Decoherence has emerged as a major challenge, both for fundamental physics, and for attempts to make solid-state qubits \[\text{1}\]. Experimental decoherence rates are often much larger than theoretical estimates \[\text{2, 3}\] in complex systems like conductors or superconductors, many low-energy excitations apart from the usual “oscillator bath” modes \[\text{4, 5}\] can cause phase decoherence. These include charged defects, dislocations, paramagnetic and nuclear spins, external flux and voltage noise, and junction resistance fluctuations. The advantages of solid-state systems (stable circuits, scalability, etc.) cannot be exploited until decoherence effects are suppressed.

It is often assumed that this is purely an engineering problem. However this is not true—even completely reproducible, so impurities and defects are kept to a minimum—electronic decoherence is completely absent. Here we consider a very interesting possibility, in intrinsic decoherence, and then theory is essential, to see how this can be quantified and controlled. We define the states \(|\uparrow\rangle, |\downarrow\rangle\) (eigenstates of \(\hat{S}_z\)) by \(|\pm\rangle = (|\uparrow\rangle \pm |\downarrow\rangle)/\sqrt{2}\). If the total nanomagnetic spin \(\mathbf{S}\) is not too small, these states correspond roughly to semiclassical spin coherent states \[\text{5}\], having orientations \(\mathbf{n}_\alpha\) (here \(\sigma = \uparrow, \downarrow\)), which depend on both the internal anisotropy field of the nanomagnet, and any transverse external field \(\mathbf{H}_z\). The splitting \(\Delta_\alpha\) depends sensitively on \(\mathbf{H}_z\).

The intrinsic decoherence in insulating nanomagnets comes from entanglement of the nanomagnetic spin wave function with that of the nuclear spins and phonons \[\text{6}\]. Both of these couplings are well understood. The nuclear spins \(\mathbf{I}_k\) couple to the electronic spins \(\{\mathbf{s}_j\}\) in \(\mathbf{S}\) via individual hyperfine couplings \(A_{\alpha\beta}^{jk} s_j^\alpha I_k^\beta\), and the phonons couple to \(\mathbf{S}\) via magnetoacoustic (spin-phonon) interactions \[\text{7}\]. Without yet specifying the precise form of the hyperfine couplings (which of course depend on the system being studied) we can define quite generally a vector \(\varepsilon_k^{\parallel}\) which specifies the net effect on the \(k\)-th nuclear spin \(\mathbf{I}_k\), of all the hyperfine fields coming from the individual electronic spins in \(\mathbf{S}\):

\[
\varepsilon_k^{\parallel} \equiv \omega_k^{\parallel} \tau_k = \frac{1}{2} \sum_j A_{\alpha\beta}^{jk} (\langle s_j^\alpha \rangle \tau^\beta - \langle s_j^\alpha \rangle \tau^\beta) \quad (1)
\]

Here \(\hat{\mathbf{I}}_k\) is a unit vector in the direction of this total field, with components \(\hat{I}_k^\alpha\), and \(\langle s_j^\alpha \rangle\) is the expectation value of \(s_j^\alpha\) when \(\mathbf{S} \rightarrow \mathbf{S} \mathbf{n}\). We see that when \(\mathbf{S}\) flips from \(\mathbf{S}_1\) to \(\mathbf{S}_2\), the energy change is just \(2\omega_k^{\parallel} \hat{\mathbf{I}}_k \cdot \mathbf{I}_k\), i.e., we have a diagonal coupling \(\hat{\mathbf{I}}_k \cdot \mathbf{I}_k\) between the qubit and \(\mathbf{I}_k\). One can also write this coupling as \(\hat{\mathbf{I}}_k \cdot \mathbf{I}_k\), with components \(\xi_\alpha = \omega_k^{\parallel} \hat{\mathbf{I}}_k \cdot \mathbf{I}_k\). Note that the weak interaction between nuclear spins (typically dipolar) causes slow spin...
diffusion between them. The net effect of this is to make \( \xi_x \rightarrow \xi_z(t) \), i.e., the nuclear bias field acting on the qubit fluctuates in time even when the qubit is frozen. We shall argue below that we can neglect this fluctuation in strong transverse fields.

The \( I_k \) also couple to the external field \( H_o \), with Zeeman coupling \( \omega^z_k \hat{m}_k \cdot \hat{I}_k \), where

\[
\omega^z_k \hat{m}_k = g_k^N \mu_N H_o
\]

(2)

and \( \hat{m}_k \) is a unit vector along \( H_o \).

If we now take these terms together, we can write the interaction Hamiltonian between the nanospin \( S \) and the nuclear spins \( \{ I_k \} \) in the form \( H_{NS} = H_o(\chi) + V(\chi, I_k) \), where

\[
V = \tau_x \sum_k \omega^z_k I_k \cdot \hat{I}_k + \sum_k \omega^z_k \hat{m}_k \cdot \hat{I}_k
\]

(3)

This form is particularly useful for quantifying the decoherence from the nuclear spins. When we come to particular examples we will specify the couplings in \( [13] \).

The magnetoacoustic interaction between the qubit coordinate \( \tau \) and the phonon coordinate \( x_q \) is dominated in nanomagnets \( [10, 11] \) by a non-diagonal term \( \tau_x \sum_q c_q x_q \). This term has strength

\[
c_q x_q \sim S \Omega_o(\omega_q/\theta_D)^{1/2}, \tag{4}
\]

where \( \theta_D \) is the Debye energy, \( \omega_q = q c_s \) and \( c_s \) is the sound velocity.

Our basic idea is as follows. The allowed nuclear spin bath states, when the qubit is in some given state, have a density of states which typically has Gaussian lineshape \( [11] \), with a halfwidth \( E_o \); in terms of the \( \{ \omega^z_k \} \) defined above, \( E_o \) is given trivially by

\[
E_o^2 = \sum_k \frac{I_k + 1}{3I_k} (\omega^z_k I_k)^2 \tag{5}
\]

On the other hand the acoustic phonon energy scale is the Debye energy \( \theta_D \). Now in a nanomagnetic system \( E_o/\theta_D \) can be \( \lesssim 10^{-4} \), suggesting the following tactic for suppressing decoherence. If we tune \( \Delta_o \), so that \( \theta_D \gg \Delta_o \gg E_o \), then we will be in a "coherence window", in which decoherence will be at a minimum because the qubit dynamics is too slow to disturb most phonons, but too fast for the nuclear spins to react.

Decoherence Rates: To substantiate this idea, we generalise the low field (\( \Delta_o < E_o \)) calculations of nanomagnetic dynamics \( [10] \), where incoherent tunneling relaxation is found, to the high-field regime \( \Delta_o \gg E_o \). Because at high field the couplings \( \{ \omega^z_k \} \) of the nuclear spins to the qubit are \( \ll \) the nuclear Zeeman couplings \( \{ \omega^z_k \} \), and also \( \{ \omega^z_k \} \ll \Delta_o \), this dynamics can be solved perturbatively \( [12, 13] \). We expand about the bare qubit Hamiltonian \( [3] \) to 2nd order in \( \omega^z_k/\Delta \) (assuming \( \epsilon_o = 0 \) for simplicity) to get

\[
H_{NS} = [\Delta_o \chi_x + \sum_k \omega^z_k \hat{m}_k \cdot \hat{I}_k]
\]

\[
+ \tau_x \sum_{kk'} \frac{\omega^z_k \omega^z_{k'}}{2\Delta_o} (\hat{I}_k \cdot \hat{I}_{k'})
\]

\[
+ O((\omega^z_k)^2/\Delta_o^3) \tag{6}
\]

The decoherence time \( \tau_o \) is defined as the characteristic time for decay of the off-diagonal density matrix element, starting in state \( |\uparrow\rangle \). In the present case we calculate this matrix element as a path integral over pairs of qubit trajectories \( \tau_z(t), \tau_z(t') \) (each taking values \( \pm 1 \), with occasional flips between these values), weighted by an influence functional \( F[\tau_z(t), \tau_z(t')] \) which incorporates the interactions \( [3] \). The contribution from the 2nd term in \( [3] \) to this functional is \( [12, 13] \):

\[
\ln F = - \sum_k \frac{(\omega^z_k)^2}{8\hbar^2} \int_0^t ds dse^{i\omega_s s} [\tau_z(s) - \tau_z'(s)]^2 \tag{7}
\]

From this result we can then use standard techniques developed for the spin-boson model \( [3] \), to find a contribution \( \gamma_o^c \) to the dimensionless decoherence rate \( \gamma_o = 1/\tau_o \Delta_o \) (the commonly used 'decoherence quality factor' \( [3] \)) is just \( Q_o = \pi/\gamma_o \). We find

\[
\gamma_o^c = \sum_{kk'} \left( \frac{\omega^z_k}{\omega^z_{k'}} \right) \left( \frac{I_k + 1}{I_{k'}} \right) \left( \frac{I_{k'} + 1}{I_k} \right) \omega^z_k \omega^z_{k'} \frac{1}{2\Delta_o^2} \tag{8}
\]

which decreases rapidly with increasing qubit operating frequency \( \Delta_o \).

There are 2 other contributions to \( \gamma_o \) coming from the nuclear spins \( [10, 13] \). First, in writing \( [3] \) we omitted a renormalisation of the tunneling matrix element caused by the coupling to the nuclear spins \( [14] \), which in fact describes the nuclear spin transitions induced directly by electronic spin flips. This adds a contribution \( \gamma_o^c \) to \( \gamma_o \), given by \( \gamma_o^c = \frac{1}{2} \sum_k |\vec{\alpha}_k|^2 \), where \( |\vec{\alpha}_k| = \pi/\omega^z_k/2\Omega_o \). However when \( \Delta_o \gg E_o \), the ratio \( \gamma_o^c/\gamma_o \sim O(\Delta_o^2/\Omega_o^2) \ll 1 \) (the usual WKB reduction of the tunneling amplitude), i.e., this term can always be neglected to first approximation. Second, we have neglected the intrinsic nuclear spin diffusion dynamics, caused by internuclear interactions. In low fields, when the tunneling is slow, and \( \Delta_o \ll E_o \), this intrinsic nuclear dynamics renders the tunneling dynamics incoherent \( [10, 12, 13, 21] \) (the usual WKB reduction of the tunneling amplitude). However when \( \Delta_o \gg E_o \), the nuclear fluctuations are very slow compared to the qubit dynamics (of frequency \( \Delta_o \)); they then add a "noise" contribution \( \gamma_o^N = N/\pi \Delta_o \tau_2 \) to \( \gamma_o \), where \( N \) is the number of nuclear spins in each molecule.
Typically $T_s^{-1} \sim 10 - 100 \, \text{Hz}$ at low $T$ in magnetic molecules (17), whereas we are interested in $\Delta_o \sim G \, \text{Hz}$ (see below); thus $\gamma_o^N$ will be very small.

Finally we include the phonon contribution to $\gamma_\phi$. The solution to the spin-boson problem for non-diagonal coupling to phonons gives a contribution $\gamma_\phi^{ph}$ of form (10):

$$
\gamma_\phi^{ph} = \left[(\Omega_o \Delta_o)^2 / \theta_B^2 \right] \coth(\Delta_o / k_B T)
$$

which increases rapidly with $\Delta_o$.

The phonon and nuclear spin decoherence mechanisms act independently- thus we can get a simple estimate for the optimal decoherence rate $\gamma_\phi^{\text{min}}$ by summing the two dominant contributions (5) and (6), and minimizing their sum $\gamma_\phi + \gamma_\phi^{ph}$ with respect to $\Delta_o$, assuming $k_B T < \Delta_o$. This gives

$$
\gamma_\phi^{\text{min}} \approx \sqrt{2} \Omega_o E_o / \theta_B^2
$$

at an optimal tunneling splitting $\Delta_o^{\text{opt}}$:

$$
\Delta_o^{\text{opt}} \approx \theta_B (E_o / \sqrt{2} \Omega_o)^{1/2}.
$$

We see that decoherence is minimized for a given $S$ by making $E_o$ and $\Omega_o$ small, and $\theta_B$ large, within the constraint that $\Omega_o >> \Delta_o > k_B T$. If $k_B T > \Delta_o$ we get a different (less favorable) answer.

These simple results actually give reasonably accurate results when compared with numerical calculations on real systems, as we now see.

2. Three Examples: We present quantitative results for the decoherence rates in 3 different materials. We give most details for the Fe-8 example, to illustrate our method.

(i) The Fe-8 molecule: This well-characterized molecule behaves below $\sim 10 \, \text{K}$ as an electronic spin-10 system, with biaxial effective Hamiltonian $\mathcal{H}_o(S) \sim -D S_z^2 + E S_x^2 + K_4 (S_x^4 + S_y^4) - g_\mu_B H_{\perp} \cdot S$, with $D/k_B = 0.23 \, \text{K}$, $E/k_B = 0.094 \, \text{K}$, and $K_4/k_B = -3.28 \times 10^{-5} \, \text{K}$. The small oscillation frequency $\Omega_o$ is $2SC_4 \sqrt{DE} \sim 4.6 \, \text{K}$, where $C_4 = 1.56$ includes the effects of the $K_4$ term; $T$-independent tunneling dynamics appears below $\sim 0.4 \, \text{K}$. The tunneling amplitude $\Delta_o(H_{\perp})$ is trivially determined by diagonalisation of $\mathcal{H}_o(S)$ (Fig. 1), as are the spin orientations $n^1, n^4$ defined previously. At a critical transverse field $H_{\perp}$ the barrier is destroyed and $n^1, n^4$ merge. When $H_{\perp} = 0$ is along the hard $\bar{z}$-axis (so the azimuthal angle $\phi = 0$ or $\pi$), one has $g_\mu_B H_{\perp} = 2S(D + E)$ for this Hamiltonian, giving $H_{\perp} \sim 4.8 \, \text{T}$. We find by comparing exact diagonalisation with semiclassical calculations, that the latter are accurate up to $H_{\perp} \sim 3.3 - 4.3 \, \text{T}$ (for $\phi = 90^\circ, 0^\circ$ respectively).

The hyperfine interactions between the 8 $^{57}Fe$ ions and the 205 nuclear spins in the molecule (213 if $^{57}Fe$ isotopes are substituted for $^{56}Fe$ nuclei) are of 2 kinds. The $Fe$ electronic spins interact with any $^{57}Fe$ ions via contact hyperfine interactions, which we assume to be the same as for $^{57}Fe$ ions in similar materials (24). On the other hand the hyperfine couplings $A_{jk}^{z} \gamma_o I_j^{z}$ to all the other nuclear spins are thought to be dominated by purely dipolar terms (16).

Using the known nuclear positions and moments, the hyperfine interactions and the nuclear Zeeman couplings are then quantified numerically, to find the $\{\omega_k^\perp\}$ and hence $E_o$ (see Fig. 2), and also the $\{\omega_k^\perp\}$. In spite of the large number of protons, $E_o$ is quite small (particularly when we substitute $^2H$ nuclei for $^1H$), making Fe-8 a reasonable candidate for coherent dynamics in high fields. One may also calculate the nuclear dynamics (16), but here we simply note that NMR experiments (17) indicate that at low $T$, $T_1 \sim 10 - 30 \, \text{msec}$ for $^1H$ nuclei in magnetic molecules. We then calculate numerically the different nuclear spin contributions to $\gamma_\phi$ (i.e., the dominant contribution $\gamma_k^\perp$, plus the smaller contributions $\gamma_k^\parallel$ and $\gamma_k^N$), and also the phonon contribution $\gamma_\phi^{ph}$, as a function of $H_{\perp}$.

The results are shown for high fields in Fig. 3. Actually almost all experiments on the quantum dynamics of Fe-8 have been done in the regime $\Delta_o < E_o$, i.e., where we expect nuclear spins to cause incoherent tunneling - which is what is found experimentally (16, 17). However once $\Delta_o$ exceeds $E_o$, all nuclear spin decoherence should fall off very fast. The nuclear noise contribution $\gamma_k^N < 10^{-7}$ once $|H_{\perp}^x| > 3.2 \, \text{T}$ (for $\phi = 0^\circ$) or $> 2 \, \text{T}$ (for $\phi > 90^\circ$), so we can safely ignore it. The estimate then gives $\gamma_\phi^{\text{min}} \approx 5.8 \times 10^{-5}$ at $\Delta_o^{\text{opt}} \approx 0.14 \, \text{K}$, when $\phi = 0$ and $H_{\perp}^x = 3.45 \, \text{T}$ (assuming the optimal set of nuclear isotopes, with $^2H$ instead of $^1H$, no $^{57}Fe$, etc).

In Fig. 3 we calculate $\gamma_\phi$ numerically, adding all nuclear spin and phonon contributions. For $\phi = 0$ we get a nu-
Numerical value $\gamma_{\phi}^{\text{min}} = 6 \times 10^{-5}$, at a $\Delta \phi_{\text{opt}} = 0.135 \text{ K}$ when $H_{\phi} = 3.45 \text{ T}$; thus the estimate works well for $\phi = 0$. The further reduction of $\gamma_{\phi}^{\text{min}}$ to $2.4 \times 10^{-6}$, when $\phi \rightarrow 90^\circ$ ($H_{\phi} = 2.93 \text{ T}$), comes from decreases in both $\Omega_o$ and the effective $S$ (since both the tunneling barrier and the change $|S^\uparrow - S^\downarrow|$ decrease). Fig. 3 clearly illustrates the window for coherent dynamics which opens up at high transverse fields ($H_{\phi} \sim 2.9 - 3.4 \text{ T}$, depending on $\phi$) in Fe-8.

(iii) Ho ions: Not all systems will have an optimal intrinsic decoherence described by (10). Consider the $\text{LiHo}_2\text{Y}_{1-x}\text{F}_{3}$ system [20], in which sharp absorption lines are seen [21, 22] at low $x$. The $\text{Ho}^{+3}$ ions (with spin = 8) have a lowest doublet state, with splitting $\Delta_o \propto |H_{\phi}^{\parallel}\rangle$ in a transverse field- only one other state, at an energy $\Omega_o \sim 10.6 \text{ K}$, is important for the low-energy physics (all other electronic spins levels are at energies $\gtrsim 100 \text{ K}$). The new feature here, which renders (10) inapplicable, is a very strong hyperfine coupling $H_{\phi}^{\parallel} = 0.039 \text{ K}$ to the $I = 7/2$ Ho nuclear spin- whereas the couplings to the “satellite” Li, Y, and F nuclear spins are very weak. Thus instead of having a Gaussian line-shape, the hyperfine multiplet has a “toothcomb” structure with 15 Ho lines spaced in intervals of 0.039 K, each weakly broadened (by less than 1 mK) by the other nuclei. The quantity $E_o$ is then not well-defined [11]. However one may instead just calculate the coupled Ho-nuclear spin dynamics in 2nd-order perturbation theory, since $\Omega_o \gg$ all hyperfine couplings, and all Ho transitions go via the single intermediate level. We then find the optimal strategy is to (a) freeze the Ho nuclei by cool-
tactic reaches its limit once \( S_f \) purification of nuclear spins, our existent ones, it pays to keep \( \Delta_0 \) to 30 mK (using \( H_c^+ \sim 0.3 T \)) gives a contribution \( \gamma_{\phi}^{min} \sim 1.8 \times 10^{-5} \) from the \( Ho \) nuclei (phonons giving a contribution < 10^{-9} here). Some residual decoherence also comes from the ‘satellite’ nuclear spins- in this context it is interesting that \( Ho \) ions can be prepared in hosts with almost no other nuclear spins (eg., \( CaWO_4 \), with isotopically purified W; see [21]). Thus we can expect a reduction of \( \gamma_{\phi}^{min} \) to very low levels, limited only by a weak phonon effect, external noise, and any spin impurities.

3. Discussion: In insulating systems where nuclear spin decoherence is suppressed by a transverse field, decoherence optimisation is well described by \( \gamma_{\phi} \). This means having a small \( S \), small \( E_o \), and a “stiff” system (high \( \theta_o \)). Surprisingly it also means low \( \Omega_o \), ie., weak magnetic anisotropy. By taking all these measures, very low values of \( \gamma_{\phi} \) can be attained.

Our results are thus good news for coherence in small nanomagnets, and magnetic qubits. In most materials a correct choice of parameters requires strong transverse fields, but in systems like \( LiHo_o Y_{1-x} F_4 \) with one very strong hyperfine coupling \( \omega_0 \), and otherwise weak or non-existent ones, it pays to keep \( \Delta_0 \) low (ie., not too strong fields), and to have \( kT \ll \omega_0 \| \) (to suppress thermal nuclear spin noise).

For genuinely macroscopic superpositions of magnetic states, the result [10] is not such good news. Although one may stave off decoherence for large \( S \) by isotopic purification of nuclear spins, our \( Ni \) example shows this tactic reaches its limit once \( S \sim 10^5 - 10^6 \).

So far experiments on nanomagnets have concentrated on field ranges where \( \Delta_0 < E_o \), and so incoherent tunneling is observed- any experiments on, eg., \( Fe \)-8 in the range \( |H_c^+| \sim 3 - 4 T \) would be of great interest. In almost all experiments so far, on molecules or rare earth ions, collective tunneling caused by inter-spin dipolar interactions complicate the interpretation [21] [21] [22]. A clean observation of coherence, with a measurement of \( \gamma_{\phi} \), will thus involve manipulation at microwave frequencies, on a properly isolated single system.

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