Suppression of Macroscopic Quantum Coherence in Magnetic Particles by Nuclear Spins

Anupam Garg
Department of Physics and Astronomy, Northwestern University, Evanston, Illinois 60208
(Received 28 March 1994)

The Néel vector of a small antiferromagnetic particle can in principle resonate between opposite directions. Nuclear spins strongly suppress this and related macroscopic quantum coherence (MQC), even when magnetic nuclei have only 2% abundance. The resonance signal breaks up into well separated groups of lines, and the highest frequency signal height *increases* with temperature as $\sim\exp(-T_0^2/T^2)$. For a recent claim to have seen MQC in ferritin, less than 3.5% of the particles are found capable of contributing to the signal.

PACS numbers: 75.60.Jp, 03.65.Db, 76.20.+q, 76.60.Jx

Resonance between degenerate states of a complex system, or macroscopic quantum coherence (MQC) as Leggett [1] has termed it, becomes progressively harder to see as the system becomes larger. The principal reason is that macroscopic systems are almost always coupled to an environment which is sensitive to different system states, and *dynamically* suppresses MQC [2]. Recently, Awschalom *et al.* claim to have seen MQC [3] in particles of ferritin, an iron storage protein in the form of a hollow shell of 75 Å inner diameter, that can be filled with an inorganic compound close in composition and structure to ferrihydrite or hydrated α-Fe_2O_3 [4]. This core is believed to be antiferromagnetic, and Awschalom *et al.* ascribe the peak in their ac susceptibility to resonance of the Néel vector between opposite easy axes.

In this Letter we show that nuclear spins strongly suppress MQC in magnetic particles. We stress at the outset that this study is quite different from a previous one [5] of the effect of nuclear spins on macroscopic quantum *tunneling* (MQT) in magnetic particles. It is essential to discriminate between MQC and MQT. The latter refers to the decay of a metastable state. MQC is a far more delicate phenomenon than MQT, as it is much more easily destroyed by an environment, and by very small c-number symmetry breaking fields that spoil the degeneracy. The present results follow these general expectations. While nuclear spins do suppress MQT [5], their effect can be reduced by using elements such as Fe and Ni with low natural abundances of the magnetic nuclear species (^{57}Fe, ^{61}Ni). By contrast we will find that even with such elements, the effect on MQC is severe.

Prokof'ev and Stamp [6] have also studied the effect of environmental spins on MQC. They also conclude that MQC is suppressed, but their emphasis is rather different. They focus on topological effects, and on a much larger range of environmental spin frequencies with a view to studying the conceptually intriguing transition from weak to strong coupling. We will limit ourselves to the frequency range relevant to nuclear spins, and focus on the ac susceptibility $\chi''(\omega)$ as the quantity of greatest experimental interest. Our Hamiltonian [Eqs. (6) and (7) below] is in fact buried in Eq. (7) of Ref. [6], but the remarkable resulting behavior of χ'' is not realized there. We find that the spectral line at the bare tunneling frequency Δ_0 is chopped up into a large number of lines with $\omega \le \Delta_0$. Almost all the spectral weight is at $\omega \ll \Delta_0$, but the line at Δ_0 persists, and has a weight given by the fraction of magnetic particles with no net nuclear spin polarization p. Since this fraction increases with temperature T, a surprising result is that MQC may be enhanced with increasing T in some range. It also suggests double resonance experiments—driving p to zero by a strong rf pulse at ω_n would lead to strong transient enhancement of the MQC signal.

These results also contrast strikingly with those for the two level problem with an Ohmic bath [7]. There, the bare line is pulled down (possibly to $\omega = 0$) and broadened, and acquires a low frequency tail, but it remains one line. It is interesting that our bath strongly violates the Caldeira-Leggett condition [8] under which any bath is equivalent to a set of harmonic oscillators, viz. that any *one* bath degree of freedom be weakly perturbed by the system. This condition *is* met by nuclear spins for MQT [5], elastic waves for both MQT and MQC [9], and Stoner excitations for domain wall MQT in metallic magnets [10]. The differences offer a valuable lesson in the variety of ways in which an environment can affect MQC and MQT.

We will treat only antiferromagnetic MQC [11]. The results extend trivially to ferromagnetic MQC through 180° [6,12]. We consider a small uniaxially anisotropic antiferromagnetic particle with N_e ($\lesssim 10^4$) atomic moments or spins, each of magnitude s. Denoting the spin directions on the two sublattices by unit vectors $\hat{\mathbf{n}}_1$ and $\hat{\mathbf{n}}_2$, we have the obvious Hamiltonian [11b,13]:

$$\mathcal{H}_e = J\hat{\mathbf{n}}_1 \cdot \hat{\mathbf{n}}_2 - K(\hat{\mathbf{n}}_{1z}^2 + \hat{\mathbf{n}}_{2z}^2), \tag{1}$$

where $J \gg K > 0$. Adiabatic elimination of the total moment ($\propto \hat{\mathbf{n}}_1 + \hat{\mathbf{n}}_2$) leads to the following Euclidean

 0031-9007/95/74(8)/1458(4)$06.00

action for the Néel vector $\hat{\mathbf{l}}$:

$$S_0[\hat{\mathbf{l}}(\tau)] = \frac{(N_e \hbar s)^2}{8J} \int (\dot{\theta}^2 + \sin^2\theta\, \dot{\phi}^2 + \omega_e^2 \sin^2\theta)\, d\tau\,, \tag{2}$$

where θ and ϕ are the polar angles of $\hat{\mathbf{l}}$ and $\omega_e = 4(JK)^{1/2}/N_e \hbar s$ is the antiferromagnetic resonance frequency. Instanton methods [11,13] give a tunneling frequency, $\Delta_0 = (\omega_e/\pi)e^{-B}$, where $B = 2N_e s(K/J)^{1/2}$. The instanton itself is given by

$$\sin\theta = \operatorname{sech}\omega_e\tau\,, \qquad \dot{\phi} = 0\,. \tag{3}$$

For typical values of J and K, $\omega_e/2\pi \sim 10^{11}$–$10^{13}$ Hz. Even for a weak ferromagnet such as α-Fe_2O_3, $\omega_e/2\pi \sim$ 10 GHz. Because $B \propto N_e$, it is unlikely that Δ_0 will exceed more than a few MHz, if $N_e \gtrsim 5000$.

Next, let N of the magnetic ions have magnetic nuclei. Each nuclear spin $\mathbf{I}_i$ couples to the electronic spin $\mathbf{s}_i$ on the same atom, and the total interaction Hamiltonian can be written as

$$\mathcal{H}_{en} = -A \sum_{i=1}^{N} \mathbf{I}_i \cdot \mathbf{s}_i\,. \tag{4}$$

The nuclear Larmor frequency is given by $\omega_n = As/\hbar$. For simplicity we consider only $I = \frac{1}{2}$. For ferritin, the relevant nucleus is ^{57}Fe, I *is* $\frac{1}{2}$, and the hyperfine field is known from Mössbauer data [4] to be 50 ± 2 T, giving $\omega_n/2\pi = 68.5$ MHz ($\equiv 3.3$ mK). These are typical hyperfine fields for magnetic ions [14], and we can safely assume that $\omega_e \gg \omega_n \gg \Delta_0$.

In lieu of $\chi''(\omega)$, we will find the correlation function ($\mathbf{M}$ is the total uncompensated moment of a particle with $M_0 = |\mathbf{M}|$):

$$C(t) = \langle \mathbf{M}(t) \cdot \mathbf{M}(0) \rangle = M_0^2 \langle \hat{\mathbf{l}}(t) \cdot \hat{\mathbf{l}}(0) \rangle\,. \tag{5}$$

To do this, we will first analyze the system composed of the Néel vector restricted to the two states $|\hat{\mathbf{l}} = \pm\hat{\mathbf{z}}\rangle$ and the nuclear spins, and obtain approximately the energy level spectrum. Since the system is finite, these levels will be sharp. We will then put in relaxation mechanisms by hand in the form of a nuclear T_1 time T_{1n}. Processes that give rise to T_{1n} include phonons, paramagnetic impurities, and other nuclear species' spins.

To find the spectrum, let us first represent the states $|\hat{\mathbf{l}} = \pm\hat{\mathbf{z}}\rangle$ as pseudospin states $|+\rangle$ and $|-\rangle$ and write the Hamiltonian using corresponding spin operators σ_x, σ_y, and σ_z. $\mathcal{H}_e$ is evidently mapped on to $\hbar\Delta_0\sigma_x/2$. To map $\mathcal{H}_{en}$, we use a left handed system of nuclear spin axis on one sublattice, effectively inverting $\mathbf{s}_i$, so that $\mathbf{i} \cdot \mathbf{s}_i \to \mathbf{i} \cdot \hat{\mathbf{l}}$, for all i in Eq. (4). If Eq. (4) is projected onto the states $|\pm\rangle$, we evidently get a term proportional to σ_z, so the total Hamiltonian is

$$\mathcal{H}_0 = \frac{1}{2}\hbar\Delta_0\sigma_x - \frac{1}{2}\hbar\omega_n\sigma_z \sum_{k=1}^{N} \mu_k^z\,, \tag{6}$$

where $\mu_k = 2\mathbf{I}_k$ are the Pauli spin operators for the nuclei. Note that, as discussed in Sect. I of [7(b)] [see the remarks preceding Eq. (1.4) there], Eq. (6) does not have any terms coupling the bath to σ_x or σ_y. Such terms describe processes wherein the bath changes the overlap of $|+\rangle$ and $|-\rangle$. They are themselves of order Δ_0 or smaller, and thus negligible compared to the σ_z term. The nonlinearity of our bath and the difference with the spin-boson problem [7] is now manifest: According to Eq. (6), the z component of each nuclear spin is conserved, and so, therefore, is the polarization $p = \sum_k \mu_k^z$. (Note that p is defined to be an integer.) If we label the states as $|\pm, p, \alpha\rangle$, where p is the polarization, and α is the remaining set of quantum numbers, $|+, p, \alpha\rangle$ and $|-, -p', \alpha'\rangle$ are nondegenerate by $|p - p'|\omega_n$, which far exceeds Δ_0, if $p \neq p'$. The mixing between such states is thus negligible, and, since p is conserved, we obtain the very simple result that *the dominant tunneling occurs only when* $p = 0$, *and then at the bare frequency* Δ_0.

Let us now consider the states with $p \neq 0$. Since the states $|+, p, \alpha\rangle$ and $|-, -p, \alpha'\rangle$ for a given p are all degenerate by the above argument, let us seek an effective Hamiltonian describing how each group is split by the small amount of tunneling that does occur. We see from Eq. (4) that to flip a nuclear spin, the local field seen by it, which is parallel to $\hat{\mathbf{l}}$, must have components in the x-y plane. In other words, nuclear spins can only coflip with $\hat{\mathbf{l}}$. This point becomes very clear if one computes the path integral for the total partition function of our system using the action (2) and the coupling (4). This path integral is still dominated by instantons for $\hat{\mathbf{l}}$. Between instantons, each nuclear spin sees a field along $\hat{\mathbf{z}}$, and cannot flip. During an instanton, it sees a field in the x-y plane of magnitude $\hbar\omega_n/\gamma_n$ (γ_n is the nuclear spectroscopic splitting ratio) for a time $\sim\omega_e^{-1}$. Since $\omega_n/\omega_e \ll 1$, the amplitude Δ_k for k spins to coflip is of order $\Delta_k \sim \Delta_0(\omega_n/\omega_e)^k$ from perturbation theory. (A more accurate calculation will be given later.) The desired coflip Hamiltonian which is block diagonal by polarization can thus be written as

$$\mathcal{H}_{cf} = \frac{1}{2}\hbar\sigma_x \sum_{p=1}^{N} \Delta_p Q_p\,,$$

$$Q_p = \sum_{i_1,\ldots,i_p} \pi_p(\mu_{i_1}^x \mu_{i_2}^x \cdots \mu_{i_p}^x)\pi_p\,. \tag{7}$$

Here, π_n is a projection operator onto states with $p = n$ *and* $p = -n$.

It is straightforward to diagonalize Eq. (7) by rewriting Q_p in terms of the total nuclear spin. Let us denote the *tunnel splittings* and corresponding degeneracies within each polarization block by $\Omega_{p,k}$ and $g_{p,k}$. We find

$$\Omega_{p,k} = \Delta_p \binom{N_+ - k}{p}, \tag{8}$$

$$g_{p,k} = \binom{N}{k} - \binom{N}{k-1}, \tag{9}$$

where $k = 0, 1, \ldots, N_-$, and $N_\pm = (N \pm |p|)/2$. The second moment (about zero) within each group is $\mathrm{Tr}(Q_p^2/2)$, which gives

$$\langle \Omega_p^2 \rangle = \Delta_p^2 \binom{N_+}{p}. \tag{10}$$

A closed form can also be found for $\langle \Omega_p^4 \rangle$. Odd moments are harder. For $p = 1$, 2, and 3, e.g., as $N \to \infty$, we have

$$\frac{\langle \Omega_p \rangle}{\Delta_p} \approx \frac{\sqrt{2\pi N}}{4}, \qquad \frac{N}{4}, \qquad \frac{\sqrt{2\pi N^3}}{32}. \tag{11}$$

We thus see that the spectrum of χ'' consists of a series of groups of lines centered at $\langle \Omega_p \rangle$ plus a single line at Δ_0. Each group is lower than the previous one by the ratio $\sim N^{1/2}\omega_n/\omega_e$, instead of ω_n/ω_e, but this is still small compared to unity if $N \sim 100$ or so. The relative weights of the lines in a group are given by Eq. (9), and the total weight of a group is proportional to the thermal distribution of the polarization:

$$f_p = (2\pi\sigma_p^2)^{-1/2} e^{-(p-\overline{p})^2/2\sigma_p^2}, \tag{12}$$

where $\overline{p} = N \tanh(\beta\epsilon_n/2)$, $\sigma_p = N^{1/2}\,\mathrm{sech}(\beta\epsilon_n/2)$, $\beta = 1/k_BT$, and $\epsilon_n = \hbar\omega_n$.

Relaxation will broaden the lines as follows. Suppose we are in a state $|+, \{\mu^z\}\rangle$, with some value of p at $t = 0$. This state will resonate with others of the same energy at a superposition of frequencies taken from the set $\Omega_{p,k}$. This resonance will persist until time t only if *all* the nuclear spins maintain relative phase coherence, i.e., provided *none* of them suffers a T_1 process. If T_{1n} is the relaxation time for one nuclear spin, the probability of this happening is $\exp(-Nt/T_{1n})$, and we expect $C(t)$ to decay with this factor. Since T_{1n} can be as long as a few seconds, it is possible for the lines at zero and nearby polarizations to not be overly broadened, but lines with $|p| \geq 3$ or so are unlikely to be distinguishable from a broad background. For practical purposes therefore, $C(t)$ from one particle can be written as

$$C(t) = f_0 M_0^2 \cos(\Delta_0 t) e^{-Nt/T_{1n}} + C_{\mathrm{low}}(t), \tag{13}$$

where C_{low} is the frequency contribution from $p \neq 0$. A better formula for f_0 than Eq. (12) is

$$f_0 = (2\pi N)^{-1/2} [\cosh(\beta\epsilon_n/2)]^{-N}. \tag{14}$$

For $k_BT \gg \epsilon_n$, $f_0 \sim \exp(-T_0^2/T^2)$, with $T_0^2 = N\epsilon_n^2/8k_B^2$.

An important proviso to the above results is that ω_n be very nearly the same for all nuclear spins in a particle. If the dispersion in ω_n is comparable to or more than $N^{-1/2}\Delta_0$, the line at Δ_0 will also be shifted down and split, as the different $p = 0$ states will not be degenerate. Also, the spread $\Delta\Omega_p$ within a group will be reduced for larger p values.

Let us now discuss the ferritin experiment [3] in light of these results. We have previously noted [15] that the signal seen in [3] is too large and difficult to reconcile with elementary calculations that do not explicitly account for dissipation. If nuclear spins are included, the expected signal height should be reduced still further by f_0. Each ferritin particle in [3] has $N_e = 4500$ Fe ions, giving $N = 101$, using a 2.25% abundance for ^{57}Fe, and $T_0 = 11.7$ mK, using a 50 T hyperfine field. At $T = 29.5$ mK, the lowest temperature studied in [3], and the one at which the peak in χ'' is shown, $f_0 = 0.034$. (Fluctuations in N from one particle to another have negligible effect on f_0.) Thus the upper bound P_{sat} on the peak power absorption calculated in [15] should be lowered to 5.1×10^{-23} W, and the direct estimate to 2.4×10^{-25} W. The actual absorption is much larger: 10^{-21} W. This makes the interpretation of the data in terms of MQC even more implausible.

One should expect an even smaller signal height, in fact, due to proton spins by an additional factor which we now estimate. Each ferritin particle contains about $N_p = 8000$ protons. The local field at every proton site reverses when $\hat{\mathbf{l}}$ flips, so the same considerations of coflipping, etc. apply to the protons as to the ^{57}Fe nuclei. The local field at the protons is unknown and probably distributed, but a mean value of 100 G of dipolar origin is not unreasonable. This gives $\epsilon_n = 20\ \mu$K or 0.43 MHz in frequency units, compared to $\Delta_0 = 950$ kHz. If we allow states differing by less than 2Δ to mix (although the frequency spread is larger than seen), only particles with proton polarization $|p| \leq 5$ can contribute. This fraction is $\sim 11(2\pi N_p)^{-1/2} = 0.05$. Note that this is almost purely entropic, so it is not too sensitive to uncertainties in the local field.

We conclude by calculating Δ_p. For $k_BT \ll \hbar\omega_e$, the partition function is given by the path integral over all closed paths obeying $\hat{\mathbf{l}}(0) = \hat{\mathbf{l}}(\beta\hbar) = \hat{\mathbf{z}}$:

$$Z(\beta) = 2 \oint [d\hat{\mathbf{l}}] e^{-S_0[\hat{\mathbf{l}}(\tau)]/\hbar} \Lambda[\hat{\mathbf{l}}(\tau)], \tag{15}$$

$$\Lambda = z_0^{-N} \mathrm{Tr}_n \left[T_\tau \exp\left(-\hbar^{-1} \int_0^{\beta\hbar} \mathcal{H}_{en}(\tau)\, d\tau \right) \right], \tag{16}$$

where $z_0 = 2\cosh(\beta\epsilon_n/2)$, and Tr_n denotes a trace over the nuclear spins. Without the latter, the dominant paths consist of instantons with a width $\sim\omega_e^{-1}$, and separations $\gg\omega_e^{-1}$. The j instanton path (where j must be even) has an action jB. The fluctuations around these paths can be thought of as composed of independent sets of fluctuations around each instanton, and give a factor D^j, where $D \sim \omega_e$ is the factor (or fluctuation determinant) for one instanton. Integrating over the locations of the instanton centers gives $(\hbar\beta)^j/j!$, and summing over j we obtain $\tilde{Z} = 2\cosh(\Delta_0/2k_BT)$, with $\Delta_0 \propto De^{-B}$.

A key point is that the mean instanton-instanton separation is $\sim\Delta_0^{-1}$. If we had a nondegenerate problem, where the energy of the $\hat{\mathbf{l}} = -\hat{\mathbf{z}}$ exceeded that of the $\hat{\mathbf{z}}$ state by ϵ, with $\epsilon \gg \Delta_0$, we would find that the mean durations of the paths in the $-\hat{\mathbf{z}}$ state would shrink to order ϵ^{-1}. Each instanton would be effectively bound to an anti-instanton, and j instanton paths would add to give a contribution of $O(\beta^{j/2})$ and not $O(\beta^j)$. The same holds when nuclear spins are included. The important paths are those where instantons join degenerate states, and which require as few nuclear spin flips as possible. Let us denote Λ for a j instanton path by Λ_j, the contribution to Λ_j from states with polarization p by $\Lambda_{j,p}$, and define Z_j and $Z_{j,p}$ analogously. For the case $j = 0$, $\hat{\mathbf{l}}(\tau) = \hat{\mathbf{z}}$, and $\Lambda_0 = 1$. Next, let us consider $j = 2$, with instanton centers at τ_1, τ_2, and $\tau_2 - \tau_1 \gg \omega_e^{-1}$. Suppose the initial state is one of $|+, p, \alpha\rangle$. This has N_+ up spins and N_- downs. At τ_1, p spins must flip from up to down to maintain degeneracy. These can be chosen in

$$\binom{N_+}{p}$$

ways (assuming $p > 0$), but the same p spins must flip back to up at τ_2. Since the magnetic field seen by one nucleus during the instanton at τ_i is $\hbar\omega_n\gamma_n^{-1}\hat{\mathbf{l}}(\tau - \tau_i)$, and since $\omega_e^{-1} \ll \hbar\beta$, the contribution of this state to Λ_2 is

$$\frac{e^{p\beta\epsilon_n/2}}{z_0^N}\binom{N_+}{p}(U_2^{+-}U_1^{-+})^p, \qquad (17)$$

where U_1^{-+} is the one coflip amplitude:

$$U_1^{-+} = \left\langle - \left| T_\tau e^{-\omega_n \int \mathbf{I}\cdot\hat{\mathbf{l}}_1 \, d\tau} \right| + \right\rangle, \qquad (18)$$

and U_2^{+-} is similarly defined. As $\omega_n/\omega_e \ll 1$, perturbation theory suffices, and we get

$$U_1^{-+} = \frac{\omega_n}{2}\int_{-\infty}^{\infty} \hat{\mathbf{l}}_{1+}(t)\, dt. \qquad (19)$$

For U_2^{+-}, we get $\hat{\mathbf{l}}_{2-}$ instead; $\hat{\mathbf{l}}_\pm = \hat{\mathbf{l}}_x \pm i\hat{\mathbf{l}}_y$. Note that we can extend the limits in Eq. (19) to $\pm\infty$, as $\hat{\mathbf{l}}_\pm \simeq 0$, if $\omega_e|\tau - \tau_i| \gg 1$. Multiplying Eq. (17) by the total number of states with polarization p, we get

$$\Lambda_{2,p} = f_p\binom{N_+}{p}\left(\frac{\omega_n}{2}\right)^{2p}\left[\int_{-\infty}^{\infty}\int_{-\infty}^{\infty} dt\, ds\, \sin\theta_1(t) \times \sin\theta_2(s) e^{i\phi_1(t) - i\phi_2(s)}\right]^p, \qquad (20)$$

where θ_i and ϕ_i are polar angles for $\hat{\mathbf{l}}_i$. The corresponding term in the partition function is

$$Z_{2,p} = 2\oint [d\hat{\mathbf{l}}] e^{-S_0[\hat{\mathbf{l}}]/\hbar}\Lambda_{2,p}. \qquad (21)$$

The paths are now restricted to have two instantons, and only their shape needs to be found. Since we only need the answer asymptotically as $N_e \to \infty$, the important point is that the factor of $\Lambda_{2,p}$ in Eq. (21) does not modify the least action path, just as in one-dimensional Laplace integrals of the type $\int h(t)e^{xu(t)}\, dt$, the function $h(t)$ does not modify the critical points $u'(t_c) = 0$, which give the leading $x \to \infty$ behavior. Note in particular that the ω_n^{2p} factor multiplies the entire expression and thus cannot affect the time scale of the instanton. We can thus use Eq. (3) for $\sin\theta$ in Eq. (21), but ϕ_1 and ϕ_2 are no longer independent, and $\Lambda_{2,p}$ is evidently largest when $\phi_1 = \phi_2$. The factor in square brackets in Eq. (20) is then $(\pi/\omega_e)^2$. Integration over the instanton centers yields a factor of $(\hbar\beta)^2/2$, and, since $Z(\beta)$ is a sum of terms of the form $2\cosh(\beta\hbar\Omega_{p,k}/2$, Eq. (21) is the β^2 term in its Taylor expansion. The remaining factor in Eq. (21) is evidently $f_p\langle\Omega_p^2\rangle$. Comparing with Eq. (10), we get

$$\Delta_p = \Delta_0(\pi\omega_n/2\omega_e)^p. \qquad (22)$$

This work is supported by the National Science Foundation through Grant No. DMR-9306947.

[1] See A.J. Leggett, in *Chance and Matter,* edited by J. Souletie, J. Vannimenus, and R. Stora (North-Holland, Amsterdam, 1987), for a review of MQT and MQC.

[2] M. Simonius, Phys. Rev. Lett. **40**, 980 (1978); R.A. Harris and L. Stodolsky, J. Chem. Phys. **74**, 2145 (1981).

[3] D.D. Awschalom *et al.,* Phys. Rev. Lett. **68**, 3092 (1992).

[4] See P.M. Harrison and T.H. Lilley in *Iron Carriers and Iron Proteins,* edited by T.M. Loehr (VCH Publishers, New York, 1989), for an up-to-date review on ferritin.

[5] A. Garg, Phys. Rev. Lett. **70**, 1541 (1993).

[6] N.V. Prokof'ev and P.C.E. Stamp, J. Phys. Condens. Matter **5**, L663 (1993).

[7] (a) S. Chakravarty and A.J. Leggett, Phys. Rev. Lett. **52**, 5 (1984); (b) A.J. Leggett *et al.,* Rev. Mod. Phys. **59**, 1 (1987).

[8] (a) A.O. Caldeira and A.J. Leggett, Phys. Rev. Lett. **46**, 211 (1981); (b) Ann. Phys. (N.Y.) **149**, 374 (1984); **153**, 445(E) (1984). See Sect. 2 and App. C.

[9] A. Garg and G.-H. Kim, Phys. Rev. Lett. **63**, 2512 (1989); Phys. Rev. B **43**, 712 (1991).

[10] G. Tatara and H. Fukuyama, Phys. Rev. Lett. **72**, 772 (1994); (unpublished).

[11] (a) B. Barbara and E.M. Chudnovsky, Phys. Lett. A **145**, 205 (1990); (b) I.V. Krive and O.B. Zaslavskii, J. Phys. Condens. Matter **2**, 9457 (1990).

[12] By applying external fields, degenerate easy directions that are at any angle θ can be created: See model II in E.M. Chudnovsky and L. Gunther, Phys. Rev. Lett. **60**, 661 (1988) and A. Garg, Europhys. Lett. **22**, 205 (1993). If θ is not too close to π, the nuclear spins do not need to coflip; this case is rather different and will be discussed elsewhere.

[13] J.-M. Duan and A. Garg, Physica (Amsterdam) **194–196B**, 2017 (1994). To detect the resonance via the total moment **M**, the two sublattices must have unequal total spin. As Eq. (1) has uniaxial symmetry, **M** is rigorously conserved, and to describe its tunneling, one must include transverse anisotropy. This work will be published elsewhere. That we will still use Eq. (1) as the detailed form of the instanton is unimportant for us; only $\omega_e \gg \omega_n$ matters.

[14] E.A. Turov and M.P. Petrov, *Nuclear Magnetic Resonance in Ferro- and Antiferromagnets* (Wiley, New York, 1972).

[15] A. Garg, Phys. Rev. Lett. **71**, 4249 (1993); see also D.D. Awschalom *et al., ibid.* **71**, 4276 (1993).