Dissipation and interference effects in macroscopic magnetization tunneling and coherence

Anupam Garg
Department of Physics and Astronomy, Northwestern University, Evanston, Illinois 60208
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Macroscopic quantum coherence (MQC) and tunneling (MQT) of the total moment of a ferromagnetic particle are considered in the presence of a magnetic field, and the topological quenching, or spin-parity effect in the tunneling rate that has been found in recent papers is shown to follow from a selection rule due to an underlying rotational symmetry in both cases. The oscillation in the tunneling rate with magnetic field is considered more carefully for the MQT porblem. In addition to the rotational symmetry, this oscillation is shown to require that the intermediate state obtained immediately after the magnetization has tunneled out of its metastable orientation have a narrow decay width. The tolerances on the decay width and misalignment of the magnetic field are derived, and the decay of the intermediate state is qualitatively discussed, along with its implications for the free induction decay of the moment in a small particle.

I. INTRODUCTION

The phenomena of macroscopic quantum tunneling (MQT) and coherence (MQC) have been actively studied for almost 15 years now.[1] The two kinds of systems which are regarded as the most promising are those based on Josephson junctions[2,3] and small magnetic particles.[4,5] The macrovariable is the phase difference of the Cooper pairs on the two sides of the junction [or the related quantity, the embraced flux in the rf superconducting quantum interference device (SQUID)] in the former and the total magnetic moment (or the Néel vector if the particle is antiferromagnetic) in the latter. It has recently been pointed out[6–9] that magnetic systems can in principle display interference effects that are intrinsically absent in the Josephson-junction-based systems. These effects are ultimately rooted in the different quantum nature of spin as opposed to position and momentum, and have been related to the Berry phase in the spin coherent-state path integral.[10–12]

In this paper we reexamine two problems studied in Refs. 8 and 9, and provide a somewhat more prosaic explanation[13] for the interference effects in terms of symmetries of the Hamiltonian.[14] For both problems, we consider a small ($\lesssim 50$ Å radius), single-domain, ferromagnetic particle, with hard, medium, and easy axes of magnetization along $\hat{\mathbf{x}}$, $\hat{\mathbf{y}}$, and $\hat{\mathbf{z}}$, respectively.[15] If the temperature is low enough to freeze out spin waves, we need only consider the dynamics of the direction $\hat{\mathbf{M}}$ of the total magnetic moment $\mathbf{M}$, which can be viewed as a single large spin J, proportional to $|\mathbf{M}|$. The simplest model Hamiltonian with the correct anisotropy structure is

$$\mathcal{H}_0 = k_1 J_x^2 + k_2 J_y^2 \ , \tag{1.1}$$

where $k_1 > k_2 > 0$. If, as in Ref. 8, we now apply a magnetic field $\mathbf{H}$ along the hard axis $\hat{\mathbf{x}}$, we displace the easy directions from $\pm\hat{\mathbf{z}}$ toward $\hat{\mathbf{x}}$, and the problem is one of MQC, i.e., resonance between degenerate states. If, on the other hand, following Ref. 9, the field is applied along $-\hat{\mathbf{z}}$, the problem is one of MQT, i.e., escape from the metastable direction $\hat{\mathbf{z}}$ toward the absolutely stable one, $-\hat{\mathbf{z}}$.

In Ref. 8, it was shown that the tunnel splitting oscillated as H varied, vanishing at $2J$ values of the field, including zero and negative values.[16] We recover this result in Sec. II by an analysis reminiscent of electron paramagnetic resonance and Mössbauer spectra studies.

Similarly, in Ref. 9, Chudnovsky and DiVincenzo suggested that the escape rate Γ would also oscillate with H, although the period of the oscillations was not obtained. This is a less obvious conclusion and one which the present author specifically ruled out in Ref. 8. What is the source of these opposite conclusions? We will show that it is the way in which the states of the system in the deep well (i.e., that centered on $-\hat{\mathbf{z}}$) are treated. In Ref. 8, these states are implicitly taken to form a continuum following the usual discussion[17] of MQT for a point particle moving in a one-dimensional potential such as

$$V(q) = Aq^2 - Bq^3 \ . \tag{1.2}$$

This potential is unbounded below and is in general only an approximation to the true potential, which must be bounded below for any real system. The approximation is nevertheless believed to work on the grounds that the system will not return to the state near $q=0$ or even to the exit point once it has escaped, so that it is enough to focus on small values of q. Here Eq. (1.2) (or a similar formula) is an adequate approximation for the purpose of studying the small-t behavior of a wave packet created near $q=0$ at $t=0$. This approach is also implicit in the initial treatment[4] of magnetic MQT.

In Ref. 9, on the other hand, the deep well states are taken to be discrete, and the oscillation derives from a selection rule for transitions between an initial state in the shallow well to a nearly degenerate final state in the deep well. As the magnetic field is increased, this transition is alternately allowed and forbidden—that is the oscillation. Strictly speaking, however, this treatment is valid only for an isolated particle, in which case it is

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meaningless to talk of an *escape* rate.[18] In particular, the "no return" assumption does not hold if the particle is treated as an isolated system. For energy conservation then implies that an initial state with $\mathbf{M}\|\hat{\mathbf{z}}$ either has very little overlap with the deep well states and thus very little amplitude to tunnel or, if this amplitude is large, will flip-flop back and forth. In either case, the system cannot truly *escape* from the $\hat{\mathbf{z}}$ direction. If the particle is not isolated, its energy is not conserved, and all energy levels except that of the ground state near $-\hat{\mathbf{z}}$ acquire a nonzero width. If the levels are broadened so much that adjacent levels overlap, then the states in the deep well effectively form a continuum, and we recover the standard picture of MQT, with no oscillations in Γ.

The opposite conclusions in Refs. 8 and 9 can thus be viewed as arising from very different assumptions about dissipation or coupling of the particle to its environment. The escape rate does not oscillate with H if the dissipation is strong, while it does if the dissipation is weak. Once this is realized, the interesting issues are clearly to determine what constitutes "strong" or "weak" dissipation, whether the weak dissipation limit can be achieved in reality, and what other practical obstacles are likely to be present to the observation of escape rate oscillations.

We study these questions in Sec. III. To orient the discussion, we first examine (Sec. III A) the simpler problem of a *lightly* damped point particle moving in a one-dimensional asymmetric double well. We model damping by assigning energy widths to the deep well states. We find that even here there can be an oscillation in Γ as the energy difference between the two wells is varied. There is obviously no selection rule at work in this case, and the oscillation merely reflects the fact that if the deep well states have a narrow enough decay width, the initial state in the shallow well will have a shorter lifetime if it is almost exactly degenerate with a state in the deep well, than if it is not degenerate. The oscillations in this case are due to the structure in the density of states in the deep well. This type of oscillation is not as interesting as that due to the selection rule and will also be present in the magnetic particle if the selection rule is absent, or is not exact, for example. We will discuss how these two oscillations might be distinguished shortly, but it is apparent that requiring them to be observable leads to essentially the same condition on the dissipation.

The simple model of Sec. III A suggests that we distinguish between three types of states: the *initial* state with $\mathbf{M}\|\hat{\mathbf{z}}$, a group of *intermediate* states in the deep well with $\mathbf{M}\nparallel\hat{\mathbf{z}}$, but nearly degenerate with the initial state, and the *final* state with $\mathbf{M}\|-\hat{\mathbf{z}}$. The picture of escape for a lightly damped particle is then that $\mathbf{M}$ tunnels from $\hat{\mathbf{z}}$ to an intermediate direction, precesses about $\hat{\mathbf{z}}$ while losing energy gradually, and eventually rings down to $-\hat{\mathbf{z}}$. Oscillations in Γ (of either kind) will be observable only if the intermediate states have lifetimes in excess of the inverse of the intermediate level spacing. We find this level spacing and thus obtain a condition on their lifetime in Sec. III B. The selection rule oscillations will also be wiped out if the symmetry in question is broken by a strong enough perturbation. We show in Sec. III C that a misalignment of the magnetic field by a milliradian constitutes such a perturbation for a particle with 10^4 or so moments. We do not know if the magnetic axes of such a small particle can be determined with this accuracy or if stray magnetic fields, due to impurities outside the particle, e.g., can be eliminated to the very high degree required, but in general we are somewhat less sanguine about the feasibility of this experiment than the authors of Ref. 9.

We consider the decay of the intermediate state for real magnetic particles in Sec. III D. The answers here are less clear-cut, and it is hard to reach general conclusions. The intermediate state can be viewed as one where all the individual magnetic moments in the particle precess homogeneously about $\hat{\mathbf{z}}$. It decays toward the state with $\mathbf{M}\|-\hat{\mathbf{z}}$ primarily via the creation of magnons with nonzero wave vector. Such magnons, however, cost a minimum energy $\sim L^{-2}$ for a particle of linear size L. Depending on the exchange and coercive fields for a given particle, their creation may or may not be energetically allowed, and even if it is, the number of states into which the uniformly precessing state can decay may be very small. It is therefore conceivable that the intermediate state decay width can be small enough to yield an oscillatory structure to the decay rate of the initial state, even if the selection rule is vitiated by stray or misaligned fields. If these oscillations are ever observed, they could be easily distinguished from those due to a selection rule by deliberately applying a small misaligning field.

The possibility of a slow decay rate for the uniformly precessing mode has important implications for the relaxation of the magnetization in a dilute assembly of small particles, even in the absence of MQT. Let us imagine that the easy axes of all the particles are parallel to $\hat{\mathbf{z}}$, say, and that they are all magnetized along this axis initially. If the external field is now reversed, the initial orientation of the moments of some of particles may be absolutely unstable as a result of nonuniformity in particle sizes and magnetic properties, but if the decay toward $-\hat{\mathbf{z}}$ is to occur via tunneling or thermal activation across a succession of small barriers, the relaxation of the net magnetization of the system could easily acquire the characteristics of slow relaxation in glasses and cloud the interpretation of MQT.

We conclude this section with one final comment. Chudnovsky and DiVincenzo note, and we concur, that the MQT rate in ferromagnets is small unless the external field is close to the coercive field. A large applied field, however, precludes the observation of the Kramers selection rule that forbids tunneling from $\hat{\mathbf{z}}$ to $-\hat{\mathbf{z}}$ for half integer J and $H=0$. Chudnovsky and DiVincenzo then state that this effect may be easier to observe in antiferromagnetic particles with a small uncompensated moment.[5] We shall have nothing to say about antiferromagnetic particles in this paper, except to note that the problem is now much closer to MQC than MQT. It is this author's view that MQC is a far more delicate phenomenon than MQT in general. This is supported by quantitative calculations of the effect of Ohmic dissipation in the Josephson junction systems. The same shunt resistance affects MQC (Ref. 19) much more than MQT.[17] In the particular case of magnetic particles, nuclear spins are an important source of dissipation,[20] and the same nuclear spin environment suppresses MQC [Refs. 21 and 20(b)] (or tunneling of the moments through large angles) far more than MQT.[20]

II. THE MQC PROBLEM

In this section we will rederive the result of Ref. 8 on the oscillation of the tunnel splitting. To the Hamiltonian (1.1) we add a term describing a field $\mathbf{H}\|\hat{\mathbf{x}}$, giving

$$\mathcal{H}_C=k_1J_x^2+k_2J_y^2-g\mu_BHJ_x \ , \tag{2.1}$$

where g is a g factor. (The subscript C stands for coherence.) For $H<2K_1/\gamma J$, the classical energy (expectation value of $\mathcal{H}_C$) has minima at $\theta=\pi/2\pm\theta_0$, $\phi=0$, where $u_0=\cos\theta_0=g\mu_BH/2k_1J$. It is convenient to take $k_1=1/J^2$ and write $k_2=\lambda k_1$, $\lambda<1$. We can then write

$$\mathcal{H}_C=\frac{1}{J^2}(J_x^2+\lambda J_y^2)-\frac{2u_0}{J}J_x \ . \tag{2.2}$$

Under a 180° rotation about $\hat{\mathbf{x}}$, $J_{y,z}\rightarrow -J_{y,z}$ and $J_x\rightarrow J_x$, and so the Hamiltonian (2.2) is invariant. Denoting the eigenstates of J_z by $|m\rangle_z$, or simply $|m\rangle$, we also have that under this operation, $|m\rangle\rightarrow|-m\rangle$ up to an m-independent phase factor. Thus the Hamiltonian breaks up into two disjoint spaces V_+ and V_-, spanned by the states $|m,+\rangle$ and $|m,-\rangle$, where

$$|m,\pm\rangle=2^{-1/2}(|m\rangle\pm|-m\rangle) \ . \tag{2.3}$$

(If J is an integer and $m=0$, $|0,+\rangle\equiv|0\rangle$, and $|0,-\rangle$ does not exist.) It then follows that as the field u_0 is changed, the energy levels of states belonging to the V_+ and V_- subspaces can cross. The only question is if the lowest two states do this, and if so, how often. To answer this, let us first put $\lambda=0$. The Hamiltonian is then

$$\mathcal{H}_{C,0}=\frac{1}{J^2}J_x^2-\frac{2u_0}{J}J_x \ . \tag{2.4}$$

This is diagonal in the J_x basis. Writing the J_x eigenstate with eigenvalue k as $|k\rangle_x$, we have for the energy

$$E_{k,0}=k(k-2Ju_0)/J^2 \ . \tag{2.5}$$

These curves are drawn in Fig. 1(a) for $J=2$ and $u_0\geq 0$. It is obvious that as u_0 changes from -1 to 1, the lowest eigenvalue changes in the sequence $k=-J,-J+1,\ldots,J-1,J$, giving $2J$ field values where it is doubly degenerate, i.e., where the tunnel splitting vanishes.

Let us now turn on λ. By the symmetry of $\mathcal{H}_C$ mentioned earlier, it follows that not all of the crossings for $\lambda=0$ will become anticrossings. [See Fig. 1(b).] It is easy to see that the states $|k\rangle_x$ belong to $V_\pm$ as follows:

$$\begin{aligned} &k=J,J-2,\ldots\in V_+ \ , \\ &k=J-1,J-3,\ldots\in V_- \ . \end{aligned} \tag{2.6}$$

In particular, since the crossings between the lowest two states for $\lambda=0$ occur between neighboring values of k, they persist when $\lambda\neq 0$. This proves the desired result.

We can also see that the vanishing of the tunnel splitting is destroyed if the field is slightly off the x axis, as $\mathcal{H}_C$ no longer has any rotation symmetry. If the misalignment is small, then as long as $\mathbf{H}$ is in the x-y plane, we have two degenerate classical minima, but the amplitudes of the two main interfering trajectories have unequal magnitudes, and we expect that the tunnel splitting will oscillate without going to zero. The degree of misalignment which can be tolerated is quite small, however, and we shall calculate it for the MQT problem, where the same situation arises. If the field also has a z component, the degeneracy is spoiled, and the problem is closer to one of MQT, which we discuss in the next section.

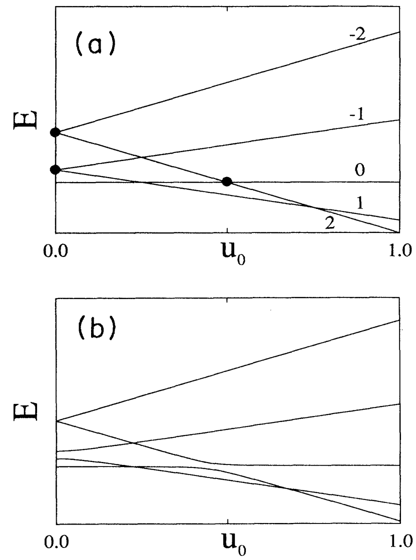

FIG. 1. (a) Energy levels of the MQC Hamiltonian (2.2) for $J=2$ and $\lambda=0$. The levels are labeled by their J_x eigenvalues. The heavy black dots indicate crossings that are eliminated when $\lambda\neq 0$. (b) Same as (a), but with $\lambda=0.2$. The splitting between the two uppermost curves when $u_0=0$ is not zero, but cannot be resolved on the scale of this figure.

III. THE MQT PROBLEM

In this section we will study the problem when a field is applied to our particle along $-\hat{\mathbf{z}}$. This problem has so far[4] been formulated as one of classic MQT,[17] i.e., escape from a metastable state into a continuum of states in which the range of values available to the coordinate or tunneling variable is unbounded. In the case of the magnetic particle, the tunneling variable is the moment $\mathbf{M}$, and this is clearly bounded, and the energy spectrum is discrete if the particle is regarded as isolated. It is therefore desirable to study how the metastable state decays when this restriction, i.e., the finiteness of the range of values available to $\mathbf{M}$, is taken into account. In so doing, we shall gain a clearer understanding of the conclusion of an oscillatory escape rate in Ref. 9 and the conditions required to observe this oscillation.

To orient the discussion, we will first consider the simpler analogous problem for a particle in a one-dimensional double well (see Fig. 2) with unequal minima.[22] We will discuss how escape should be properly defined by coupling the system to a bath. We will find that the bounded or discrete character of the deep well states is irrelevant if the dissipation is large. By contrast, if the dissipation is sufficiently weak, we will find that this system also shows oscillations in the escape rate as the bias V_0 between the wells is varied. In this case, the interesting question is the period (in V_0) of these oscillations. With this background, we will then study the isolated magnetic particle quasiclassically and obtain the oscillation period. We will then study the sensitivity of the oscillations to misalignment of the field and conclude with a discussion of the decay width of the excited levels in the deeper well.

A. Particle in an asymmetric one-dimensional double well

Let us consider a particle moving in a well as shown in Fig. 2, and let it have eigenstates as depicted.[22] We suppose first that V_0 is such that the exact eigenstate $|\psi_0\rangle$ is localized mostly in the left-hand well and that the $|\psi_n\rangle$, $|\psi_{n+1}\rangle$, etc., states are localized mostly in the right-hand well. Suppose now that the particle is prepared at $t=0$ in a state $|\chi_i\rangle \approx |\psi_0\rangle$ and orthogonal to the other predominantly left-well states. It is physically obvious that it cannot escape into the deeper well for ever.[18] Formally, we can show this as follows. Up to an over all phase, the amplitude for finding the particle in $|\chi_i\rangle$ at a time t is

$$A(t)=|c_0|^2+\sum_{n\neq 0}|c_n|^2 e^{-i(E_n-E_0)t/\hbar}, \tag{3.1}$$

where $c_n=\langle\psi_n|\chi_i\rangle$, and we have separated the largest term $|c_0|^2$ from the sum. Writing $|c_0|^2=1-\delta$, where $\delta \ll 1$, we have $|A(t)|\geq 1-2\delta>0$ for all t.

To define a decay or an escape rate, we must provide a mechanism for energy loss. In classical terms, a particle oscillating in the right well with some amplitude should eventually be able to come to rest. Let us again prepare the system in the state $|\psi_0\rangle$ at $t=0$ and ask for the probability $P(t)$ to find it in the same state at a latter time t. We can understand the essential behavior of $P(t)$ as a function of the damping strength and bias without constructing or studying detailed models for the damping. Let us first imagine that the barrier between the two wells is high so that we can regard the two wells as uncoupled.

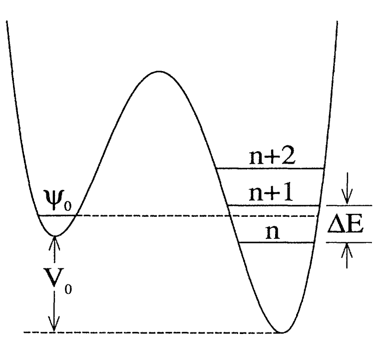

FIG. 2. An asymmetric double well, showing some of the energy levels.

The state $|\psi_0\rangle$ is then the ground state of the left well and cannot decay. Damping will induce a small real shift[23] of the energy ϵ_0, but we are not interested in its value and we simply absorb it in the definition of ϵ_0. On the other hand, the states $|\psi_n\rangle$, $|\psi_{n+1}\rangle$, etc., in the right well will decay, and the energy levels $\epsilon_n, \epsilon_{n+1}, \ldots$ will acquire widths $\gamma_n, \gamma_{n+1}, \ldots$. (The shifts of the real part can again be viewed as having been absorbed in the definitions of ϵ_n, ϵ_{n+1}, etc.) Let us now turn on the coupling between the wells and write the amplitude per unit time (which could be calculated by WKB methods, e.g.,) to tunnel from $|\psi_0\rangle$ to $|\psi_n\rangle$ as $-i\Delta_{0n}/\hbar$. Note that dissipation will reduce the value of Δ_{0n}, but we can again view this effect as having been included in the definition of Δ_{0n}. We assume that the initial state has essentially zero overlap with any excited states in the left well, and in this way we arrive at the effective Hamiltonian

$$\begin{pmatrix} \epsilon_0 & \Delta_{0n} & \Delta_{0,n+1} & \cdots \\ \Delta_{0n} & \epsilon_n - i\gamma_n/2 & 0 & \\ \Delta_{0,n+1} & 0 & \epsilon_{n+1}-i\gamma_{n+1}/2 & \\ \vdots & & & \ddots \end{pmatrix}. \tag{3.2}$$

Let us also assume for the moment that $\gamma_n \gg \Delta_{0n}$, etc. This is likely to hold in most situations, as the Δ's are tunneling matrix elements and thus exponentially small. None of the energy denominators are then small, and the eigenvalue $E_0 \approx \epsilon_0$ can be found by perturbation theory. The decay rate, in particular, is given by

$$\Gamma_0=-2\,\mathrm{Im}E_0=\sum_n \Delta_{0n}^2 \frac{\gamma_n}{(\epsilon_0-\epsilon_n)^2+\gamma_n^2/4}. \tag{3.3}$$

This is an entirely unsurprising result. If we recall that the density of states $g(\epsilon)$ in the right well is given by

$$g(\epsilon)=\frac{1}{2\pi}\sum_n \frac{\gamma_n}{(\epsilon-\epsilon_n)^2+\gamma_n^2/4}, \tag{3.4}$$

then Eq. (3.3) is precisely what one would write by analogy with the current across a tunnel junction between two normal metals.

Equation (3.3) shows that as the bias V_0 is varied, the decay rate will peak whenever one of the levels ϵ_n coincides with ϵ_0. The distance between peaks, or the period of oscillation, is the energy level difference ΔE between neighboring levels. The peaks will be completely washed out, however, if γ_n is comparable to or bigger than ΔE. In other words, the decay maps out the density of intermediate states in the deep well. If the levels are broadened so much that they begin to overlap, then individual levels are no longer discernible, and we effectively decay into a structureless continuum. The important quantity thus is the degree of damping of large amplitude oscillations in the deep well.

The second point is that as long as $\gamma_n \gg \Delta_{0n}$, there is no shift of order Δ_{0n} in the real part of E_0; i.e., there is no coherent flip-flop between the left and right wells, even when ϵ_0 coincides with ϵ_n. To see this, let us put $\epsilon_n=\epsilon_0$ in Eq. (3.2), in which case we can keep only the 2×2 block in the upper left corner. The eigenvalues of this block are

$$\epsilon_0 - \frac{i\gamma_n \pm \sqrt{-\gamma_n^2 + 16\Delta_{0n}^2}}{4} . \tag{3.5}$$

If $\gamma_n \gg 4\Delta_{0n}$, the eigenvalue $E_0 \approx \epsilon_0 - 2i\Delta_{0n}^2/\gamma_n$. There is no shift in the real part, and the imaginary part is in accordance with Eq. (3.3). On the other hand, if $\gamma_n \ll 4\Delta_{0n}$, the energy eigenvalues are

$$\epsilon_0 \pm \Delta_{0n} - i\gamma_n/4 , \tag{3.6}$$

from which it follows that we obtain a damped resonance with

$$P(t) \approx \cos^2(\Delta_{0n} t) e^{-\gamma_n t/2} . \tag{3.7}$$

The absence of a MQC resonance when $\gamma_n \gg 4\Delta_{0n}$ can be also be qualitatively understood as follows. Imagine that the particle is prepared in the state $|\psi_0\rangle$. It tunnels out at some later time, and the resulting wave packet makes one back and forth transit of the deep well. Because of dissipation, though, the particle returns to the vicinity of the exit point with an energy that is decreased by an amount of order at least γ_n. Since $\gamma_n \gg \Delta_{0n}$, the wave packet on return can no longer mix strongly with the state $|\psi_0\rangle$, and there is little probability for it be retrapped in the initial state.

We thus arrive at the following picture for the time-dependent behavior of an asymmetric double well system: (a) If $\gamma_n = 0$, we are dealing with an unphysical closed system, and there is no escape as $t \to \infty$. (b) If $0 < \gamma_n \ll 4\Delta_{0n}$, we get damped coherent oscillations such as those in Eq. (3.7) if $|\epsilon_0 - \epsilon_n| \lesssim 2\Delta_{0n}$, while we get essentially exponential decay with the rate (3.3) otherwise. In this case we get dramatic changes in the behavior as the basis is swept through a resonance. (c) If $4\Delta_{0n} \lesssim \gamma_n \ll |\epsilon_n - \epsilon_{n+1}|$, there is never any coherent oscillation between the wells, but the decay rate, again given by Eq. (3.3), shows oscillations as the bias is varied. (d) If $|\epsilon_n - \epsilon_{n+1}| \ll \gamma_n$, the oscillations in the decay rate are also washed out.

The different conclusions of Refs. 8 and 9 regarding the MQT escape rate thus hold depending on whether one is case (d) or (c), respectively. Case (b) is essentially MQC and extremely difficult to obtain in this author's view, for reasons well discussed elsewhere.[19–21]

B. The isolated magnetic particle

Let us now return to our magnetic particle. To the Hamiltonian (1.1) we add a term for a field along $-\hat{\mathbf{z}}$, giving

$$\mathcal{H}_T = k_1 J_x^2 + k_2 J_y^2 + g\mu_B H J_z . \tag{3.8}$$

(The subscript T stands for tunneling.) This Hamiltonian is invariant under 180° rotations about $\hat{\mathbf{z}}$,[24] and so it is best to work in the J_z basis $\{|m\rangle\}$. The energy eigenstates again divide into two disjoint subspaces $V_\pm$, with

$$\begin{aligned} m &= J, J-2, \ldots \in V_+ , \\ m &= J-1, J-3, \ldots \in V_- . \end{aligned} \tag{3.9}$$

To understand the eigenstates of $\mathcal{H}_T$, let us write its mean value in the coherent state $|\theta,\phi\rangle$ as $E(\theta,\phi)$. Dropping terms of order J compared to J^2, we have

$$E(\theta,\phi) = (k_1\cos^2\phi + k_2\sin^2\phi) J^2 \sin^2\theta + g\mu_B H J \cos\theta . \tag{3.10}$$

This energy is sketched in the easy (y-z) plane in Fig. 3, along with a few energy levels. Let us first suppose that the lowest-energy state that is localized primarily in the shallow well, $|\psi_0\rangle$, is not too close in energy to any of the states in the deep well. It is apparent that $|\psi_0\rangle \simeq |J\rangle \in V_+$. It is also plausible (and we shall show below) that the states in the deep well alternatively belong to V_+ and V_-. As H is increased, the states in the deep well will move down in energy, and different states will come into resonance with $|\psi_0\rangle$. Since the latter can only mix with the V_+ states, however, it follows that the interval ΔH between successive resonances is given by

$$\Delta H = (E_{n+2} - E_n)/2g\mu_B J = \Delta E / g\mu_B J . \tag{3.11}$$

For the particle of the previous subsection, of course, the energy interval between successive resonances would be ΔE, not $2\Delta E$.

We can now state the sense in which Chudnovsky and DiVincenzo's suggestion[9] of an oscillating escape rate is true: As the bias magnetic field is varied, the mixing or resonance between the state $|\psi_0\rangle$ and the states of the deep well takes place with an energy interval that is twice what one would get for a particle tunneling through a spatial barrier. Every other resonance is absent.

But, as discussed in Sec. III A, to go from the above result to an oscillation in the true *escape rate* requires including dissipation and showing that the deep well levels are not broadened by more than $2\Delta E$. Before doing this, we show the V_+, V_- alternation of the states in the deep well and also calculate ΔE. We also note, as in Sec. II, that this oscillation is a consequence of the rotational symmetry of $\mathcal{H}_T$, and we will find below the amount of misalignment of the field required to destroy it.

To show the alternation of levels, let us replace k_1 by k_2 in Eq. (3.8). $\mathcal{H}_T$ is then diagonal in the J_z basis, and the energies in the deep well are ordered according to the

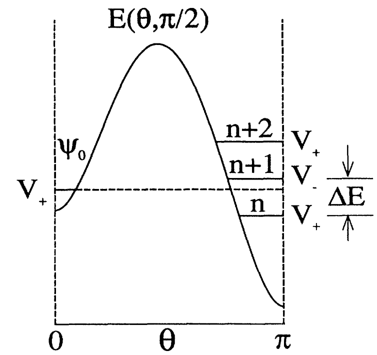

FIG. 3. Magnetic particle MQT potential $E(\theta,\phi)$ in the easy plane, showing energy levels as in Fig. 2, along with the subspace to which they belong.

m values and, consequently, alternately belong to V_+ and V_-. We now turn on the perturbation $(k_1-k_2)J_x^2$. Levels belonging to V_+ and V_- can now cross, but since $\mathcal{H}_T$ possesses no symmetries other than the 180° rotation about $\hat{\mathbf{z}}$, these crossings are accidental and cannot all happen at the same value of k_1-k_2. Indeed, continuity arguments suggest that the crossings will only occur for states with energies near the top of the barrier. Thus, except for isolated "hiccups," the desired alternation of levels in the deep well is proved.[25]

To find the energy level difference ΔE, we exploit the correspondence principle to relate it to ω, the frequency of periodic motion with energy near E_n in the deep well, by $\Delta E=\hbar\omega$. To find ω, we regard $E(\theta,\phi)$ [Eq. (3.10)] as a Hamiltonian with conjugate variables ϕ and $J\hbar\cos\theta$. This gives a semiclassical equation of motion:

$$-J\hbar\sin\theta\frac{d\phi}{dt}=\frac{\partial E(\theta,\phi)}{\partial\theta}\ , \tag{3.12}$$

integrating which gives

$$\frac{2\pi}{\omega}=\int_0^{2\pi}\frac{d\phi}{\dot{\phi}}=J\hbar\int_0^{2\pi}d\phi\left[\frac{\partial E}{\partial(\cos\theta)}\right]^{-1} . \tag{3.13}$$

For $E=g\mu_B HJ$ in Eq. (3.10), the required integration is elementary, and we get

$$\Delta E=\hbar\omega=2k_1J[\lambda(1-h)(1-\lambda h)]^{1/2}\ , \tag{3.14}$$

where $\lambda=k_2/k_1$ as before and $h\equiv H/H_c$; $H_c=2k_2J/g\mu_B$ is the value of the field at which the barrier in Fig. 3 disappears completely.

Combining Eqs. (3.11) and (3.14), we get

$$\frac{\Delta H}{H_c}=\frac{1}{J}[(1-h)(\lambda^{-1}-h)]^{1/2}\ . \tag{3.15}$$

The limit $h\approx 1$ is particularly interesting, for otherwise the barrier between the two wells is large and the tunneling rate is very small. The WKB exponent in the tunneling rate varies as $J(1-h)^{3/2}$ as $h\to 1$, and for $J=10^4$, we require $1-h\simeq 10^{-2}$ to get an appreciable tunneling rate.[26] Taking these values of J and h, $\lambda=O(1)$, and $H_c=10^4$ G, we get $\Delta H\sim 0.1$ G. One may hope to get larger ΔH (~ 1 G) for smaller particles, with $J\lesssim 10^3$, but of course that makes the experimental problem harder.

This calculation also answers another question asked by Chudnovsky and DiVincenzo. For half integer J and $H=0$, there is no tunneling between $\hat{\mathbf{z}}$ and $-\hat{\mathbf{z}}$ by Kramers' theorem. This effect will be destroyed when the field is large enough to make the ground state in one well degenerate with the first excited state in the other well, i.e., when $g\mu_B HJ=\Delta E(h=0)$. From Eq. (3.14), it follows that the tolerable limit on the field is $2H_c/\lambda^{1/2}J$, which agrees with Ref. 9 in its J dependence and also fixes the scale factor.

C. Field misalignment

An important issue in any attempt at experimental verification of the forbidden transitions is field misalignment. Let us suppose that the field has a small component $H_y \ll H_z$ along $\hat{\mathbf{y}}$. This is clearly the more dangerous component as compared to H_x, as the y direction is the medium direction of magnetization and the escape takes place in the y-z plane. We wish to obtain a criterion for H_y which will ensure that the selection rule is not wiped out.

Although it is possible to formally solve this problem for any value of H_z, we will concentrate on the limit when $H_z\simeq H_c$, as otherwise the WKB exponent S_0 for tunneling out of the initial state becomes very large for any reasonably sized particle. For a particle with total spin $J=10^4$ and $\lambda=0.1$ (nearly easy plane anisotropy), e.g., $S_0>30$ if $1-h>0.023$. In this case the exit angle is very small, and $J_z\simeq J$. We can replace the commutator $[J_x,J_y]=i\hbar J_z$ by the position-momentum commutator $[P,Q]=-i\hbar$, if we identify $J_x=-P$ and $J_y=JQ$. Adding the field along $\hat{\mathbf{y}}$, the Hamiltonian (3.8) is equivalent to the following Hamiltonian for a particle moving in one dimension:

$$\mathcal{H}_T=(k_1-k_2)P^2+k_2J^2\left[(1-h)Q^2-\frac{1}{4}Q^4-2\frac{H_y}{H_c}Q\right] . \tag{3.16}$$

Let us denote the amplitudes for tunneling out of the $Q=0$ state along the $+Q$ and $-Q$ directions by A_+ and A_-, respectively, and also introduce the ratio $\zeta=|A_-/A_+|$. When $H_y=0$, $A_+=A_-\propto\exp(-8U/3\hbar\omega_0)$, where U is the barrier height and ω_0 is the small oscillation frequency near $Q=0$. (Note that the tunneling *rate* $\sim|A_\pm|^2$, and so $S_0=16U/3\hbar\omega_0$.) For $H_y\neq 0$, the leading corrections to $A_\pm$ are given by still regarding the potential as a quartic for positive and negative Q, but with unequal barrier heights $U_\pm$. Writing $\Delta U=U_- - U_+$, we get

$$A_-/A_+=\exp(-8\Delta U/3\hbar\omega_0)\ . \tag{3.17}$$

The selection rule can be understood as due to interference between the semiclassical trajectories which exit from the $J_z=J$ state along positive and negative J_y, and meet in the x-z plane in the classically allowed region. When $H_y=0$, $\zeta=1$, and perfectly constructive or destructive interference occurs if the final state belongs to V_+ or V_-, respectively. If $H_y\neq 0$ and $\zeta\ll 1$, the interference will be practically nil, and we will get substantial tunneling into the V_- state. It is easy to show that

$$\Delta U=4[2(1-h)]^{1/2}k_2J^2(H_y/H_c)\ , \tag{3.18}$$

$$\omega_0=2J[k_2(k_1-k_2)]^{1/2}(1-h)^{1/2}\ . \tag{3.19}$$

Let us take $\zeta>\zeta^*$ as an operational criterion for the observability of a selection rule based oscillation in Γ, where ζ^* is a small number, but not too close to zero. A good choice is $\zeta^*=0.1$.[27] This leads to the following tolerance criterion for the misalignment:

$$\frac{H_y}{H_c}<\frac{3\sqrt{2}}{32J}\left[\frac{k_1-k_2}{k_2}\right]^{1/2}|\ln\zeta^*|\ . \tag{3.20}$$

Taking $\zeta^*=0.1$, we find that even with $k_1=10k_2$, the field must be aligned to better than 0.1 mrad for $J=10^4$.

In absolute terms, with $H_c = 10^4$ G, we require $H_y < 1$ G. A field of this size could easily be produced by a few paramagnetic impurities lying within, say, 5 Å of the particle's surface, and so this point must be carefully attended to in any actual attempt to see this effect.

A different criterion for the observability of forbidden transitions can be obtained as follows. Let us consider the two states that are localized in the deep well in the absence of any field along $\hat{\mathbf{y}}$ and that are closest in energy to the initial, metastable state, and let us denote them by $|\psi_-\rangle$ and $|\psi_+\rangle$ depending on which subspace V_- or V_+ they belong to. Let us now consider $\mathcal{H}' = -g\mu_B J_y H_y$ as a perturbation to Eq. (3.8) and write

$$\mathcal{H}'_{+-} = \langle \psi_+ | \mathcal{H}' | \psi_- \rangle \ . \tag{3.21}$$

If this matrix element is comparable to or exceeds the energy difference ΔE in Eq. (3.14), then the states $|\psi_-\rangle$ and $|\psi_+\rangle$ will be strongly mixed by the perturbation, and all transitions will be allowed.

To find the matrix element $\mathcal{H}'_{+-}$, let us write the general eigenfunction of $\mathcal{H}_T$ as $|\psi\rangle = \sum_m c_m |m\rangle$. Schrödinger's equation is then equivalent to a three-term recursion relation for the c_m's which can be solved by a discrete WKB method.[28] The quasiclassical wave function for periodic motion with energy E within the deep well is given by

$$c_m = \left[\frac{2\omega(E)}{\pi v(m)} \right]^{1/2} \cos\Phi_E(m) \ , \tag{3.22}$$

where

$$\Phi_E(m) = \int_{m_t}^{m} \phi(m';E)dm' + \pi m_t + \frac{\pi}{4} \ . \tag{3.23}$$

Further, $m = J\cos\theta$, $v(m) = dm/dt$ along the classical trajectory, $\omega(E)$ is the frequency of this trajectory, m_t is a turning point, and $\phi(m;E)$ can be found from the energy conservation law (3.10).

The matrix element $\mathcal{H}'_{+-}$ can now be found as a Fourier component of the classical motion following Ref. 29. We leave the details as an exercise for the reader and only quote the result:

$$|\mathcal{H}'_{+-}| = \frac{2k_2 J^2}{\pi} \frac{H_y}{H_c} \int_0^{\pi/2\omega} \sin\theta(t)\sin\phi(t)\sin(\omega t)\omega \, dt \ . \tag{3.24}$$

(We have chosen the initial condition $\phi = 0$ at $t = 0$.) For $E = g\mu_B HJ$ in Eq. (3.10), this integral can be found in terms of complete elliptic integrals. The general result is uninteresting, but in the limit $H \simeq H_c$, the condition $|\mathcal{H}'_{+-}| < \Delta E$ can be shown to be

$$\frac{H_y}{H_c} < \frac{\pi}{2J} \frac{[k_1(k_1-k_2)]^{1/2}}{k_2} \ . \tag{3.25}$$

This is a slightly weaker condition than Eq. (3.20) and should therefore be discarded in favor of the latter.

D. Decay width of the intermediate state

We now qualitatively discuss the decay of the intermediate state. In semiclassical terms, this state corresponds to a precession of the net magnetic moment about $\hat{\mathbf{z}}$ (Ref. 30) and is entirely analogous to a state with a large number of $k = 0$ magnons in a ferromagnetic resonance experiment. The problem of its decay is thus conceptually similar to the well-studied one of ferromagnetic relaxation.[31] Nevertheless, standard golden rule calculations of its width are rather difficult. Neither the interaction matrix elements nor the density of states factors can be simply calculated for a small particle. The second problem is particularly serious. Of the various relaxation processes relevant to ferromagnetic resonance in insulators, the ones important now are magnon-magnon scattering. Because of small particle size, however, the creation of a $k \neq 0$ magnon may be energetically forbidden. For a particle of linear size L, the smallest nonzero wave vector is $k_{\min} \simeq \pi/L$. Let us suppose that a magnon can be created with an attendant decrease of n in J_z. The change in energy [denoted δE to avoid confusion with Eq. (3.14)] is then given in field units by

$$\frac{\delta E}{\hbar\gamma} \simeq \frac{1}{2} H_{\mathrm{ex}} k_{\min}^2 a^2 - nH \ , \tag{3.26}$$

where H_{ex} is the exchange field and a is the lattice spacing. Taking $H_{\mathrm{ex}} = 5 \times 10^6$ G, the first term in Eq. (3.26) is 1–5×10^4 G for a particle with 10^4–10^5 moments. The important interactions correspond to $n=1$ or $n=2$, and so with $H \simeq H_c = 10^4$ G, the second term in Eq. (3.26) is comparable to the first, and we conclude that we are close to the threshold for magnon creation and may be either above or below it. A reliable estimate of this threshold is complicated by several factors. First, the exchange fields H_{ex} are not known precisely and may be quite nonuniform. In particular, they may be considerably weaker near the particle surface, and given the large surface-to-volume ratio, the lowest excitations may be closer to surface magnons. Second, in bulk, the magnon energy depends on demagnetization factors and magnon ellipticity or direction of propagation. Indeed, without these effects, the magnon dispersion relation would have zero bandwidth, and $k = 0$ magnons would never decay into other magnons. Some vestige of these effects must surely persist for all small particles, but continuum calculations are suspect, as are simple-minded assumptions for the shape of the particle. Third, anisotropy fields, which can be of comparable magnitude to demagnetizing fields, are also unknown and inhomogeneous. If $\delta E > 0$, it follows that the decay of the intermediate state may itself require tunneling through a barrier. This possibility has important implications for magnetic relaxation in an assembly of small particles, as it suggests a means for slow nonexponential decay.

Let us now discuss the decay mechanisms themselves. In bulk matter, $k = 0$ magnons can decay via two-magnon processes, which do not conserve crystal momentum and thus require some disorder, or via three- and four-magnon processes, which are rooted in the dipolar interaction and which underlie the Suhl instabilities.

Consider two-magnon processes first. For a particle with only 10^4 moments, a significant fraction ($\sim 10\%$) is at the surface, and it seems quite likely that the local anisotropy and exchange fields will differ significantly from the bulk value and from moment to moment. The exchange interaction, however, conserves total spin and cannot scatter the homogeneous precession mode, and so we must look to the anisotropy fields. Haas and Callen[31] note that variations in single-ion anisotropy, arising from differences in the local environment of the magnetic ion, have a characteristic rms value of 10^5 G in ferrospinels. This number is quite comparable to the equivalent energy difference in Eq. (3.26), so that even if the number of inhomogeneous states into which the homogeneous precession mode can decay is small, and even if the inhomogeneous states themselves have small decay widths, the uniform mode can mix appreciably with the nonuniform ones, effectively spreading out the density of accessible intermediate states in the deep well in Fig. 3.

The three- and four-magnon processes appear to be less important. Their interaction matrix elements are characterized by dipolar fields, which are of order 10^4 G and thus weaker than the random anisotropy fields considered above. Magnon-phonon and electromagnetic interactions are even less important. Phonons within the particle lie too high in energy to be excited. Stresses near the particle surface could give rise to low-lying local phonon modes, but their quantitative effect is hard to evaluate. The particle as a whole can emit elastic dipole radiation, and the corresponding energy width γ_{elast} can be estimated following Ref. 32. We get

$$\frac{\gamma_{\text{elas}}}{\Delta E} \simeq \frac{2\pi J(\omega)}{\hbar\omega} \sim \frac{v_0^2 K_{ME}\omega^2}{\hbar c_s^3} , \tag{3.27}$$

where $J(\omega)$ is the spectral density found in Ref. 32, v_0 is the particle volume, K_{ME} is the strain-induced anisotropy, and c_s is a sound speed. Taking a particle radius of 50 Å, $K_{ME} = 10^3$ ergs/cm^3, and $c_s = 3\times 10^5$ cm/sec, we get $\gamma_{\text{elas}}/\Delta E \sim 10^{-3}$. The electromagnetic radiative width can also be found and is utterly negligible.

IV. CONCLUSIONS

In summary, we have shown that the oscillations in the MQC tunnel splitting and MQT escape rate suggested in Refs. 8 and 9 are a consequence of simple symmetries of the Hamiltonian, although these oscillations can be easily destroyed by misaligning fields and, in the case of MQT, by short lifetimes of the intermediate states. We have concentrated on the MQT problem as it seems more amenable to experimental realization to us. Better calculation of the decay times of the intermediate state is highly desirable and seems to us to be an especially promising area for further work, with interesting implications for magnetization decay beyond the narrow question of MQT.

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[26]One may worry that the barrier disappears as $h \to 1$. However, the WKB answer for Γ, the metastable ground-state escape rate, is remarkably robust—it is accurate to a few percent even when n, the number of levels in the metastable well, is as little as 4. See, e.g., A. I. Larkin and Yu. N. Ovchinnikov, Zh. Eksp. Theor. Fiz. **91**, 318 (1986) [Sov. Phys. JETP **64**, 185 (1986)], or the discussion following Eqs. (3.6) and (3.7) in K. S. Chow, D. A. Browne, and V. Ambegaokar, Phys. Rev. B **37**, 1624 (1988). For our problem, $n \gtrsim J(1-h)^{3/2}/2(\lambda^{-1}-1)^{1/2}$. If $J=10^4$, $n \simeq 4$ is attainable even when $1-h$ is of order 10^{-2}.

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