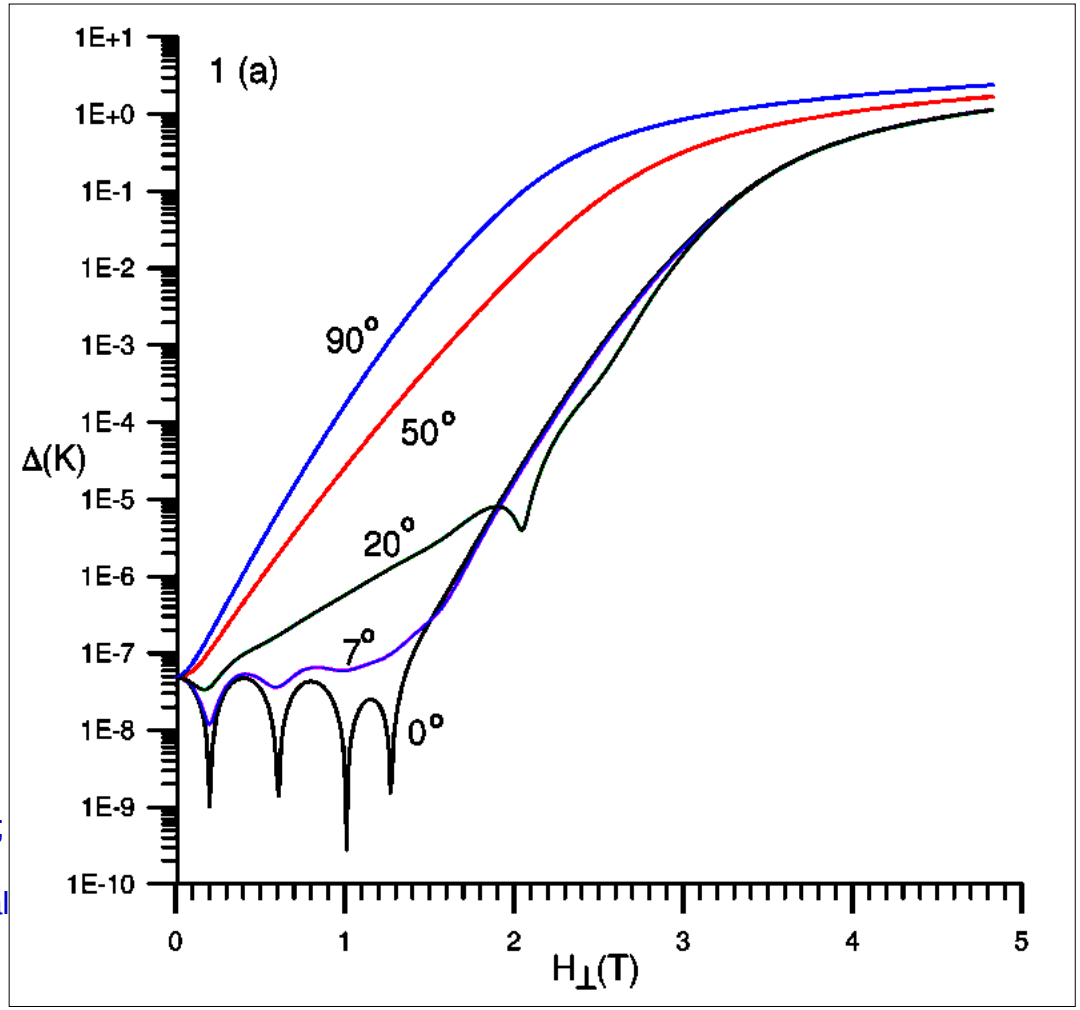
One elegant consequence of this is seen in the dynamics of a spin in a tunneling potential, where one can essentially do a 2-slit experiment with tunneling spins – this is shown on the enxt page



TUNNELING PATHS for SPINS on the BLOCH SPHERE

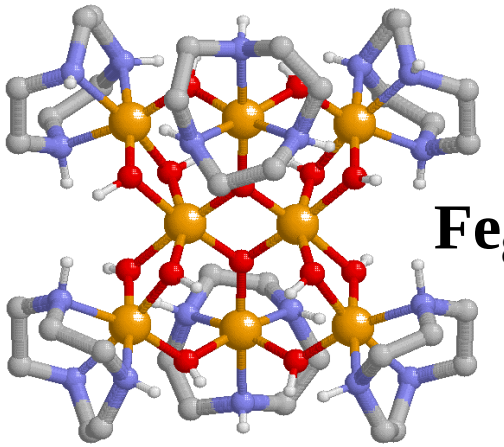


TOP: Tunneling paths for a spin on the Bloch sphere; the red paths are in zero applied field, and green in a strong field along the hard axis. The potential is biaxial – high energy is shaded dark. RIGHT: The tunneling amplitude between the minimum energy states, as a function of transverse field.



The spin moves preferentially between the 2 minimum energy states at the poles by tunneling along the pair of minimum action paths (shown as dashed lines), with amplitudes $\frac{1}{2}\Delta_\mu e^{i\varphi_\mu}$ respectively, where $\mu = 1, 2$ labels the paths, and the Δ_μ are real. An external field $\mathbf{H}_o^\perp$, applied along the hard $\hat{x}$-axis, pulls the 2 states, and the paths between them, towards $\hat{x}$, thereby reducing the enclosed area ϖ on the unit sphere. In this "symmetric" case, $\varphi_1 = -\varphi_2 = \varphi(\mathbf{H}_o^\perp)$ and $\Delta_1 = \Delta_2 = \Delta_o$. The total tunneling amplitude $\tilde{\Delta}$ is then just the sum $\frac{1}{2}\Delta_o(e^{i\varphi} + e^{-i\varphi})$, ie., $\tilde{\Delta}_o(\mathbf{H}_o^\perp) = \Delta_o(\mathbf{H}_o^\perp)\cos\varphi(\mathbf{H}_o^\perp)$.

The Fe-8 MOLECULE



Fe_8 **S = 10**

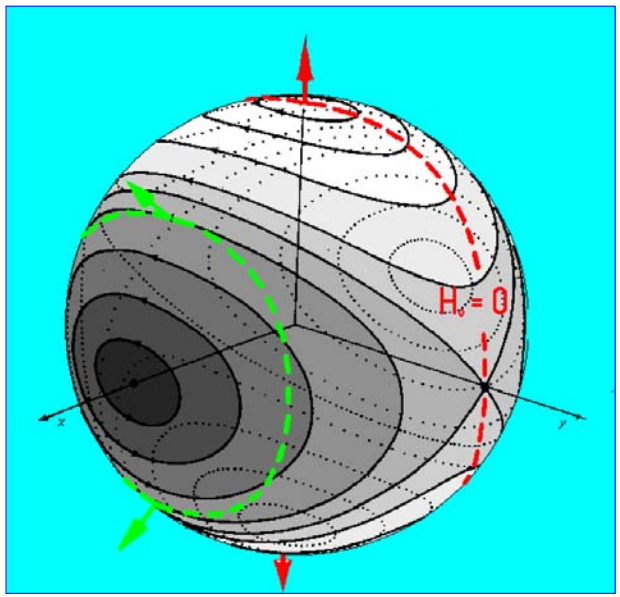
Low-T Quantum regime- effective Hamiltonian (T < 0.36 K):

$$\mathcal{H}_o(\hat{\tau}) = (\Delta_o \hat{\tau}_x + \epsilon_o \hat{\tau}_z)$$

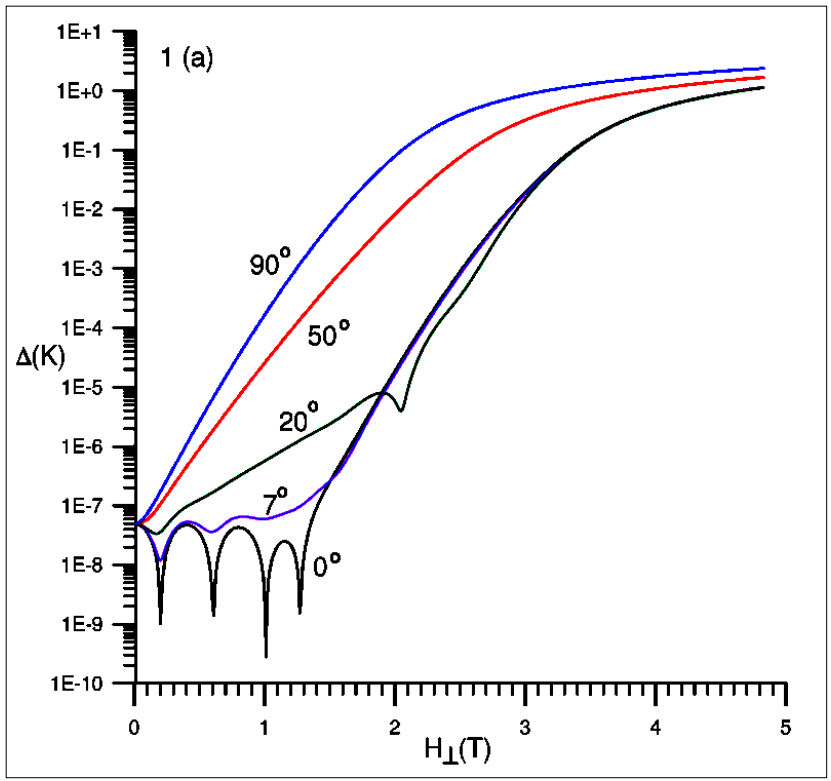
Longitudinal bias: $\epsilon_o = g\mu_B S_z H_o^z$

Eigenstates: $|\pm\rangle = [|\uparrow\rangle \pm |\downarrow\rangle]/\sqrt{2}$

Which also defines orthonormal states: $|\uparrow\rangle, |\downarrow\rangle$



Feynman Paths on the spin sphere for a biaxial potential. Application of a field pulls the paths towards the field

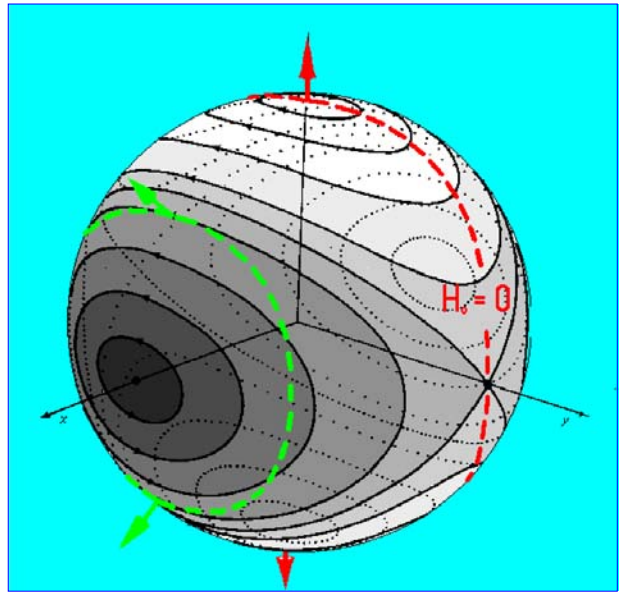


INSTANTON DERIVATION of H_{eff}

We begin by defining 'coherent states' for the central spin, which at the energy minima have form $\langle \vec{n}_1|\vec{S}|\vec{n}_1\rangle = S\vec{n}_1$ and $\langle \vec{n}_2|\vec{S}|\vec{n}_2\rangle = S\vec{n}_2$ We have an action for the giant spin of form

$$\mathcal{L}_o = -iS\dot{\theta}\varphi\sin\theta + H_o(\vec{n})$$

$$H_o(\vec{n}) = SK_{\|}\left[\sin^2\theta + \frac{K_\perp}{K_{\|}}\sin^2\theta\sin^2\varphi\right]$$ in zero field



The transition amplitude between the 2 states is

$$\Gamma^o_{\alpha\beta}(t) = \langle\vec{n}_\alpha|e^{-iH_o(\vec{S})t}|\vec{n}_\beta\rangle = \int_{\vec{n}(\tau=0)=\vec{n}_\beta}^{\vec{n}(\tau=t)=\vec{n}_\alpha}\mathcal{D}\vec{n}(\tau)\exp\left\{-\int_0^t d\tau\mathcal{L}_o(\tau)\right\}$$

We begin by ignoring fluctuations about the least action paths, so we can
Now write the Lagrangian as $\mathcal{L}_o(\theta) = \frac{S}{4K_\perp}\dot{\theta}^2 + SK_{\|}\sin^2\theta$

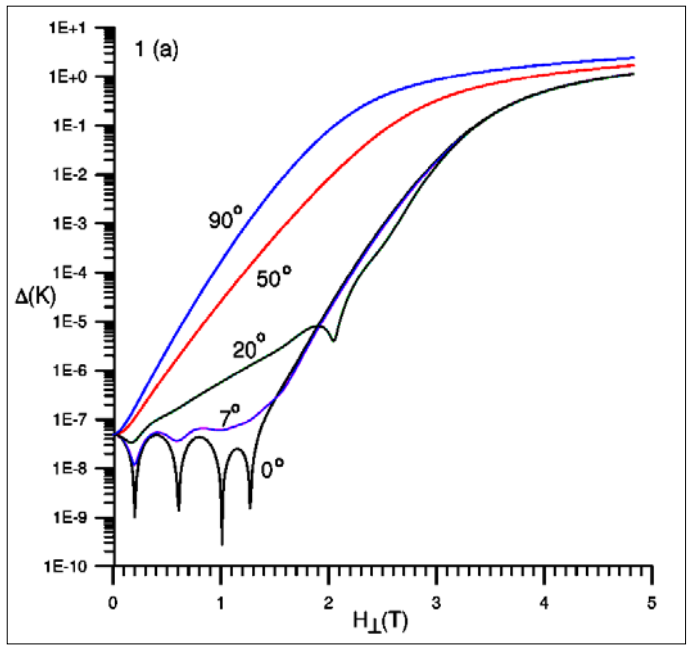
In a typical tunneling problem $K_\perp/K_{\|} \gg 1$ and we get the solution $\sin\theta(\tau) = 1/\cosh(\Omega_o\tau)$
with $\Omega_o = 2(K_{\|}K_\perp)^{1/2}$

There are 2 possible paths, which we label by $\eta = 1, -1$.
The action for these is $A_o^{(\eta)} = 2S(K_{\|}/K_\perp)^{1/2} + i\eta\pi S$

Thus we finally get a tunneling form for the effective Hamiltonian:

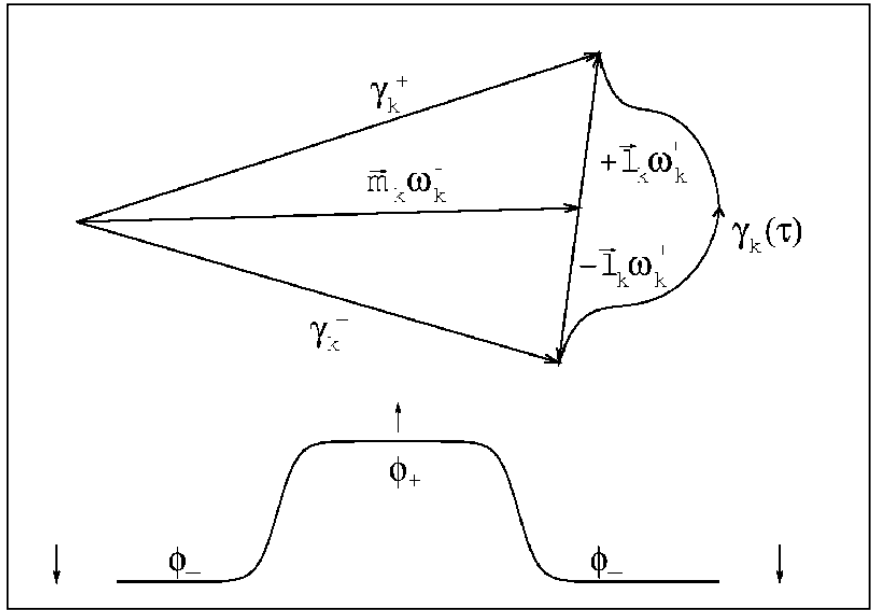
$$\hat{H}^o_{eff}(\vec{\tau}) = \Delta_o(S)\hat{\tau}_x$$

$$\Delta_o(S) = -\sum_{\eta=\pm}\sqrt{\frac{2}{\pi}ReA_o^{(\eta)}}\Omega_o\exp\{-A_o^{(\eta)}\} \equiv 2\Delta_o\cos\pi S$$



All this is easily generalized to finite applied field

Formal Derivation of "CENTRAL SPIN" Effective hamiltonian



(i) Start with the **k**-th bath spin, and define the vector field

$$\gamma_k(\tau) = h_k m_k + \omega_k l_k(\tau)$$

which varies as shown; the bath spin trajectory between the 2 end points can be calculated if we know the central spin trajectory. We also add the bath interspin interactions, to get the terms:

$$-\hat{\tau}_z \sum_{k=1}^{N} \omega_k^{\parallel} \vec{l}_k \cdot \hat{\vec{\sigma}}_k + \sum_{k=1}^{N} \omega_k^{\perp} \vec{m}_k \cdot \hat{\vec{\sigma}}_k + \sum_{k=1}^{N} \sum_{k'=1}^{N} V_{kk'}^{\alpha\beta} \hat{\sigma}_k^{\alpha} \hat{\sigma}_{k'}^{\beta}$$

(ii) Now define the "transfer matrix"

$$T_k^{\pm} = \exp\left\{ -\frac{1}{\hbar} \int_{-}^{+} d\tau \omega_k^{\parallel} \vec{l}_k(\tau) \cdot \vec{\sigma}_k \right\} \equiv e^{-i\varphi_k + \vec{\alpha}_k \cdot \vec{\sigma}_k}$$

where the scalar ϕ_k & the vector α_k are both complex.

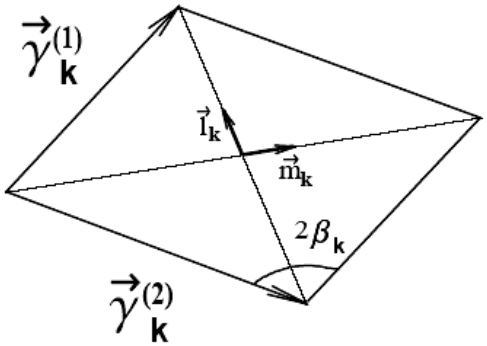
This finally gives:

$$H(\Omega_o) = \{ [\Delta\tau_+ \exp(-i \Sigma_k \alpha_k.\sigma_k) + H.c.] + \varepsilon_o \tau_z + \tau_z \omega_k.\sigma_k + h_k.\sigma_k + \text{inter-spin interactions}$$

HYPERFINE COUPLING to spin bath (NUCLEAR SPINS)

Hyperfine coupling: $\mathcal{H}_{NS}(\{\mathbf{I}_k\},\{\mathbf{s}_j\})$

$$= \left[\sum_{jk} A^{\alpha\beta}_{jk}\hat{s}^{\alpha}_{j}\hat{I}^{\beta}_{k} - \mu_N\sum_{k} g^{n}_{k}\hat{\mathbf{I}}_k\cdot\mathbf{H}_o + \sum_{k} Q^{k}_{\alpha\beta}I^{\alpha}_{k}I^{\beta}_{k}\right] + \sum_{k=1}^{N}\sum_{k'=1}^{N} V^{\alpha\beta}_{kk'}\hat{I}^{\alpha}_{k}\hat{I}^{\beta}_{k'}$$



Define the set of fields: $\gamma^{\beta}_{k} = \sum_{jk} A^{\alpha\beta}_{jk}s^{\alpha}_{j} - \mu_N g^{N}_{k}H^{\beta}_{o}$

Static component is:

$$\omega^{\perp}_{k}\hat{m}_k = g^{N}_{k}\mu_N\mathbf{H}_o$$

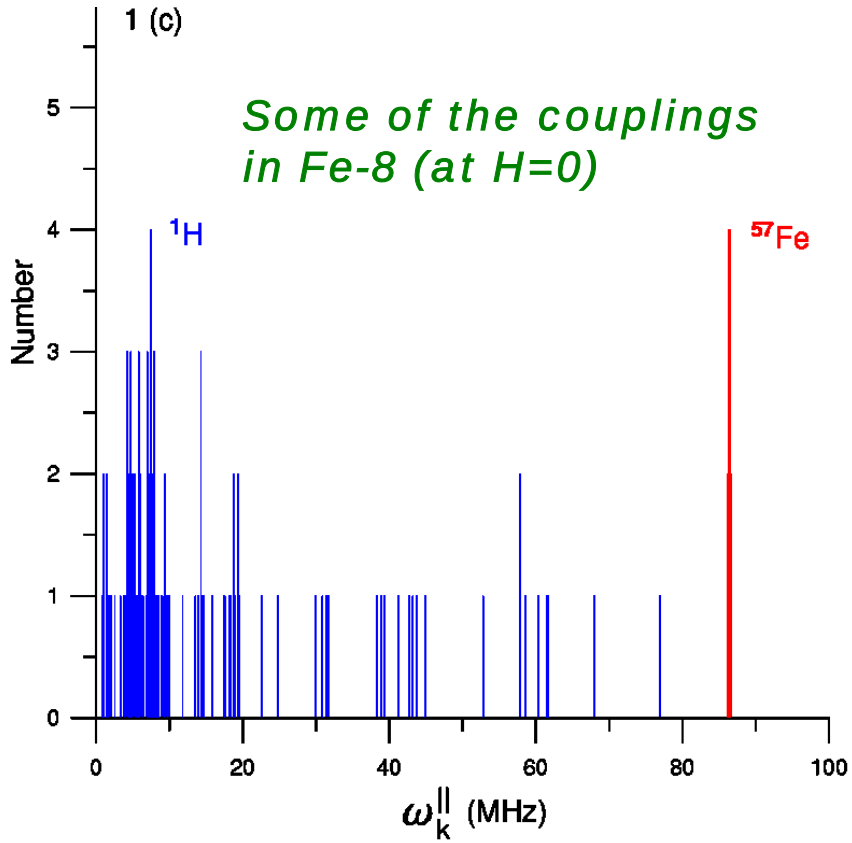
Component which flips is:

$$\omega^{\parallel}_{k}\hat{l}_k\cdot\mathbf{I}_k = \frac{1}{2}\sum_{j} A^{\alpha\beta}_{jk}\left[\langle s^{\alpha}_{j}\rangle^{\uparrow} - \langle s^{\alpha}_{j}\rangle^{\downarrow}\right] I^{\beta}_{k}$$

This gives the 'diagonal' terms in a 'central spin' Hamiltonian:

$$H^{D}_{hyp} = \hat{\tau}_z\sum_{k=1}^{N}\omega^{\parallel}_{k}\vec{l}_k\cdot\mathbf{I}_k + \sum_{k=1}^{N}\omega^{\perp}_{k}\vec{m}_k\cdot\mathbf{I}_k$$

$$+ \sum_{k=1}^{N}\sum_{k'=1}^{N} V^{\alpha\beta}_{kk'}\hat{I}^{\alpha}_{k}\hat{I}^{\beta}_{k'}$$



DERIVATION OF 'NON-DIAGONAL' COUPLING to BATH

See Tupitsyn I S, Prokof'ev N V and Stamp P C E 1997 *Int. J. Mod. Phys.* B **11** 2901

Suppose we now add a small magnetic field to the original problem. This then adds a small term to term to the Lagrangian of form

$$\delta\mathcal{L}_h = -\gamma_e S(H_o^x \sin\theta\cos\varphi + H_o^y \sin\theta\sin\varphi + H_o^z\cos\theta)$$

which upon linearization gives $\delta\mathcal{L}_h(\theta) = -\gamma_e S(H_o^x \sin\theta + \frac{i\dot\theta}{2K_\perp} H_o^y + H_o^z\cos\theta)$

The change $\delta A_h^{(\eta)}$ in the action is then $\int d\tau \delta\mathcal{L}_h(\vec S_o^{(\eta)}(\tau))$

which is easily found to be $\delta A_h^{(\eta)} = -\eta\frac{\gamma_e\pi S}{\Omega_o}(\hat{\vec x} + i\sqrt{K_\parallel/K_\perp}\hat{\vec y})\cdot\vec H_o$

Now let us consider the problem when we have an extra bath spin interacting with the giant central spin, so we have a contribution to action of form

$$\delta\mathcal{L}_\sigma(\tau) = \sum_{k=1}^N \vec\sigma_k(\tau)\cdot\vec\gamma_k(\tau)$$

with the field acting on the bath spin: $\vec\gamma_k(\tau) = \frac{\omega_k\vec S(\tau)}{2S}$

If we now write this as: $\delta A_\sigma^{(\eta)} = \sum_{k=1}^N \frac{\omega_k}{2}\int d\tau\vec\sigma_k(\tau)\cdot\vec h_{eff}^{(\eta)}(\tau) = \int d\tau\sum_{k=1}^N \delta\mathcal{L}_k^{(\eta)}(\tau)$

$$\vec h_{eff}^{(\eta)}(\tau) = \left(\eta\sin\theta^{(\eta)}(\tau),\ i\eta\sqrt{K_\parallel/K_\perp}\sin\theta^{(\eta)}(\tau),\ \cos\theta^{(\eta)}(\tau)\right)$$

We then easily find $\delta A_\sigma^{(\eta)} = \eta\sum_{k=1}^N \frac{\pi\omega_k}{2\Omega_o}(\hat{\vec x} + i\sqrt{K_\parallel/K_\perp}\hat{\vec y})\cdot\vec\sigma_k(\tau)$

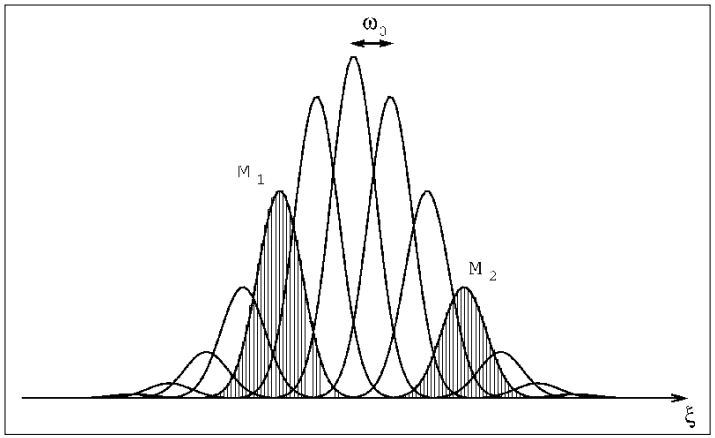
Thus we finally get the term:

$$2\Delta_o\hat\tau_- \cos\left[\pi S + \sum_k \alpha_k\vec n\cdot\hat{\vec\sigma}_k - \beta_o\vec n\cdot\vec H_o\right] + H.c.$$

$$\alpha_k\vec n = \frac{\pi\omega_k}{2\Omega_o}(-i\hat{\vec x},\ \sqrt{K_\parallel/K_\perp}\,\hat{\vec y})$$

$$\beta_o\vec n = \frac{\pi\gamma_e S}{\Omega_o}(-i\hat{\vec x},\ \sqrt{K_\parallel/K_\perp}\,\hat{\vec y})$$

Structure of NUCLEAR MULTIPLET in Fe-8



There are 215 nuclear sites in the molecule

Element	Z	$\lvert\vec{I}\rvert$	Moment $= g^n\lvert\vec{I}\rvert$ (in μ^n) $\mu^n = 7.62245\ MHz/T$	Abundance (%)
^{56}Fe	26	0	0	91.068
^{57}Fe	26	1/2	0.09	2.19
^{1}H	1	1/2	2.79267	99.985
^{2}H	1	1	0.857354	0.015
^{79}Br	35	3/2	2.0991	50.56
^{81}Br	35	3/2	2.2626	49.44
^{14}N	7	1	0.40365	99.63
^{15}N	7	1/2	-0.283	0.37
^{12}C	6	0	0	98.88
^{13}C	6	1/2	0.70225	1.12
^{16}O	8	0	0	99.757
^{17}O	8	5/2	-1.8935	0.039

Transitions between states of different total polarisation (T1 process) driven mainly by molecular tunneling)

Total width of gaussian multiplet:

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

(NB: This decreases with increasing applied field)

For Fe-8 at H=0, width is ~7 mK

(depends on isotopic concentrations)

For all practical purposes the effective Hamiltonian for a single molecule (& indeed any qubit) coupled to the spin bath is then:

$$H_{CS}(\hat{\tau}, \{\hat{\mathbf{I}}_k\}; \mathbf{H}_o) = \epsilon\hat{\tau}_z + \left\{ \frac{1}{2}\tilde{\Delta}\hat{\tau}_+ \exp\left[i\Phi_o(S, \mathbf{H}_o^{\perp}) - i\sum_k \alpha_k \vec{n}_k \cdot \hat{\mathbf{I}}_k \right] + H.c. \right\}$$

$$+ \hat{\tau}_z \sum_{k=1}^{N} \omega_k^{\parallel} \vec{l}_k \cdot \mathbf{I}_k + \sum_{k=1}^{N} \omega_k^{\perp} \vec{m}_k \cdot \mathbf{I}_k + \sum_k \xi_k^{\alpha}(t) \hat{I}_k^{\alpha}$$

SPIN-PHONON COUPLINGS

In general we expect a coupling $V_{sp-ph} = \sum_q \sqrt{\frac{\hbar}{2NM\omega_q}} V(\vec{S})(iqb_q^+ - iqb_q)$

between Giant Spin & phonons, which we write as $\hat{V}_{sp-ph} = \sum_i \eta_i \hat{O}_i^S \hat{O}_i^P$
(a product of spin and phonon operators).

Key parameters: Debye energy

the longitudinal sound velocity $c_s = (\Theta_D k_B/\hbar)(V_c/6\pi^2)^{1/3}$

Define symmetric and antisymmetric strains:

$$\epsilon_{\alpha\beta} = (\partial_\beta u_\alpha + \partial_\alpha u_\beta)/2$$
$$\omega_{\alpha\beta} = (\partial_\beta u_\alpha - \partial_\alpha u_\beta)/2$$

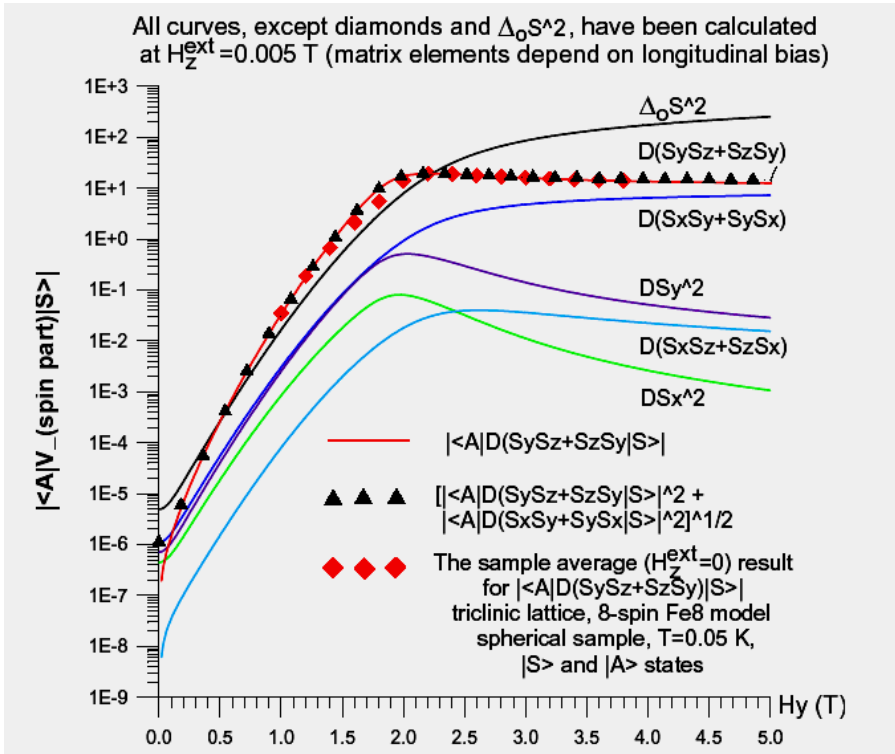
The dominant couplings for Fe-8, particularly in high transverse field, are

$$(g_1^{ph}\epsilon_{xz} + g_2^{ph}\omega_{xz})[\hat{S}_x\hat{S}_z + \hat{S}_z\hat{S}_x]$$
$$(g_3^{ph}\epsilon_{yz} + g_4^{ph}\omega_{yz})[\hat{S}_y\hat{S}_z + \hat{S}_z\hat{S}_y]$$

and simple arguments show that $g_i \sim D$

All properties can be written in terms of matrix elements

$$\mathcal{M}^2_{\mathcal{AS}} = \int \frac{d\Omega}{4\pi^2} \left| \sum_i g_i^{ph} f_i(\theta,\varphi) \langle \mathcal{A}|O_i^S|\mathcal{S}\rangle \right|^2$$



NB: One finds that for simple estimations it is sufficient to write this in the form:

$$H_{ph} = \sum_q c_q^\perp \hat{\tau}_x x_q + \sum_q \left[\frac{p_q^2}{2m_q} + \frac{1}{2}m_q^2\omega_q^2 x_q^2\right]$$

where $c_q^\perp x_q \sim S\Omega_o(\omega_q/\theta_D)^{1/2}$

2.2: QUICK NOTE on DYNAMICS of SPIN-BOSON & CENTRAL SPIN MODELS

This material is optional, and for those interested in how detailed calculations of the dynamics are done. There are review articles on the dynamics of both the spin-boson and central spin models. For those who only want a summary of the results, go to the next section in this lecture (section 2.3).

The 2 reviews in question are:

Leggett A J *et al* 1987 *Rev. Mod. Phys.* **59** 1

N.V. Prokof'ev, P.C.E. Stamp, Rep. Prog. Phys. **63**, 669 (2000)

Dynamics of Spin-Boson System

The easiest way to solve for the dynamics of the spin-boson model is in a path integral formulation. The qubit density matrix propagator is written as an integral over an "influence functional" :

$$K(1,2) = \int_{Q_1}^{Q_2} \mathrm{d}Q \int_{Q_1'}^{Q_2'} \mathrm{d}Q' \mathrm{e}^{-\mathrm{i}/\hbar(S_0[Q]-S_0[Q'])} \mathcal{F}[Q,Q']$$

The influence functional is defined as

$$\mathcal{F}[Q,Q'] = \prod_k \langle \hat{U}_k(Q,t)\hat{U}_k^\dagger(Q',t)\rangle$$

For an oscillator bath:

$$F[x(\tau),y(\tau')] = \exp -\frac{1}{\hbar}\int_{t_0}^{t} d\tau \int_{t_0}^{\tau} ds\,[x(\tau)-y(\tau)][\gamma(\tau-s)x(s)-\gamma^*(\tau-s)y(s)]$$

with bath propagator:

$$\gamma(\tau-s) \equiv \sum_\alpha (C_\alpha^2/2m_\alpha\omega_\alpha)\left[\exp -i\omega_\alpha(\tau-s) + \frac{2\cos\omega_\alpha(\tau-s)}{\exp(\beta\hbar\omega_\alpha)-1}\right]$$

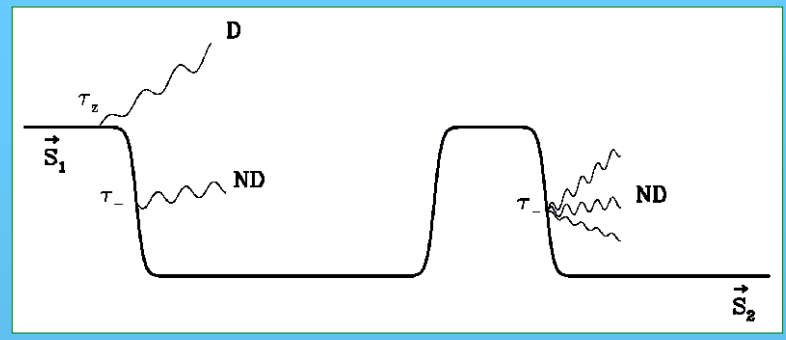
For a qubit the path reduces to

$$Q_{(n)}(s) = 1 - \sum_{i=1}^{2n}[\mathrm{sgn}(s-t_{2i-1}) + \mathrm{sgn}(t_{2i}-s)]$$

Thence

$$P_{11}(t) = \sum_{nm}^{\infty} (\mathrm{i}\Delta_0)^{2(n+m)} \int_0^t \mathrm{d}t_1 \cdots \int_{t_{2n-1}}^t \mathrm{d}t_{2n} \int_0^t \mathrm{d}t_1' \cdots \int_{t_{2m-1}'}^t \mathrm{d}t_{2m}' \mathcal{F}[Q_{(n)}, Q_{(m)'}]$$

Dynamics of Central Spin model (Qubit coupled to spin bath)



The propagators contain the following averages

Topological phase average

$$\sum_{\nu=-\infty}^{\infty} F_{\lambda'}(\nu) \int \frac{d\varphi}{2\pi} e^{i2\nu(\Phi-\varphi)}$$

Orthogonality average

$$\int_0^{\infty} dx x e^{-x^2}$$

Bias average

$$\int d\epsilon W(\epsilon) \frac{e^{-\beta\epsilon}}{Z(\beta)} \sum_M$$

The reduced density matrix, after the spin bath is integrated out, is given quite generally by:

$$\rho(Q,Q';t) = \hat{\mathscr{A}}^T(\phi)\hat{\mathscr{A}}^O(y)\hat{\mathscr{A}}^B(\epsilon,M) \times \rho^{(o)}(\phi,y,\epsilon,M;Q,Q';t)$$

Eg., for a single qubit, we get the return probability:

$$P_{11}^{(0)}(t;\Delta_M(\varphi,x);\epsilon) = 1 - \frac{\Delta_M^2(\varphi,x)}{E_M^2(\varphi,x)} \sin^2(E_M(\varphi,x)t)$$

$$E_M^2(\varphi,x) = \Delta_M^2(\varphi,x) + \epsilon^2$$

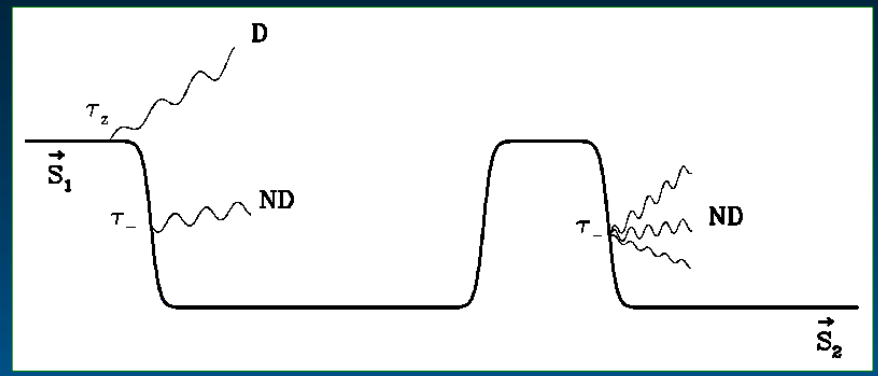
$$\Delta_M(\varphi,x) = 2\tilde{\Delta}_0 |\cos(\varphi) J_M(2x\sqrt{\gamma})|$$

$$\int \mathcal{D}\epsilon(t) e^{-\frac{1}{2}\int dt_1 \int dt_2 K(t_1-t_2)\epsilon(t_1)\epsilon(t_2)}$$

NB: can also deal with external noise

Dynamics of Central Spin Model- some key points

The easiest way to solve for the dynamics of problems like this (going back to the Kondo problem) is a path integral formulation. The effective Hamiltonian has both diagonal (D) and non-diagonal (ND) couplings in the qubit variables. A typical path is shown at right.



The standard way of doing such path integrals assumes a weak coupling to each environmental mode, assumed to be an oscillator- one then writes it as an integral over an "influence functional" (a la Feynman).

$$P_{11}(t) = \sum_{nm}^{\infty} (i\Delta_0)^{2(n+m)} \int_0^t dt_1 \cdots \int_{t_{2n-1}}^t dt_{2n} \int_0^t dt'_1 \cdots \int_{t'_{2m-1}}^t dt'_{2m} \mathcal{F}[Q_{(n)}, Q_{(m)'}].$$

The problem when one couples a qubit to a spin bath is that this assumption is no longer generally true- very often the coupling between the qubit and each bath spin is quite strong (indeed, with magnets, it can be much bigger even than Δ_o). Thus with a Hamiltonian like the Central spin model (we recall again the Hamiltonian here), we cannot use the influence functional technique.

$$H_{CS}(\Omega_0) = \left\{ 2\tilde{\Delta}\hat{\tau}_- \cos\left[\Phi - \sum_k \vec{V}_k \cdot \hat{\vec{\sigma}}_k\right] + \text{H.c.} \right\}$$
$$+\hat{\tau}_z \sum_{k=1}^{N} \omega_k^{\parallel} \vec{l}_k \cdot \hat{\vec{\sigma}}_k + \sum_{k=1}^{N} \omega_k^{\perp} \vec{m}_k \cdot \hat{\vec{\sigma}}_k + \sum_{k=1}^{N} \sum_{k'=1}^{N} V_{kk'}^{\alpha\beta} \hat{\sigma}_k^{\alpha} \hat{\sigma}_{k'}^{\beta}$$

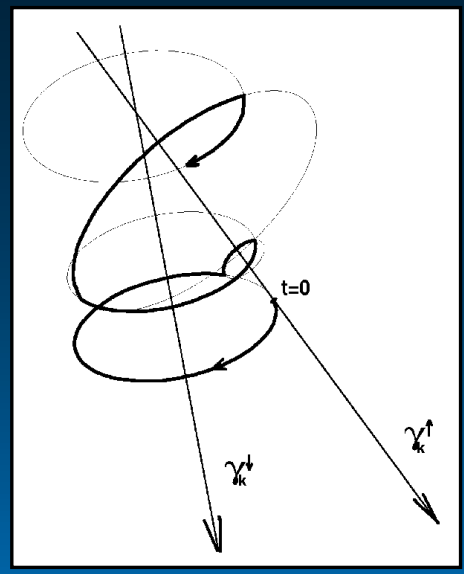
Precessional Decoherence- derivation

$$H_{\rm eff} = 2\Delta_0\tau_x + \hat{\tau}_z\omega_0^{\parallel}\sum_{k=1}^{N}\vec{l}_k\cdot\hat{\vec{\sigma}}_k + \sum_{k=1}^{N}\omega_k^{\perp}\vec{m}_k\cdot\hat{\vec{\sigma}}_k.$$

The most important physical effects are contained in the above reduced Hamiltonian. To handle this we introduce a unitary rotation between the 2 field directions

$$|\{\vec{\sigma}_k^{\,f}\}\rangle = \prod_{k=1}^{N}\hat{U}_k|\{\vec{\sigma}_k^{\rm in}\}\rangle = \hat{U}|\{\vec{\sigma}_k^{\rm in}\}\rangle.$$

$$|\vec{\sigma}_k^{\,f}\rangle = \hat{U}_k|\vec{\sigma}_k^{\rm in}\rangle = {\rm e}^{-{\rm i}\beta_k\hat{\sigma}_k^x}|\vec{\sigma}_k^{\rm in}\rangle.$$



The 2 qubit fields

The constraint of long nuclear T_1 means polarisation group M does not change- implemented in a path integral with a projection operator Π_M

$$\hat{\Pi}_M = \delta\left(\sum_{k=1}^{N}\hat{\sigma}_k^z - M\right) = \int_0^{2\pi}\frac{{\rm d}\xi}{2\pi}{\rm e}^{{\rm i}\xi\left(\sum_{k=1}^{N}\hat{\sigma}_k^z - M\right)}.$$

The dynamics of the qubit reduced density matrix are found by summing over paths, using the angle β_k (the angle between the fields γ_k^{+} and γ_k^{-}) as expansion parameter.

Physically, the bath spins precess around the 2 qubit fields, and the integration picks up the precessional phase (top right)

$$P_{M_0,M}(t) \equiv \langle R^*_{M_0,M}(t)R_{M_0,M}(t)\rangle$$
$$= \sum_{n=0}^{\infty}\sum_{m=0}^{\infty}\frac{({\rm i}\Delta_0(\Phi)t)^{2(n+m)}}{(2n)!(2m)!}\prod_{i=1}^{2n}\prod_{j=1}^{2m}\int\frac{{\rm d}\xi_i}{2\pi}$$
$$\times\int\frac{{\rm d}\xi'_j}{2\pi}{\rm e}^{-{\rm i}M_0\left(\sum_i^{2n}\xi_i - \sum_j^{2m}\xi'_j\right)}{\rm e}^{2{\rm i}M\left(\sum_{\rm i=odd}^{2n-1}\xi_i - \sum_{j=\rm odd}^{2m-1}\xi'_j\right)}\langle\hat{T}^{\dagger}_{2m}\hat{T}_{2n}\rangle.$$

$$\hat{T}^{(k)}_{2n}|\uparrow_k\rangle = {\rm e}^{{\rm i}\xi_{2n}\hat{\sigma}_k^z}{\rm e}^{-{\rm i}\beta_k\hat{\sigma}_k^x}\ldots{\rm e}^{-{\rm i}\beta_k\hat{\sigma}_k^x}{\rm e}^{{\rm i}\xi_1\hat{\sigma}_k^z}{\rm e}^{{\rm i}\beta_k\hat{\sigma}_k^x}|\uparrow_k\rangle$$
$$= {\rm e}^{{\rm i}\sum_{i=1}^{2n}\xi_i}\Bigg[(1-n\beta_k^2)|\uparrow_k\rangle + {\rm i}\beta_k|\downarrow_k\rangle\sum_{l=1}^{2n}(-1)^{l+1}{\rm e}^{-2{\rm i}\sum_{i=l}^{2n}\xi_i}$$
$$-\beta_k^2|\uparrow_k\rangle\sum_{l'=l+1}^{2n}\sum_{l=1}^{2n-1}(-1)^{l'-l}{\rm e}^{-2{\rm i}\sum_{i=l}^{l'-1}\xi_i} + {\rm O}(\beta_k^3)\Bigg],$$

The path integral splits into contributions for each **M**. They have the effective action of a set of interacting instantons

$$P_M(t) = \sum_{n=0}^{\infty}\sum_{m=0}^{\infty} \frac{(\mathrm{i}\Delta_0(\Phi)t)^{2(n+m)}}{(2n)!(2m)!} \prod_{i=1}^{2n}\prod_{j=1}^{2m}\int \frac{\mathrm{d}\xi_i}{2\pi} \times \int \frac{\mathrm{d}\xi'_j}{2\pi} \exp\{2\mathrm{i}M(\xi_{2n-1}+\xi_{2n-3}+\cdots+\xi_1) - K^{\mathrm{eff}}_{nm}(\xi_i,\xi'_j)\}.$$

$$\chi_\alpha = \sum_{\alpha'=\alpha}^{2(n+m)} 2\xi_{\alpha'} + \pi\alpha,$$

$$P_M(t) = \sum_{n=0}^{\infty}\sum_{m=0}^{\infty} \frac{(\mathrm{i}\Delta_0(\Phi)t)^{2(n+m)}}{(2n)!(2m)!} \left(\prod_{\alpha=1}^{2(n+m)}\int \frac{\mathrm{d}\chi_\alpha}{2\pi}\right) \times \exp\left\{\mathrm{i}M\sum_\alpha(-1)^{\alpha+1}\chi_\alpha - 2\kappa\left[(n+m)+\sum_{\alpha'>\alpha}\cos(\chi_\alpha-\chi_{\alpha'})\right]\right\}.$$

The effective interactions can be mapped to a set of fake charges to produce an action having the structure of a "spherical model" involving a spin **S**

$$\vec{\mathcal{S}} = \sum_{\alpha=1}^{2(n+m)} \vec{s}_\alpha, \qquad \sum_{\alpha',\alpha}\cos(\chi_\alpha-\chi_{\alpha'}) = \vec{\mathcal{S}}^2.$$

$$G(\vec{\mathcal{S}}) = \left(\prod_{\alpha=1}^{2(n+m)}\int \frac{\mathrm{d}\chi_\alpha}{2\pi}\mathrm{e}^{\mathrm{i}M(-1)^{\alpha+1}\chi_\alpha}\right)\exp\left\{-\kappa\sum_{\alpha',\alpha}\cos(\chi_\alpha-\chi_{\alpha'})\right\} = \left(\prod_{\alpha=1}^{2(n+m)}\int \frac{\mathrm{d}\chi_\alpha}{2\pi}\mathrm{e}^{\mathrm{i}M(-1)^{\alpha+1}\chi_\alpha}\right)\mathrm{e}^{-\kappa\vec{\mathcal{S}}^2}.$$

The key step is to then reduce this to a sum over Bessel functions associated with each polarisation group.

$$G(\vec{\mathcal{S}}) = \int \mathrm{d}\vec{\mathcal{S}}\mathrm{e}^{-\kappa\vec{\mathcal{S}}^2}\prod_{\alpha=1}^{2(n+m)}\int\frac{\mathrm{d}\chi_\alpha}{2\pi}\mathrm{e}^{\mathrm{i}M(-1)^{\alpha+1}\chi_\alpha}\delta\left(\vec{\mathcal{S}}-\sum_\alpha\vec{s}_\alpha\right) = \int\frac{\mathrm{d}\vec{z}}{2\pi}\int\mathrm{d}\vec{\mathcal{S}}\mathrm{e}^{-\kappa\vec{\mathcal{S}}^2+\mathrm{i}\vec{z}\cdot\vec{\mathcal{S}}}\left(\int_0^{2\pi}\frac{\mathrm{d}\chi_\alpha}{2\pi}\mathrm{e}^{-\mathrm{i}\vec{z}\vec{s}_\alpha+\mathrm{i}M\chi_\alpha}\right)^{2(n+m)} = \frac{1}{2\kappa}\int \mathrm{d}z z\mathrm{e}^{-z^2/4\kappa}J_M^{2(n+m)}(z),$$

We can now reduce the time evolution of the qubit density matrix to a sum over independently relaxing polarisation groups.

The interesting thing here is that each group has its own effective tunneling matrix element $\Delta_M(x)$.

$$P_M(t) = 2\int_0^\infty dx x e^{-x^2}\frac{1}{2}\left(1+\sum_{s=0}^{\infty}\frac{[2it\,\Delta_0(\Phi)J_M(2x\sqrt{\kappa})]^{2s}}{(2s)!}\right)$$

$$= P_M(t) = \int_0^\infty dx x e^{-x^2}(1+\cos[2\Delta_0(\Phi)J_M(2x\sqrt{\kappa})t])$$

$$\equiv 2\int_0^\infty dx x e^{-x^2} P_{11}^{(0)}(t, \Delta_M(x)),$$

a *superposition of non-interacting* correlation functions for effective tunnelling rates $\Delta_M(x)$

But.. $\Delta_M(x)$ has to be phase-averaged over a phase variable **x**. This variable represents the accumulated precessional phase of the spin bath. The total dephasing effect of this average is parametrised by the dimensionless κ.

$$\Delta_M(x) = \Delta_0(\Phi)\,J_M(2x\sqrt{\kappa}).$$

This parameter tells us the total effect of the mismatch between the 2 fields from the qubit on the bath spins (which is parametrised for each bath spin by the angle β_k). The total effect is reminiscent of the reduction of transition rates embodied in the Frank-Condon or Anderson orthogonality reduction factors.

$$e^{-\kappa} = \prod_{k=1}^{N}\cos\beta_k \sim e^{-(1/2)\sum_k \beta_k^2}.$$

Finally, we have to perform a thermal average over different polarisation groups.

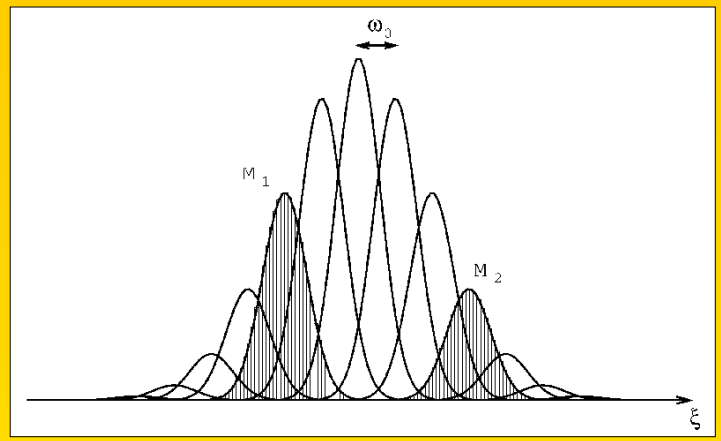
$$P_{11}(t;T) = \sum_{M=-N}^{N} w(T,M)P_M(t),$$

2.3: QUICK SUMMARY of PREDICTIONS for the DYNAMICS of a *SINGLE* QUANTUM NANOMAGNET

Here we summarize the results of calculations of the dynamics of individual qubits in both (i) the 'quantum relaxation' regime (where decoherence/dissipation from the bath dominate the dynamics, ie, where $\Delta_o << \xi_o$); & in the quantum regime, where $\Delta_o >> \xi_o$, and the system behaves coherently for long periods. We also make a few remarks on how this applies to Fe-8 and to $LiHo_xY_{1-x}F_4$.

Quantum Relaxation of a single NANOMAGNET

Structure of Nuclear spin Multiplet →



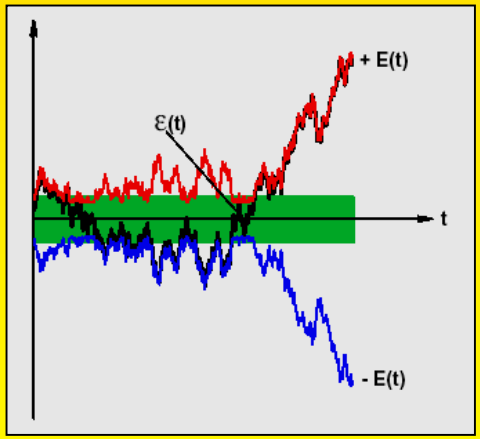
Our Hamiltonian:

$$\mathcal{H}_{NS} = \mathcal{H}_o(\hat{\tau}) + V(\hat{\tau}, \mathbf{I}_k)$$

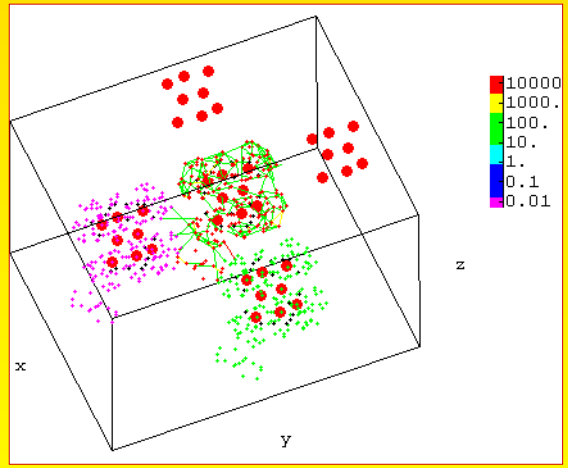
$$\mathcal{V} = \hat{\tau}_z \left[\xi^z(t) + \sum_k \omega_k^{\parallel} \hat{l}_k \cdot \hat{\mathbf{I}}_k \right] + \sum_k g_k^N \mu_N \mathbf{I}_k \cdot \mathbf{H}_o$$

When $\Delta << E_o$ (linewidth of the nuclear multiplet states around each magbit level), the magbit relaxes via incoherent tunneling. The nuclear bias acts like a rapidly varying noise field, causing the magbit to move rapidly in and out of resonance, PROVIDED

$$|g\mu_B S H_o| < E_o$$



Fluctuating noise field



Nuclear spin diffusion paths

Tunneling now proceeds over a range E_o of bias, governed by the NUCLEAR SPINmultiplet. The relaxation rate is

$\Gamma \sim \Delta^2/E_o$ **for a single qubit.**

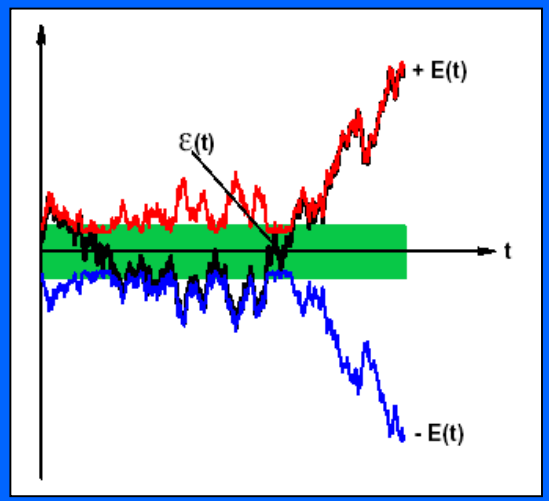
NV Prokof'ev, PCE Stamp, J Low Temp Phys 104, 143 (1996)

DYNAMICS of SPIN BATH DECOHERENCE

$$H_{CS}(\Omega_0) = \left\{ 2\tilde{\Delta}\hat{\tau}_- \cos\left[\Phi - \sum_k \vec{V}_k \cdot \hat{\vec{\sigma}}_k\right] + \text{H.c.} \right\}$$
$$+\hat{\tau}_z \sum_{k=1}^{N} \omega_k^{\parallel} \vec{l}_k \cdot \hat{\vec{\sigma}}_k + \sum_{k=1}^{N} \omega_k^{\perp} \vec{m}_k \cdot \hat{\vec{\sigma}}_k + \sum_{k=1}^{N}\sum_{k'=1}^{N} V_{kk'}^{\alpha\beta} \hat{\sigma}_k^{\alpha} \hat{\sigma}_{k'}^{\beta}$$

At first glance a solution of this seems very forbidding. However it turns out that one can solve for the reduced density matrix of the central spin exactly, in the interesting parameter regimes. From this solt^{n} the decoherence mechanisms are easy to identify:

(i) Noise decoherence: Random phases added to different Feynman paths by the noise field.

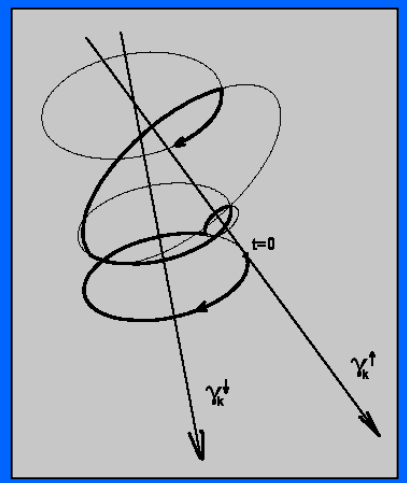


Noise decoherence source

(ii) Precessional decoherence: the phase accumulated by environmental spins between qubit flips.

(iii) Topological Decoherence: The phase induced in the environmental spin dynamics by the qubit flip itself

USUALLY THE 2ND MECHANISM (PRECESSIONAL DECOHERENCE) is DOMINANT

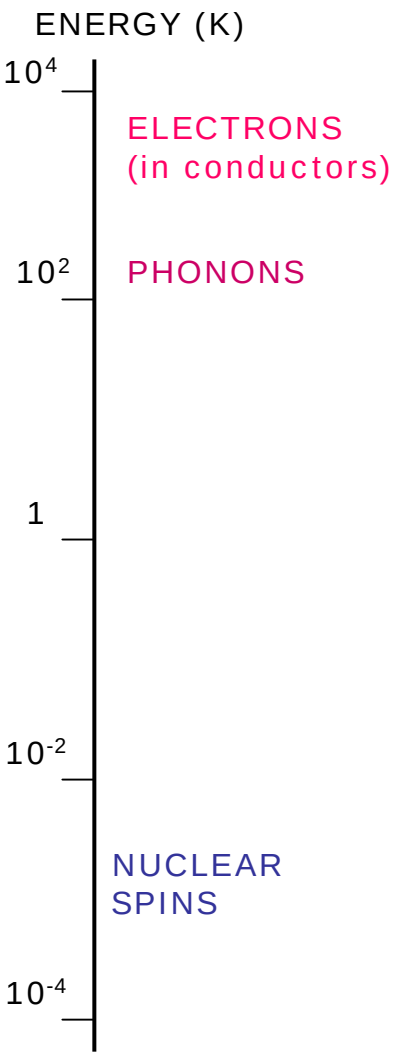


Precessional decoherence

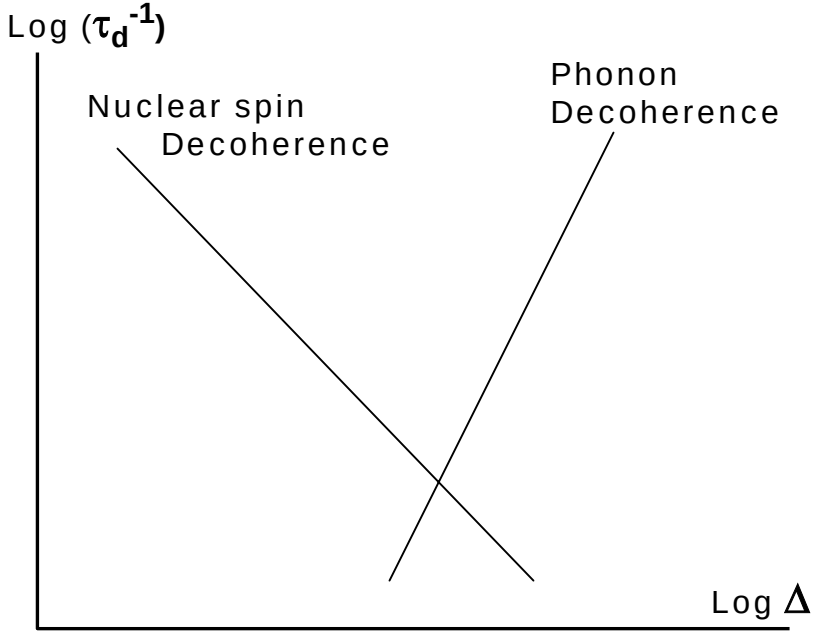
The COHERENCE WINDOW

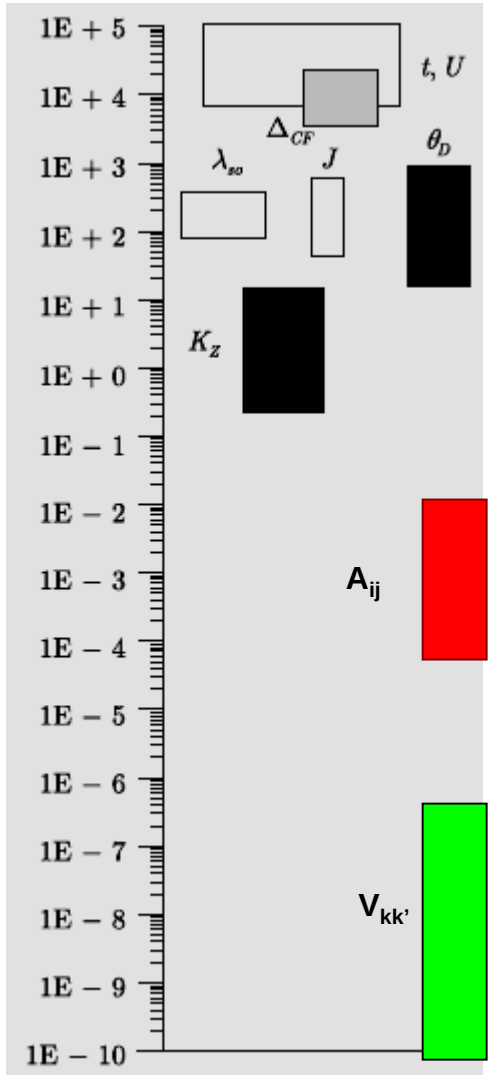
In solid-state qubit systems, the coherence window arises because of the large separation of energy scales typically existing between spin and oscillator baths. This coherence window exists in ALL solid-state systems- we look here at magnetic systems

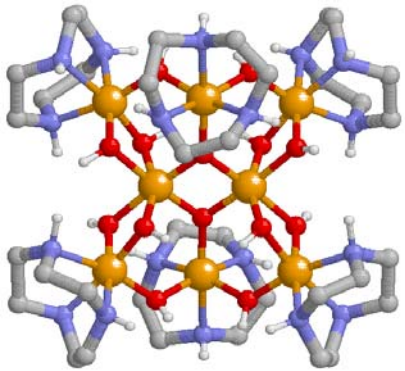
M Dube, PCE Stamp, Chem Phys 268, 257 (2001)
PCE Stamp, J Q Comp & Computing 4, 20 (2003)
PCE Stamp, IS Tupitsyn, Phys Rev B69, 014401 (2004)



If we now fix the operating frequency Δ of the qubits to lie well below the high phonon frequencies, but well above the characteristic nuclear spin frequencies (given by hyperfine couplings, then the phonons are too fast to cause decoherence, & the nuclear spins too slow.

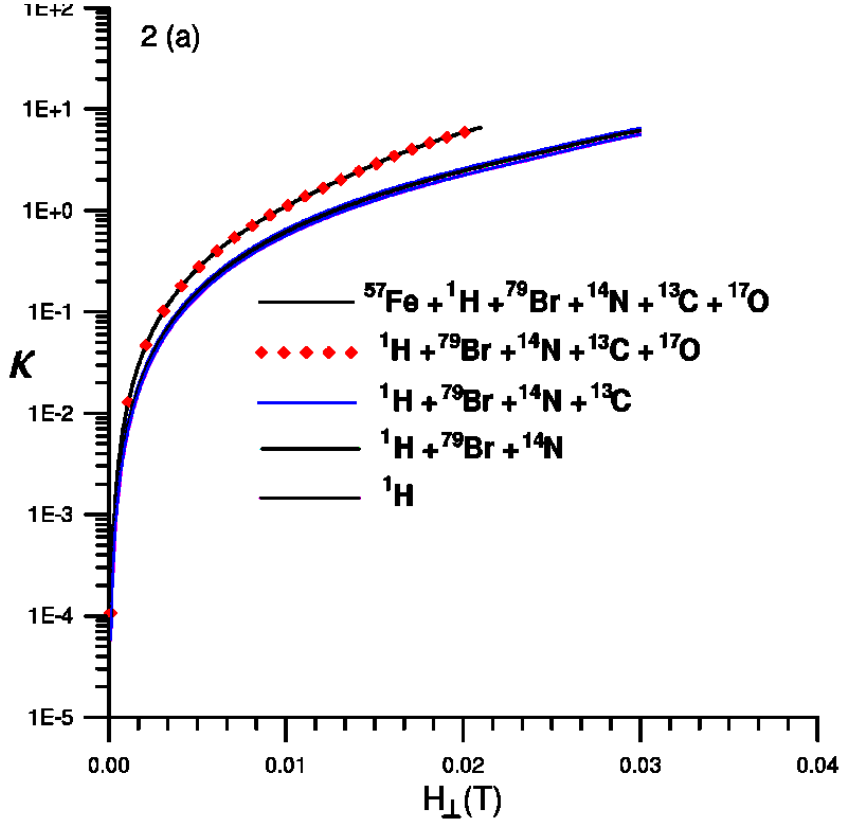


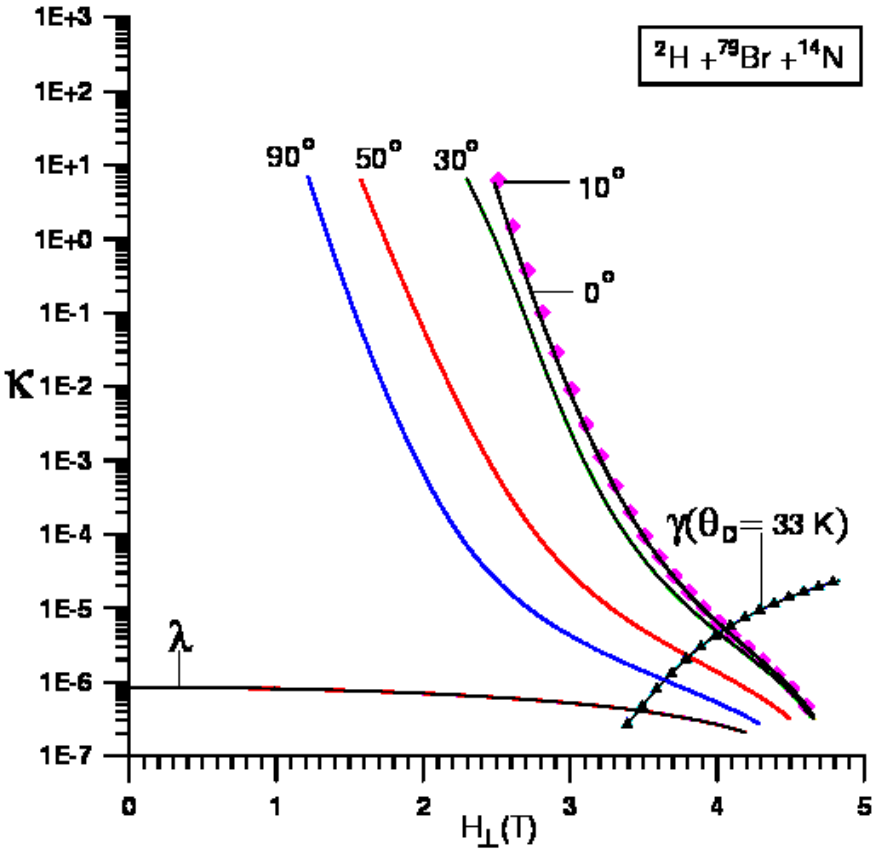




DECOHERENCE in the Fe-8 Molecule

At low applied transverse Fields, decoherence switches on very fast- expect incoherent spin relaxation:





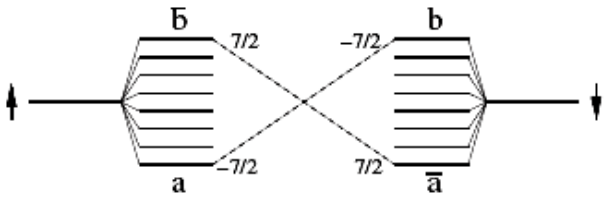
Stamp, P.C.E., Tupitsyn, I.S., Phys Rev **B69**, 014401 (2004)

However, at high fields, system can be in coherence window, in which qubit dynamics is too fast for nuclear spins to follow, but still much slower than phonons

This frequency window we call the coherence window- note that typically

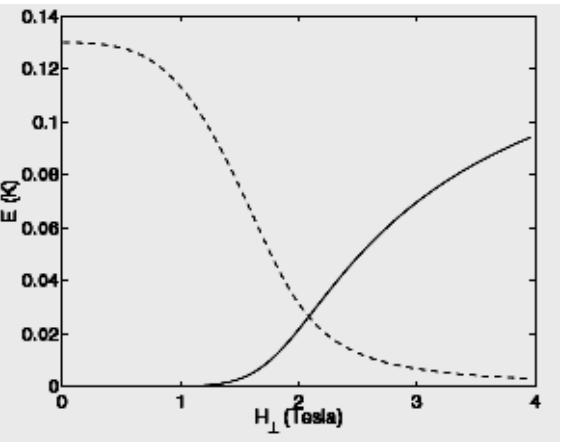
$$E_o/\theta_D \lesssim 10^{-4}$$

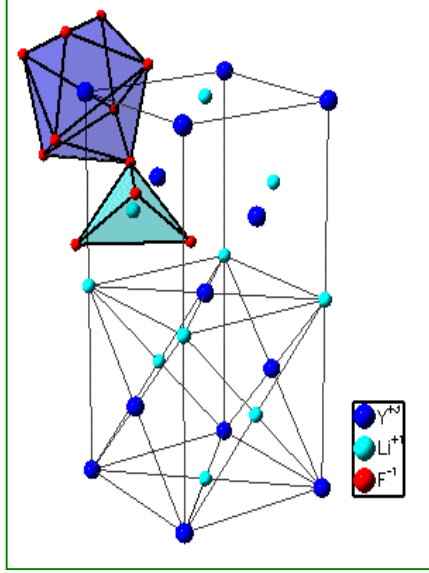
NUCLEAR SPIN BATH in MAGNETIC SYSTEMS: The $LiHo_xY_{1-x}F_4$ system



This system is usually treated as the archetypal Quantum Ising system:

$$H = -\sum_{i,j} V_{ij}^{zz}\tau_i^z\tau_j^z - \Delta_0(H_\perp)\sum_i \tau_i^x$$



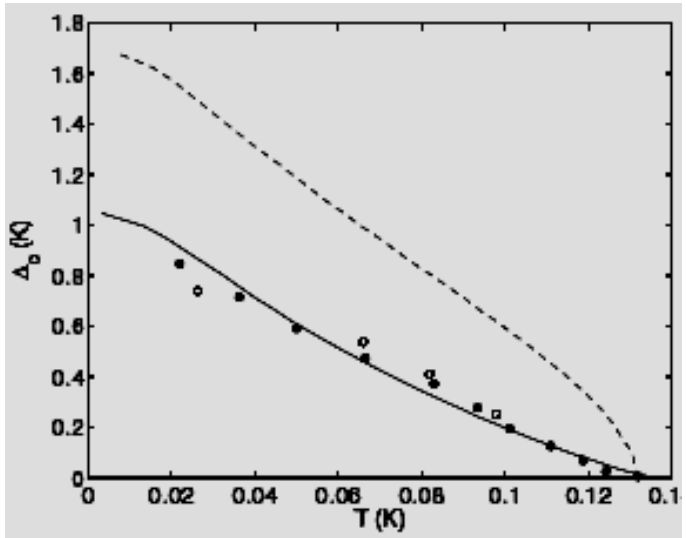


However the Ho nuclear spin actually plays a profound role in the physics:

(1) It blocks transitions until we get to very high fields (see left)

(2) The only way to understand the quantum spin glass phase is by incorporating the nuclear spins (and also the transverse dipolar terms); see below right

(3) The decoherence is completely governed by the nuclear spins down to the lowest temperatures (phonon effects disappear below roughly 250 mK)



Stamp, P.C.E., Tupitsyn, I.S., Phys Rev **B69**, 014401 (2004)

M Schechter, PCE Stamp, PRL **95**, 267208 (2005)

AND TO BE PUBLISHED

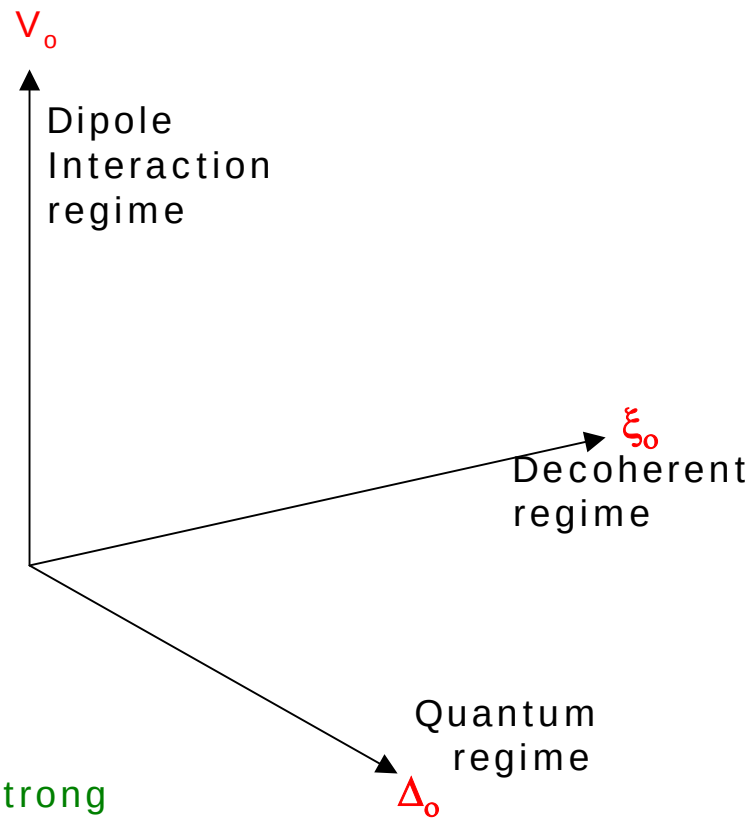
2.4: DYNAMICS of an *INTERACTING SPIN NET* coupled to an ENVIRONMENT

Until now no experiments have been performed on single magnetic qubits – they are too small to observe directly (this may well soon change). However many experiments have been done on spin nets of magnetic molecules, where the interactions tend to be dipolar. As emphasized in Christou's lectures, the great advantage one has in experiments on interacting magnetic molecules is the extreme precision with which the parameters are known and can be controlled.

Until the end of the 1990's experiments were being interpreted entirely incorrectly, in terms of non-interacting molecules – the fact that the magnetic relaxation actually looked more like that of a spin glass was ignored! However one can give a more or less complete analysis of the dynamics of the quantum relaxation regime – this was done in the period 1993-1998. Since then the quantum relaxation experiments have yielded much information on the way in which the nuclear spins, the phonons, and the dipolar interactions control the dynamics.

The coherent quantum regime, where the dynamics is controlled by the interplay between dipolar interactions and the quantum parameter Δ_o , is much less well understood. This is the quantum Ising model, but with an environment; & remember that we are not interested in simple properties like the phase diagram, but in the FULL DYNAMICS, including the N-qubit entanglement dynamics.

Different Regimes for the Spin Net System



In contrast to the single qubit problem, where we only had 2 low-T regimes (either coherent quantum or incoherent quantum relaxation), the spin net offers a range of possibilities:

(i) DIPOLE INTERACTION-DOMINATED REGIME: If one ignores the environment, this Quantum Ising system simply localises into a glass if $V_o > \Delta_o$. However the environment has a profound effect - even extremely small ξ_o will delocalise the spins, & give quantum relaxation. If we increase the quantum parameter so that $\Delta_o >> \xi_o$ (but still $\Delta_o < V_o$) then very complex multi-spin entangled dynamics ensues.

(ii) DECOHERENT RELAXATION REGIME: Even with strong decoherence/dissipation, the inter-spin correlations strongly affect the relaxational dynamics. Again, the system is never frozen, even if $\Delta_o << \xi_o$.

(iii) COHERENT QUANTUM REGIME: This is the most interesting but the most difficult to understand – we are dealing with the full quantum computation problem, with N-spin entanglement on the table. The smallest environmental coupling eventually destroys coherent dynamics – higher spin entanglement is the first to go. Many features of the dynamics here are not understood at all – this is a frontier problem of great importance. It is commonly assumed in the quantum information literature that for weak decoherence one can ignore all but uncorrelated errors (ie., single-spin decoherence coming from Interactions between individual qubits & the environment). As we shall see below this is not in general correct.

QUANTUM RELAXATION REGIME: Derivation of Kinetic Eqtn.

In both the dipolar-dominated regime and the environment-dominated regime, the dynamics is incoherent if Δ_o is small. The we can use a classical kinetic equation.

The kinetic eqtn for the magnetic qubit distribution $P_\alpha(\xi, r)$ is a BBGKY one, coupling it to the 2-qubit distribution P_2. Here r is the position of the qubit, $\alpha = +,-$ is the polarisation of the qubit along the z-axis, and ξ is the longitudinal field at r.

$$\dot{P}_\alpha(\xi,\vec{r}) = -\tau_N^{-1}(\xi)[P_\alpha(\xi,\vec{r}) - P_{-\alpha}(\xi,\vec{r})] - \sum_{\alpha'}\int\frac{d\vec{r}'}{\Omega_0}\int\frac{d\xi'}{\tau_N(\xi')}\left[P^{(2)}_{\alpha\alpha'}(\xi,\xi';\vec{r},\vec{r}') - P^{(2)}_{\alpha\alpha'}(\xi-\alpha\alpha'\tilde{U}(\vec{r}-\vec{r}'),\xi';\vec{r},\vec{r}')\right]$$

In this kinetic equation the interaction U(r-r') is dipolar, and the relaxation rate τ^{-1}_N is the inelastic, nuclear spin-mediated, single qubit tunneling flip rate, as a function of the local bias field. As discussed before, this relaxation operates over a large bias range ξ_o where typically $\xi_o \sim E_o$ (and E_o is the width of the nuclear spin muliplet introduced before)

$$\tilde{U}^{zz}_{ij} = \sum_{\mu_i\nu_j}\gamma_\mu\gamma_\nu\left[\frac{\mathbf{s}_{\mu_i}\cdot\mathbf{s}_{\nu_j}}{|\mathbf{r}_{\mu_i}-\mathbf{r}_{\nu_j}|^3} - 3\frac{(\mathbf{r}_{\mu_i}-\mathbf{r}_{\nu_j})\cdot\mathbf{s}_{\mu_i}(\mathbf{r}-\mathbf{r}_k)\cdot\mathbf{s}_{\nu_j}}{|\mathbf{r}_{\mu_i}-\mathbf{r}_{\nu_j}|^5}\right]$$

$$\tau_N^{-1}(\xi) \approx \tau_0^{-1}e^{-|\xi|/\xi_o}.$$

$$\tau_N^{-1}(\xi=0) \equiv \tau_0^{-1} \approx \frac{2\Delta_{10}^2}{\pi^{1/2}\Gamma_2}.$$

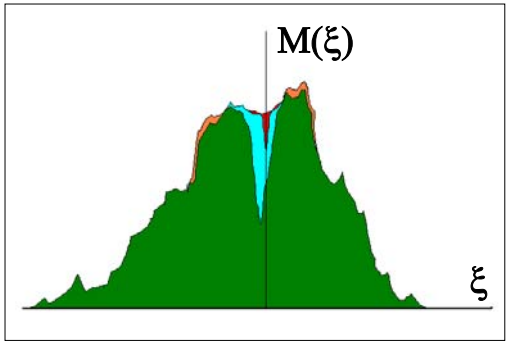
The BBGKY hierarchy can be truncated with the kinetic equation above if the initial 2-qubit distribution factorizes. This happens if the system is either (i) initially polarized, or (ii) initially strongly annealed. Then we have:

$$P_{\alpha\alpha'}(\xi,\xi',r,r';t) = P_\alpha(\xi,r,t)P_{\alpha'}(\xi',r',t) \text{ at } t=0$$

The kinetic equation can then be solved, and gives the square root short-time behaviour:

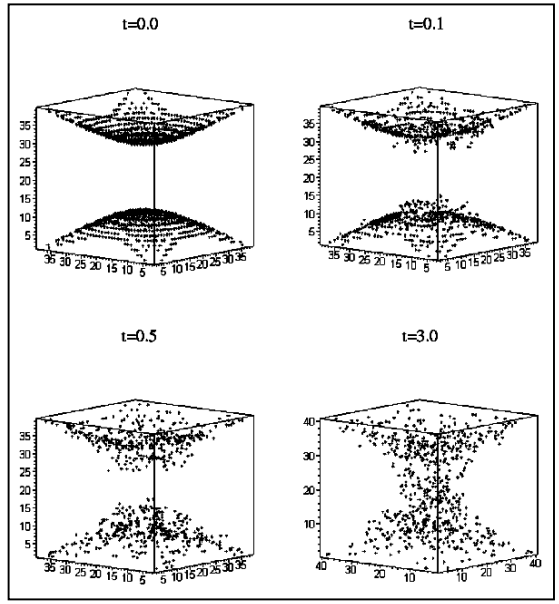
$$M(t) = M_o[1-(t/\tau_Q)^{1/2}]$$

$$\Gamma_{sqrt} \equiv \tau_Q^{-1} \sim (\Delta_o^2/V_o)\xi_o N(\epsilon_o)$$



Quantum Relaxation in a "Spin Net" of Interacting MAGBITS

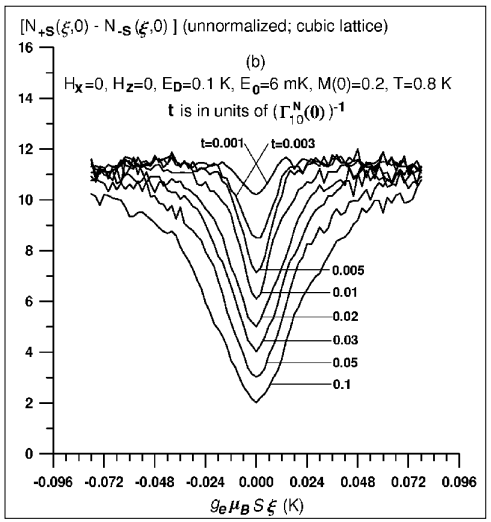
$$V_{ij} >> E_o > \Delta$$



IS Tupitsyn, PCE Stamp

At first glance the problem of a whole net of magbits, with long-range "frustrating" dipole interactions between them, looks insuperable. But actually the short-time dynamics can be solved analytically, in the quantum relaxation regime! This is because the dipole fields around the sample vary slowly in time compared to the fluctuating hyperfine fields. This leads to universal analytic predictions:

NV Prokof'ev, PCE Stamp, PRL 80, 5794 (1998)



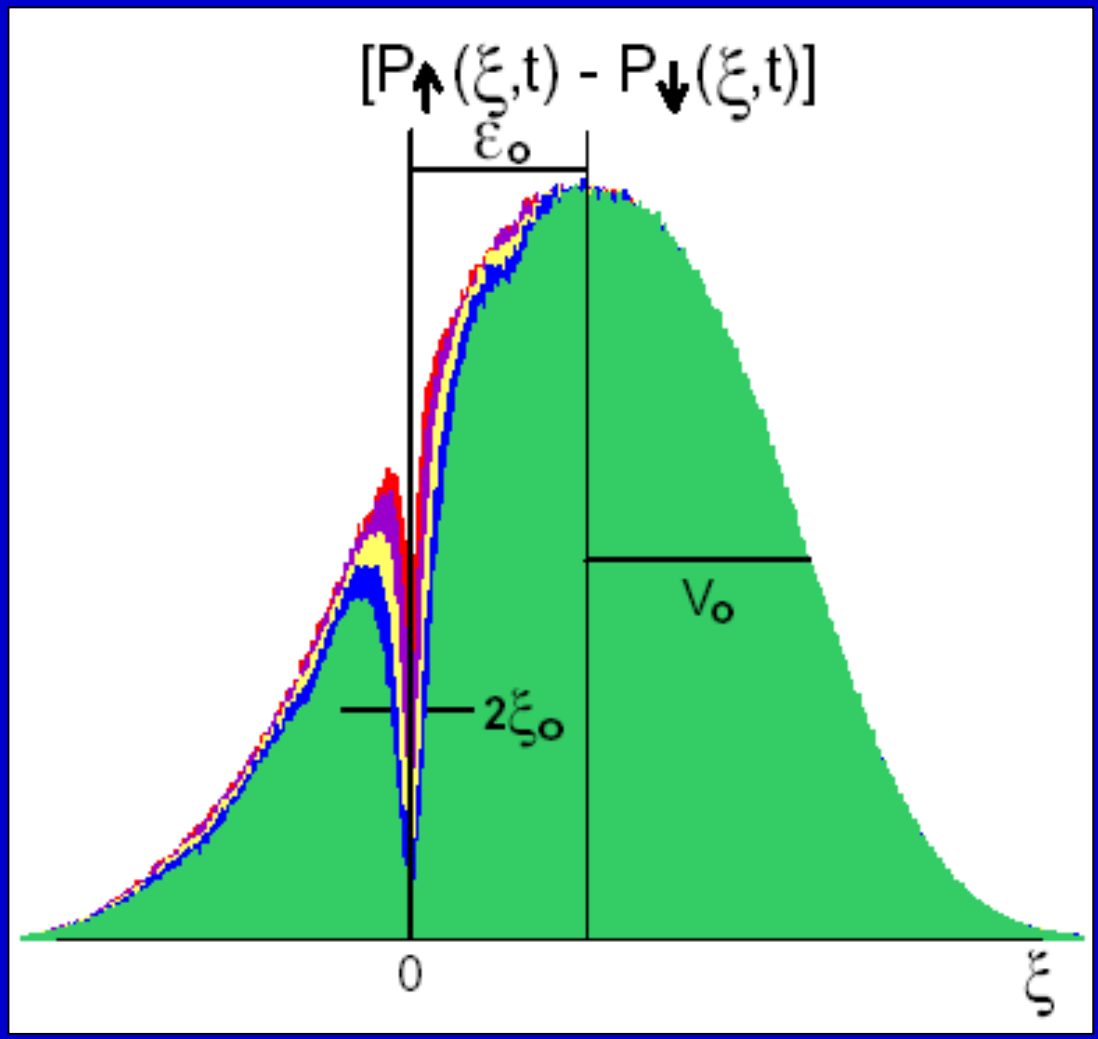
(1) Only magbits near resonance make incoherent flips
As tunneling occurs, the resonant surfaces move & disintegrate- then, for ANY sample shape

$$\delta M(t) \sim [t/\tau_Q]^{1/2} \qquad \tau_Q \sim (\Delta^2 T_2)\, E_o^{\,2} N(\xi=\varepsilon_H)/W$$

where W is the width of the dipolar field distribution, and $N(\xi)$ is the density of the distribution over bias.

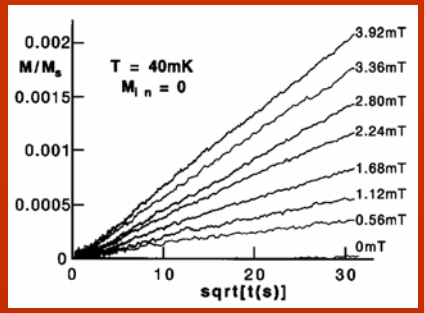
(2) Tunneling digs a "hole" in this distribution, with initial width E_o, and a characteristic spreading with time- so it depends again on the nuclear hyperfine couplings.

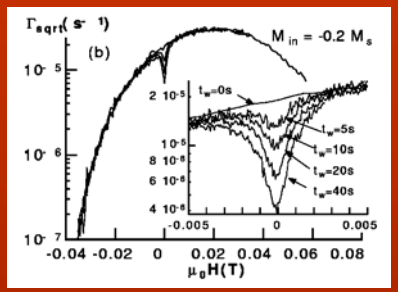
HOLE DIGGING up close

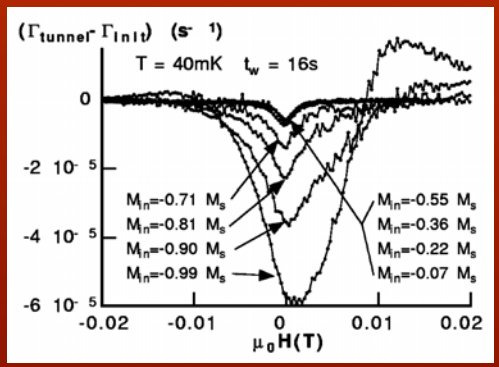


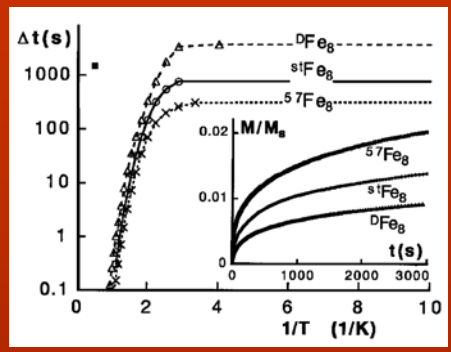
We look at the time evolution of the INTERNAL DISTRIBUTION OF BIAS FIELDS $M(\xi,t)$ (recall that ξ is the longitudinal bias field. A key feature of the theory is 'Hole-digging' in this distribution; the tunneling spins deplete the distribution. Only spins in resonance can tunnel, and this happens in a field range $2\xi_0$ (ie., controlled by the nuclear hyperfine interactions). The time evolution is non-trivial because the dipolar interactions scatter spins back into the hole (giving the square root time relaxation).

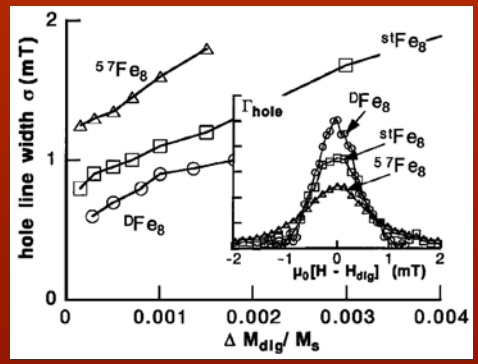
Quantum Relaxation Experiments in Magnetic Molecules

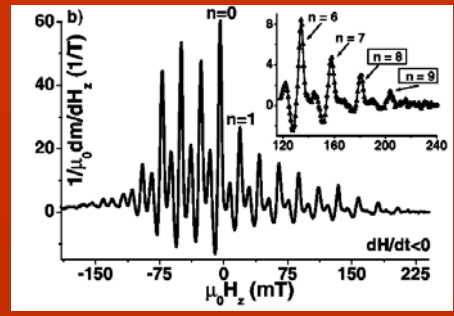






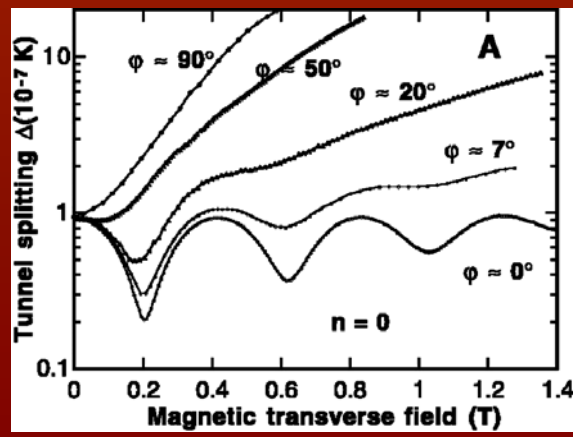






R. Giraud et al., PRL 87, 057203 (2001)

In the rare earth compound $LiHo_xY_{1-x}F_4$ the hyperfine coupling on the Ho sites is so strong that one sees the hyperfine structure directly in the quantum relaxation rate. For dilute spins (x <<1) one looks at the relaxation of individual Ho ions- and sees the nuclear multiplet structure directly!



Experiments by 4 different groups have verified all these predictions. These results on Fe-8 (of the Wernsdorfer group) show the square root relaxation, and the hole digging in the internal field distribution, Inferred from the square root relaxation rate. The hole width, and its variation with isotopes, agrees with theory. Finally, the relaxation rate oscillates with transverse field as expected for the Aharonov-Bohm oscillations in spin space.

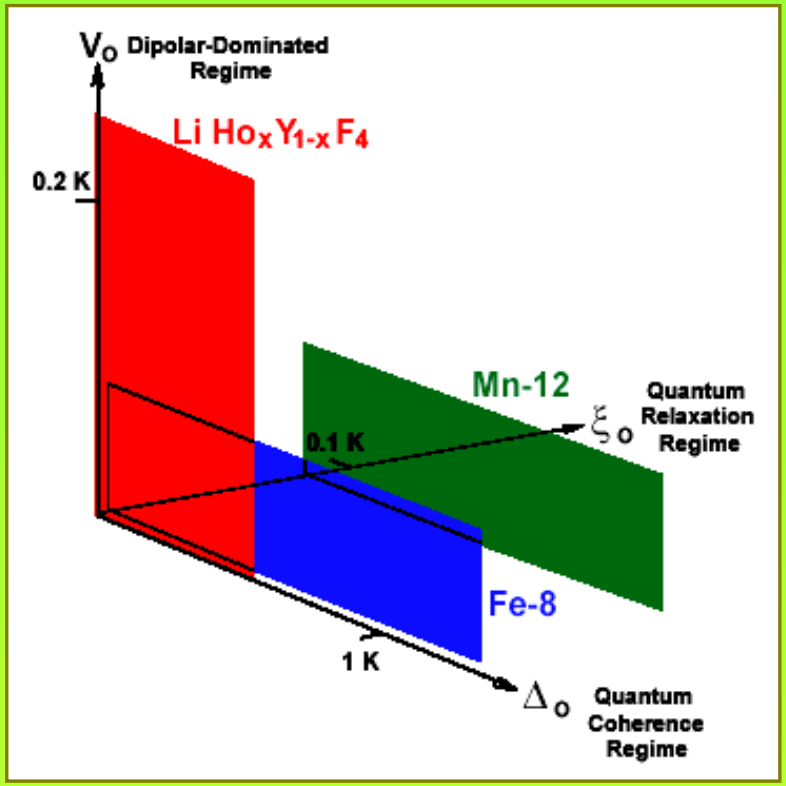
Wernsdorfer et al, PRL 82, 3903 (1999); and
PRL 84, 2965 (2000); and
Science 284, 133 (1999)

COHERENT DYNAMICS of the DIPOLAR SPIN NET

The dipolar spin net is of great interest to solid-state theorists because it represents the behaviour of a large class of systems with "frustrating" interactions (spin glasses, ordinary dipolarglasses). It is also a fascinating toy model for quantum computation:

$$H = \Sigma_j (\Delta_j \tau_j^x + \varepsilon_j \tau_j^z) + \Sigma_{ij} V_{ij}^{dip} \tau_i^z \tau_j^z + H_{NN}(I_k) + H_\phi(x_q) + \text{interactions}$$

For magnetic systems this leads to the picture at right.



Almost all experiments so far have been done in the region where Δ_o is small- whether the dynamics is dipolar-dominated or single molecule, it is INCOHERENT QUANTUM RELAXATION.

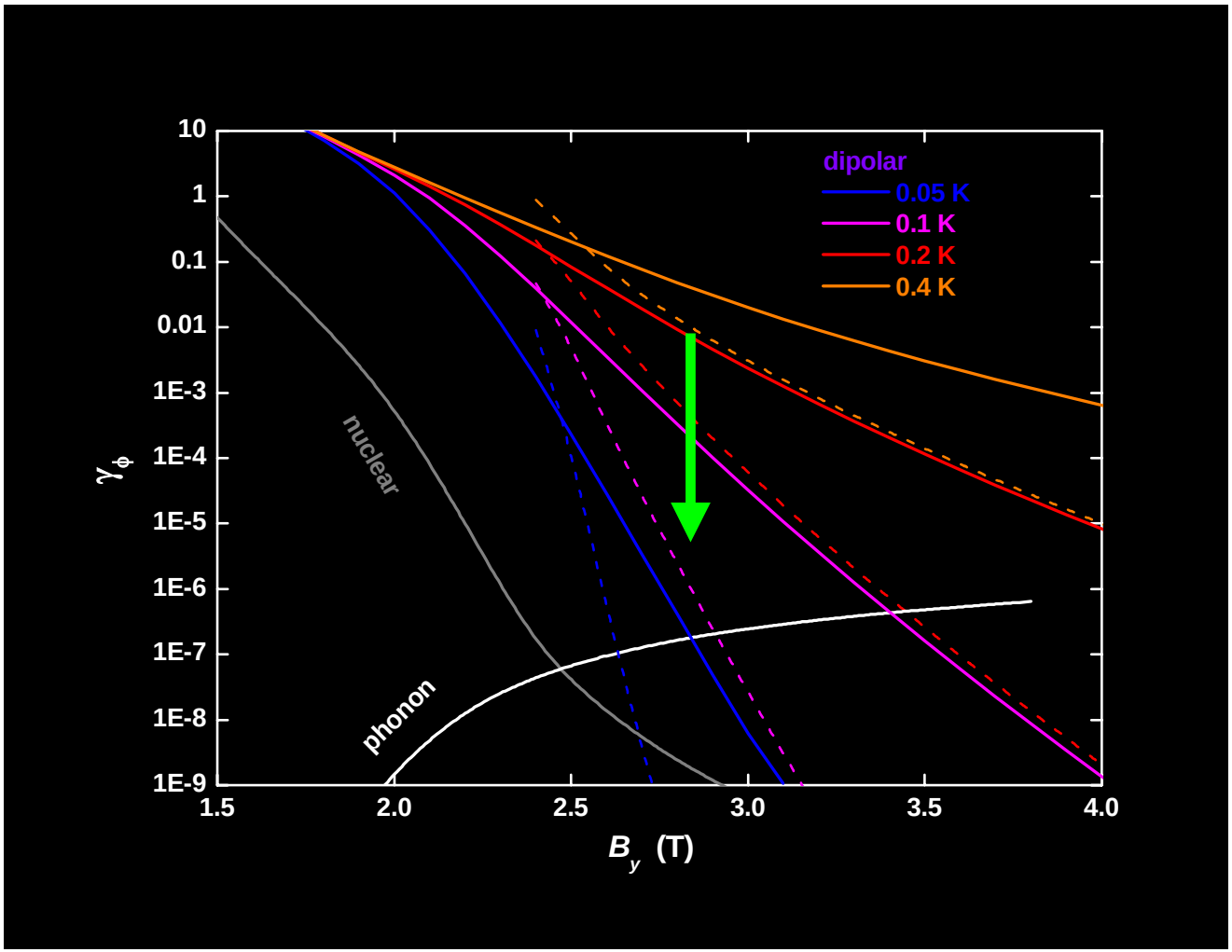
The next great challenge is the dynamics in the QUANTUM COHERENCE REGIME

Actually this is terribly complex, because we have to deal with entanglement between qubits up to the N-qubit density matrix. A lot of what has been written in the literature on this is just completely wrong.

Here I just describe the results for the single example of the Fe-8 system. Here one finds that the most important source of decoherence arises from CORRELATED ERRORS COMING FROM THE INTER-QUBIT INTERACTIONS.

RESULTS for DECOHERENCE in the Fe-8 SPIN NET

A very startling result emerges when one looks at the low-T decoherence in a dipolar spin net. Even for rather low T, te decoherence is dominated by correlated errors (ie., coming from pairs of qubits). This runs contrary to all the quantum information dogma.



Here we see results for the Fe-8 system. Note that at low T we can still get very high coherence:

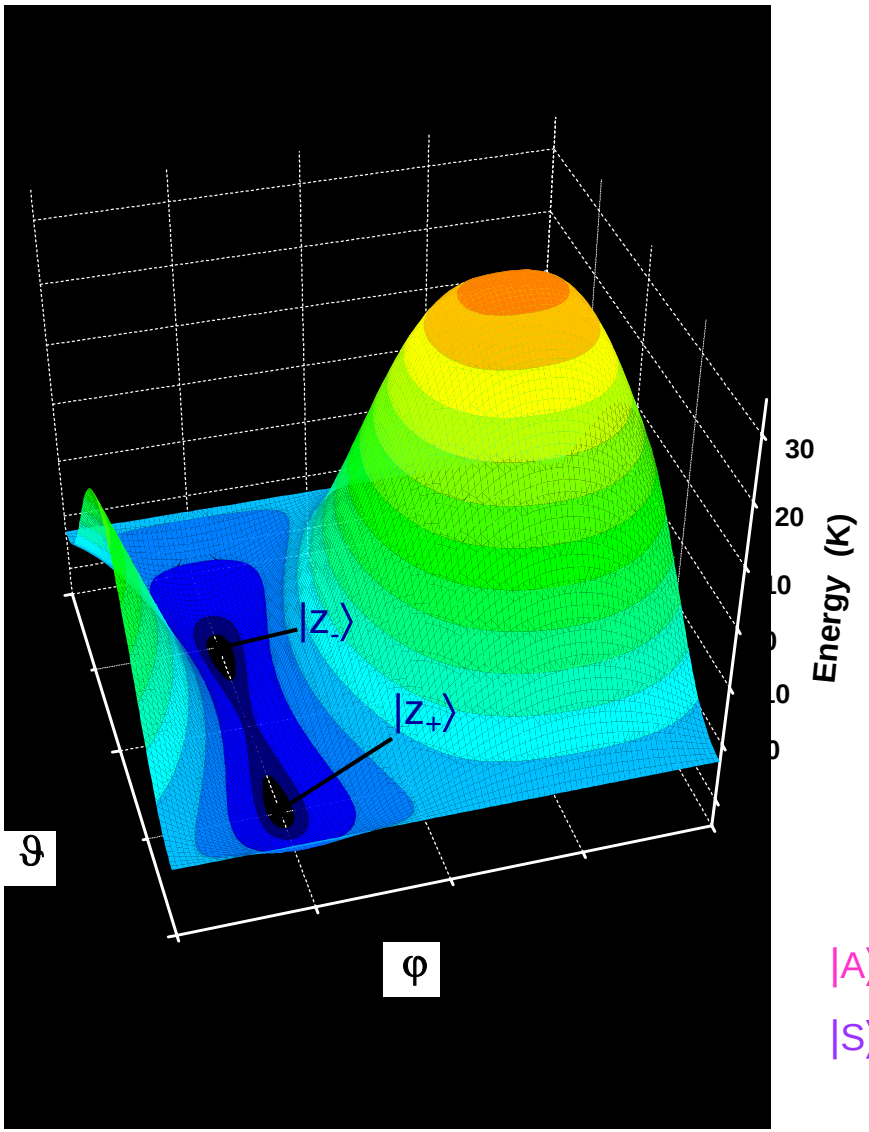
optimal coherent operation point at T = 50 mK

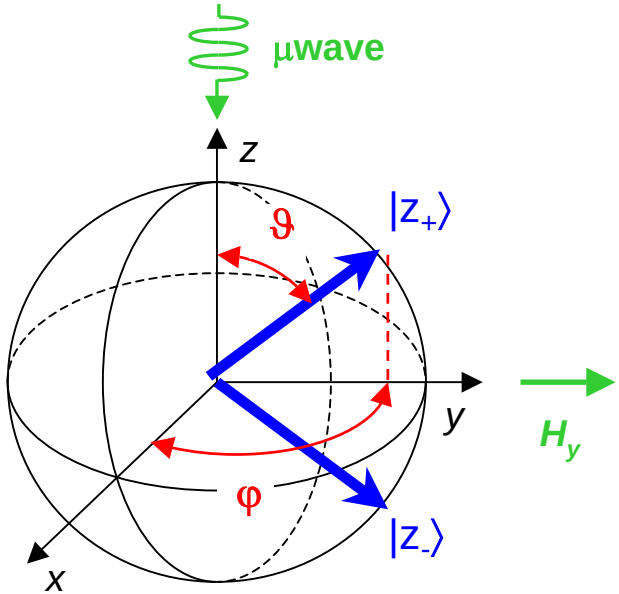
$Q \sim 10^7$

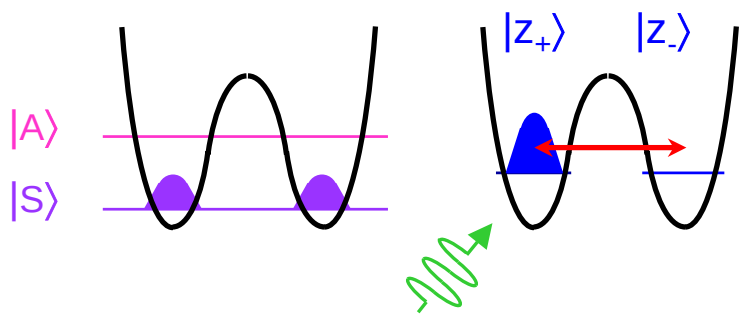
A. Morello, P.C.E. Stamp and I.S. Tupitsyn, cond-mat/0605709 (2006)

PROPOSED EXPERIMENT to observe COHERENCE in Fe-8

For details see the paper (ref below).







A. Morello, P.C.E. Stamp and I.S. Tupitsyn, cond-mat/0605709 (2006)