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LETTER TO THE EDITOR

Giant spins and topological decoherence: a Hamiltonian approach

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Abstract. We study the topological decoherence, arising from environmental spins, which acts to suppress the quantum coherent behaviour of grain magnetization. It is found that this decoherence mechanism is so effective that even under the most favourable conditions, it will be very difficult to see quantum coherent behaviour of grains having a quantum number S greater than $\sim O(10)$. This conclusion does not affect the possibility of seeing tunnelling behaviour for larger grains.

1. Introduction

Despite its long history, the problem of spin tunnelling was not solved correctly until 1986 by van Hemmen and Suto [1] and Enz and Schilling [2]. Both solutions were semiclassical WKB calculations, applicable to 'giant spins', for which the spin quantum number $S \gg 1$. Such giant spins are believed to correctly describe ferromagnetic grains, and there has been great theoretical interest recently in the possible tunnelling behaviour of both grains and magnetic domain walls [3], as well as a number of experiments [4, 5].

A more delicate situation arises when either the giant spin or the domain wall is tunnelling between two degenerate states; this is the situation of 'macroscopic quantum coherence' (MQC) [6]. As noted in [1] and [2], and discussed more recently from an instanton viewpoint by a number of authors [7–9], the straightforward tunnelling of a giant spin is only possible if $S = n$, where n is an integer. In semiclassical language, tunnelling paths of opposite sense (e.g. clockwise and anticlockwise) give opposite 'topological phase' contributions $\pm i\pi S$ to the tunnelling action; in the MQC problem they interfere, so that the original 'tunnelling splitting' energy Δ_o now becomes

$$\tilde{\Delta} = \Delta_o|e^{i\pi S} + e^{-i\pi S}| = 2\Delta_o|\cos\pi S|.$$

Thus 1/2-integer giant spins ($S = n + 1/2$) do not show a MQC splitting, as one expects from Kramers' theorem [1, 2].

This immediately leads to the question of how the coupling of the giant spin to any 'environmental' spins (electronic or nuclear) will affect the coherent tunnelling. In this paper we wish to study this problem directly, starting from a microscopic Hamiltonian; it has already been analysed indirectly in [9]. We deal quantitatively with a new kind of 'topological decoherence' which is very effective in destroying MQC for giant spins; it is also of theoretical interest because it cannot be described using any kind of Caldeira–Leggett model, in common with a number of other dissipative and decoherence mechanisms operating in magnets [10, 11].

The basic idea is straightforward. If the giant spin, via its coupling to the environmental spins, causes some to flip as it tunnels, then an extra phase will be accumulated in the tunnelling action. In the strong coupling limit, where the environmental spins tunnel rigidly with the giant spin, this simply increases S to a larger effective value. However, for intermediate and weak coupling, the environmental spins only flip some of the time, and different phases are incurred each time the giant spin tunnels. This randomizes the phases (or 'winding numbers') and coherent motion is destroyed.

2. Theory

We now show how this works quantitatively. We start from a Hamiltonian

$$H = H_0(\boldsymbol{S}, \boldsymbol{H}) + \sum_{k=1}^{N} \left(H_k^{(0)}(\sigma_k, \boldsymbol{H}_0) + \gamma_k(\boldsymbol{S})\sigma \right) \tag{1}$$

in which H_0 describes the giant spin (a ferromagnetic grain, possibly in an external field $\boldsymbol{H}_0$), interacting with a set $\{\sigma_k\}$ of environmental spins. The environmental spins couple to $\boldsymbol{S}$ through the 'field' $\gamma_k(\boldsymbol{S})$, and also have their own independent dynamics, governed by the $H_k^{(0)}$. We are interested in the case of coherent tunnelling (MQC) in which H_0 has a potential with two degenerate minima: a typical example would be

$$H_0 = K_{\parallel} S_z^2 + K_{\perp} S_y^2 + \gamma \boldsymbol{H}_0 \boldsymbol{S} \tag{2}$$

where $K_{\parallel}$ and $K_{\perp}$ are anisotropy energies. The coupling $\gamma_k \sigma_k$ can be to either nuclear or electronic spins, not already included in $\boldsymbol{S}$. In general, γ_k must be an operator, but for the giant spin considered here we shall treat it as a vector—this does not affect the following [12]. In this example, $\boldsymbol{S}$ tunnels between orientations $\boldsymbol{S}_1 = (\theta = \pi/2, \phi = -\pi)$ and $\boldsymbol{S}_2 = (\theta = \pi/2, \phi = \pi)$, when $K_{\parallel} > K_{\perp} > 0$.

The Zeeman splitting $\omega_k = 2|\gamma_k|$ (for spin-1/2 environmental spins), thus varies enormously, as shown schematically in figure 1. Nuclear spins in the grain itself can have ω_k ranging from $\omega_k > 10^9$ Hz (for rare-earth hyperfine coupling) to roughly 10^6 Hz (dipole couplings); the frequencies of nuclear spins outside the grain have no lower limit (for distant nuclei), and the same is true for any paramagnetic electronic spins outside the grain (in, e.g. a substrate or surrounding dielectric). There may also b 'loose' spins, i.e. electronic spins which are only weakly coupled (by, say, superexchange or dipole interactions) to the grain magnetization—these would exist on the grain surface, and at dislocations or defects in the grain. We shall return to discuss the actual values of the $\{\omega_k\}$ below.

2.1. Single environmental spin

We start with a single spin-1/2 coupled to $\boldsymbol{S}$; for simplicity we ignore any static fields at the spin-1/2 site, to end up with a Hamiltonian $H = H_0(\boldsymbol{S}) + \boldsymbol{h}(\boldsymbol{S})\sigma$ (quite generally $\boldsymbol{h}(\boldsymbol{S}) = -\boldsymbol{h}(-\boldsymbol{S})$). Since we are only interested in the possible slow tunnelling dynamics

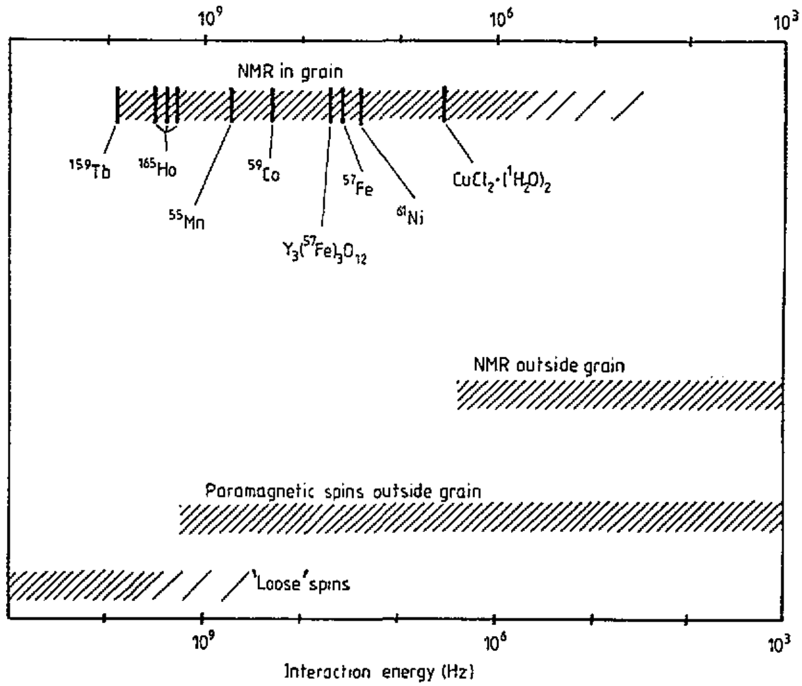

Figure 1. The range of interaction energies E_k between the environmental spin σ_k and S. A variety of nuclear spin energies are shown, for different magnetic materials. The nuclear and paramagnetic spin energies outside the grain are essentially dipole, but the 'loose spin' energies can arise from a combination of exchange, superexchange, and electron transfer energies.

of $\boldsymbol{S}$, we truncate the actual Hilbert space for the giant spin to its two lowest levels, so that $H_0(\boldsymbol{S}) \rightarrow \tilde{H}_0(\boldsymbol{S})$:

$$\tilde{H}_0 = 2\Delta_0 \cos \pi S \hat{\tau}_x \tag{3}$$

in which $\hat{\tau}$ operates on the giant spin. For $S = n + 1/2$ (i.e. 1/2-integer), the ground state is a doublet, in accordance with Kramers' theorem [7–9]. This procedure is rather standard in the non-interacting case; however here it generates new terms in the effective Hamiltonian since the environmental spin can flip during the tunnelling motion of $\boldsymbol{S}$. Now the instanton action is an operator in the subspace of the environmental spin, and must be written in general form (with $\boldsymbol{n}$, $\boldsymbol{n}'$ unit vectors) as

$$\exp\{-\hat{A}(\pm)\} = \exp\{-A_0\beta\hat{\boldsymbol{\sigma}}\boldsymbol{n}' + \delta\}\exp\{\pm\mathrm{i}(\pi S + \alpha\hat{\boldsymbol{\sigma}}\boldsymbol{n} + \phi)\}.$$

The first factor describes the effect of the environmental spin state on the potential barrier for tunnelling and gives rise to a renormalized instanton (see, for example [13]). Although this effect itself can be important (e.g. in the adiabatic limit described below), it cannot influence the interference between topologically distinct instantons, and can be absorbed into the renormalized value of A_0. The last factor can be considered as the effect on $\boldsymbol{\sigma}$ of the tunnelling $\boldsymbol{S}$, described by the transfer matrix $\hat{T}_{\pm} = \mathrm{e}^{\pm\mathrm{i}(\alpha\hat{\boldsymbol{\sigma}}\boldsymbol{n}+\phi)}$, where α and ϕ depend only on $\omega_0 = 2|\boldsymbol{h}(\boldsymbol{S})|$, and Ω, the bounce frequency of $\boldsymbol{S}$ coming from $H_0(\boldsymbol{S})$. To simplify the example, we assume $h(S)$ is parallel to S in which case the transfer matrix is written as $\hat{T}_{\pm} = e^{\pm i\alpha\hat{\sigma}_x}$. The particular form of α depends on the instanton trajectory [12], but in all cases one has (i) $\alpha \ll 1$ if $\Omega \gg \omega_o$ (sudden perturbation), in fact $\alpha = \omega_o/\Omega$ in this limit; and (ii) if $\Omega \ll \omega_o$ (adiabatic limit), then $\alpha \to \pi/2$, so that σ follows S. One may now write an effective Hamiltonian for the 'spin complex':

$$\begin{aligned} H_{\mathrm{eff}} &= 2\Delta_o \hat{\tau}_x \cos(\pi S + \alpha\hat{\sigma}_x) + \tfrac{1}{2}\omega_o \hat{\tau}_z \hat{\sigma}_z \\ &= 2\Delta_o \hat{\tau}_x \left[\cos\pi S \cos\alpha - \hat{\sigma}_x \sin\pi S \sin\alpha\right] + \tfrac{1}{2}\omega_o \hat{\tau}_z \hat{\sigma}_z. \end{aligned} \tag{4}$$

Then there are two possible cases, namely:

(a) S = integer: Let $S = n$. Then σ appears only in the diagonal term, and cannot flip; its energy changes by $\pm\omega_o$ when S tunnels. Thus H_{eff} becomes

$$H_{\mathrm{eff}}^{S=n} = \tilde{\Delta}_{\mathrm{eff}} \hat{\tau}_x \pm \omega_o \hat{\tau}_z \tag{5}$$

with $\tilde{\Delta}_{\mathrm{eff}} = 2\Delta_o \cos\alpha$. This is a Landau–Zener form; when $\omega_o/\tilde{\Delta}_{\mathrm{eff}} > 1$ the coherent tunnelling rate rapidly collapses to zero.

(b) S = 1/2-integer: Now, in the absence of σ, we have no MQC, since $\cos\pi S$ in (3) is zero. Adding σ gives

$$H_{\mathrm{eff}}^{S=n+1/2} = 2\Delta_o \sin\alpha\, \hat{\tau}_x \hat{\sigma}_x \tag{6}$$

so, if σ does not flip, nor can S. They flip together, with an effective splitting $\tilde{\Delta}_{\mathrm{eff}} = 2\Delta_o \sin\alpha$. For weak coupling, $\tilde{\Delta}_{\mathrm{eff}} = 2\Delta_o\omega_o/\Omega$, and for strong coupling ($\omega_o > \Omega$), $\tilde{\Delta}_{\mathrm{eff}} \simeq 2\Delta_o$. To summarize, as the coupling between S and σ is switched on, we get a smooth change in the behaviour of S, from a spin of quantum number S to one of $S + 1/2$.

From this example we learn that coupling in the range $\Delta_o < \omega_o < \Omega$ is very effective in suppressing MQC. In this range any coherent delocalization of S is connected with the transitions between the states of σ having nearly the same energy before and after the tunnelling transition. We therefore define states $\{\chi_\sigma^{(i)}\}$ as eigenfunctions of different Hamiltonians $H^{(i)}\chi_\sigma^{(i)} = E_\sigma^{(i)}\chi_\sigma^{(i)}$, where $H^{(1,2)} = H^{(0)}(\sigma) + h(S_{1,2})\sigma$, and S_1, S_2 are two degenerate states of S. Then the condition for observing MQC has a form $E_\sigma^{(1)} \simeq E_\sigma^{(2)}$, and the corresponding tunnelling amplitude is multiplied by the overlap integral $\langle\chi_\sigma^{(1)}|\chi_\sigma^{(2)}\rangle$ if σ does not flip while S tunnels, or by $\langle\chi_{-\sigma}^{(1)}|\chi_\sigma^{(2)}\rangle$ otherwise. In the case $H^{(0)} = 0$ considered above (which is, of course, an extreme one), the states $\chi_\sigma^{(1)}$ and $\chi_\sigma^{(2)}$ are orthogonal, and the resonant amplitude is zero for S integer. On the other hand $\chi_{-\sigma}^{(1)} = \chi_\sigma^{(2)}$, but the matrix element of simultaneous rotation of S = 1/2-integer and σ has an additional small factor $\omega_o/\Omega \ll 1$.

Suppose we add a term $\omega_1\hat{\sigma}_z$ to the Hamiltonian (4), and $\omega_1, \omega_o > \Delta_o$. Now all four states of the 'spin complex' have different energies, and are therefore localized irrespective of the value of the topological phase. Unlike many other MQC problems we obviously lack here the condition that the energies of the initial and final states can be matched at all, because there is no special symmetry for the spectra of the environmental spin $\{E_\sigma^{(1)}\}$ and $\{E_\sigma^{(2)}\}$ to coincide. This is disastrous, because now even one spin can suppress MQC completely!

We face here a rather general question of whether it is possible to prepare a system so precisely that the frequencies ω_1 and ω_o can be neglected. In this connection, note that the giant spin is also very sensitive to any magnetic field fluctuations in the direction parallel to

$S_1 - S_2$. With $\Delta_o \sim 1$ MHz and $S \sim 10^3$, for example, this fluctuation should be less than $\delta H = \Delta_o/\gamma|S_1 - S_2| \sim 10^{-4}$ G in order to maintain MQC. One could try applying some compensating magnetic field to preserve the degeneracy in $H_o(\boldsymbol{S}, \boldsymbol{H})$, and, if necessary, to adjust at least two levels of the spin complex with accuracy of the order of Δ_{eff} (we return to experiments in the conclusions).

2.2. *N environmental spins*

Consider now the general case, including a possible external field perpendicular to $\boldsymbol{S}_1 - \boldsymbol{S}_2$, and dropping the condition $H_k^{(0)} = 0$. This gives an effective Hamiltonian

$$H_{\text{eff}} = 2\Delta_o \hat{\tau}_x \cos\left(\Phi + \sum_{k=1}^{N}(\alpha_k \hat{\sigma}_k n_k + \phi_k)\right) + \sum_{k=1}^{N}\left(H_k^{(0)}(\boldsymbol{\sigma}_k, \boldsymbol{H}) + \frac{1}{2}\omega_k \hat{\sigma}_k^z \hat{\tau}_z\right) \tag{7}$$

where $\Phi(\boldsymbol{H}_o)$ is a magnetic field dependent topological phase, $\Phi(0) = \pi S$ [14], and for the angles α_k and ϕ_k one has (i) $\alpha_k, \phi_k \to 0$, when $E_k \ll \Omega$ (where E_k is the energy splitting of the eigenvalues of $\boldsymbol{\sigma}_k$; of course when $\boldsymbol{H}_o$ and $H_k^{(0)}$ are zero, then $E_k = \omega_k$), and (ii) in the adiabatic limit $\alpha_k \to \pi/2$ and $\phi_k \to \phi_k^{\text{B}} - \pi/2$, where ϕ_k^B is the Berry phase accumulated by $\boldsymbol{\sigma}_k$ while rotating adiabatically with the giant spin. Assuming Φ to be an arbitrary number we simply absorb the sum $\sum_{k=1}^{N}\phi_k$ into a new definition of topological phase: $\Phi \to \bar{\Phi}$

We proceed now in two steps. First, we discuss the second term in (7) and show that under rather general conditions this term suppresses MQC in exactly the same way as it does in the case of one environmental spin. Then we solve exactly the Hamiltonian with only the first term being non-zero—this corresponds to the purely topological decoherence when *all* states have the same energy and phase randomization is the only reason for decoherence.

(i) It is clear from the behaviour of α_k and ϕ_k that we have to consider only those environmental spins having $E_k \leqslant \Omega$, otherwise they will rotate adiabatically with $\boldsymbol{S}$. The crucial point is that usually $\omega_k/E_k \sim 1$. (In the Caldeira–Leggett formalism one assumes $\omega_k/E_k \to 0$ for each environmental mode, and only the collective effect of all modes gives an observable result). In order to tunnel in a resonant way we have either to flip $\boldsymbol{\sigma}_k$, which gives a small factor $\alpha_k \ll 1$, and/or to project its states with the same energy, which for $\omega_k/E_k \sim 1$ gives a value of the overlap integral $R_k = \langle\chi_{\sigma'}^{(1)}|\chi_{\sigma}^{(2)}\rangle$ between 1 and 0 (recall that $R_k \to 0$ when $E_k \to \omega_k$, as demonstrated by the Hamiltonian (4)). Thus in the best case we estimate the effective tunnelling amplitude as

$$\Delta_{\text{eff}} \sim \Delta_o \prod_{k=1}^{N} R_k \ll \Delta_o. \tag{8}$$

If we try to tunnel without adjusting the environmental wavefunction to the same energy, then we face an energy fluctuation between initial and final states of order $\sum_k \omega_k \sim \omega_o N^{1/2} > \Delta_o$ (we assume here a high-temperature random energy distribution for $\{\boldsymbol{\sigma}_k\}$), which prevents coherent delocalization. Note that the condition of adjusting the levels of the spin complex is now more severe; $\delta H = \Delta_{\text{eff}}/\gamma|S_1 - S_2|$. One now requires $\Delta_o \gg 1$MHz and $|S_1 - S_2| \ll 10^3$ to observe MQC in any experiment; in fact coherence is only realistic for $S \sim 10$ or less (i.e. no longer macroscopic!). Moreover, experiments on such small grains are impossible (at least with present technology); multi-grain experiments would be required, and unless all such grains were exactly aligned together, with exactly degenerate states present in all grains (!), coherence would again be suppressed.

(ii) Now we turn to the most intriguing case when any tunnelling event is a resonant transition, which corresponds to considering the Hamiltonian

$$H_{\text{top}} = 2\Delta_o \hat{\tau}_x \cos\left(\bar{\Phi} + \sum_{k=1}^{N} \alpha_k \hat{\sigma}_{n_k}\right). \tag{9}$$

We study the probability $P_{11}(t)$ to find the giant spin in state $|1\rangle$ at time t if it was there at $t = 0$. Then the series generated by (9) has the form

$$P_{11}(t) = \frac{1}{2} + \frac{1}{2}\left\langle \cos\left[4\Delta_o \cos\left(\bar{\Phi} + \sum_{k=1}^{N} (\alpha_k \hat{\sigma}_{n_k})\right)\right]\right\rangle \tag{10}$$

where $\langle\cdots\rangle$ stands for the trace over the environmental states. Alternatively, we can present the average as

$$\sum_{nm=0}^{\infty} (-1)^{n+m} \left\{ \frac{(2\Delta_o t)^{2n}(2\Delta_o t)^{2m}}{(2n)!(2m)!} e^{-i\bar{\Phi}(2n-2m)} \left\langle \prod_{k=1}^{N} e^{-i\alpha_k \hat{\sigma}_{n_k}(2n-2m)} \right\rangle \right.$$
$$\left. - \frac{(2\Delta_o t)^{2n+1}(2\Delta_o t)^{2m+1}}{(2n+1)!(2m+1)!} e^{-i\bar{\Phi}(2n+1-2m-1)} \left\langle \prod_{k=1}^{N} e^{-i\alpha_k \hat{\sigma}_{n_k}(2n-2m)} \right\rangle \right\}. \tag{11}$$

Introducing the function

$$F(n-m) = \prod_{k=1}^{N} \cos\left(2\alpha_k(n-m)\right) \tag{12}$$

we can evaluate the series to give

$$P_{11} = \frac{1}{2}\left[1 + J_0(4\Delta_o t) + 2\sum_{\nu=1}^{\infty} (-1)^{\nu} F(\nu) \cos(2\nu\bar{\Phi}) J_{2\nu}(4\Delta_o t)\right] \tag{13}$$

where $J_{2\nu}$ is a Bessel function. From this expression the non-interacting case ($F(\nu) = 1$) is easily recovering to give $P_{11} = 1/2\{1 + \cos[4\Delta_o \cos(\bar{\Phi})t]\}$. The adiabatic limit is obtained by noting that with $\alpha_k = \pi/2$ the effect of $F(\nu) = i^{2n-2m}$ can be entirely absorbed into the topological phase $\Phi_{\text{ad}} = \Phi + \sum_{k=1}^{N}(\phi_k^B - \pi/2) + N\pi/2 \equiv \Phi + \sum_{k=1}^{N} \phi_k^B$.

The most destructive contribution comes from spins with $\alpha_k, \pi - \alpha_k \sim 1$. With only a few spins in this region the F function rapidly collapses to $F(\nu) = \delta_{\nu,0}$. In this case the answer is universal, depending on neither Φ nor the couplings;

$$P_{11} \to \tfrac{1}{2}\left[1 + J_0(4\Delta_o t)\right]. \tag{14}$$

Thus, the phase randomization does lead to the decay of coherent oscillations, although the decay law $t^{-1/2}$ is much weaker then one might expect. The reason is clearly seen from the structure of (11): the basic contribution to the result (14) comes from trajectories with equal number of clockwise and anticlockwise instantons. All phase factors completely cancel for these trajectories, but the fraction of such trajectories goes to zero at long time as $(2N)!/(N!N!2^{2N}) \sim N^{-1/2}$, where $N \sim \Delta_o t$. The spectral function of (14) is that of a 1D tight-binding model—instead of a sharp resonance peak at some frequency $\omega = \Delta_{\text{eff}}$ we find a broad (but finite) structure for $0 < \omega < 8\Delta_o$. As discussed above, if the second term in the Hamiltonian (7) is non-zero it will certainly suppress MQC.

3. Conclusions and experiments

We have analysed here a new kind of decoherence mechanism, in a Hamiltonian framework, for the case of ferromagnetic grains. This mechanism is quite outside the Caldeira–Leggett framework for the description of envirr .mental decoherence, since the effect of each environmental spin mode, in destroying phase coherence, is strong, except under very special circumstances. This makes the prognosis for experimental observation of MQC in grains very pessimistic: quite apart from any asymmetry in the potential, caused by stray fields $\delta \boldsymbol{H}$ parallel to $\boldsymbol{S}_1 - \boldsymbol{S}_2$, we have to deal with the decoherence caused by environmental spin flips, discussed here. In the general case, where crystal and external fields act on the environmental spins (the 2nd term in (7)), this effectively kills quantum coherence except for microscopic spins ($S \leqslant 10$). In the special case where these fields are absent, we still have 'pure' topological decoherence, which also destroys MQC, and leads to an interesting long-time tail in $P_{11}(t)$. In any case, this decoherence proves fatal to MQC if more than a few environmental spins are coupled to $\boldsymbol{S}$ in the energy range between Δ_o and Ω.

It is nevertheless interesting to see if it is possible to evade this topological decoherence in some way, on some experiment. Since [3] typically $\Omega \geqslant 10^8$ Hz, and $\Delta_o \leqslant 10^6$ Hz (with Ω/Δ_o typically 10^4 or greater) then it is clear that nuclear spins will be the worst source of topological decoherence. This suggests three possible strategies, namely:

(i) Choose a magnet with extremely high NMR frequencies. The best candidate seems to be ^{159}Tb, with an NMR frequency of over 8 GHz, or ^{165}Ho, with quadrupole-split NMR frequencies [15] of 2439, 3108, and 3776 MHz (at $T = 1.5$ K); it has an easy axis in the basal plane. One may then hope that all E_k lie *above* Ω; the main problem here is to find a substrate or solvent for the grains having no E_k, ω_k in the range from Δ_o to Ω.

(ii) Choose a magnet with nuclear spin $I = 0$. The most common isotopes of Fe and Ni do the job, but it will be necessary to remove the $\simeq 2\%$ of finite-spin isotopes; and one still has the problem of substrate/solute nuclear spins.

(iii) Apply a strong magnetic field $\boldsymbol{H}_o$ perpendicular to $\boldsymbol{S}_1 - \boldsymbol{S}_2$, which not only lowers the energy barrier, but also suppresses ω_k/E_k, leaving only pure topological decoherence.

It is possible that a combination of (i) or (ii) with (iii) might do the trick—otherwise we shall have to wait for MQC experiments on SQUIDS [16]. We emphasize here that none of what we have said will affect ordinary *tunnelling* of grain magnetization.

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