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Topologically Quenched Tunnel Splitting in Spin Systems without Kramers' Degeneracy.

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Abstract. – Spin systems can undergo quantum tunneling with classically degenerate minima. In some cases, the amplitudes from symmetry-related trajectories can interfere destructively, quenching the tunneling rate. This quenching need not be related to Kramers' degeneracy. This is shown by studying a problem (relevant to macroscopic quantum phenomena in ferromagnetic particles) with biaxial symmetry and an external field. The tunnel splitting is found to oscillate with the field, vanishing at certain values. Spin path integrals are used to prove Kramers' theorem, and an extension in which a subset of the energy levels is systematically doubly degenerate.

Problems in which spin degrees of freedom tunnel between various states have been the subject of many studies over the last few years. At very low temperatures, *e.g.*, the total moment of a small (~ 100 Å diameter) ferromagnetic particle may resonate between degenerate easy axes, or out of a metastable easy direction [1], thus providing other instances of macroscopic quantum phenomena (MQP). (See [2] for reviews of MQP.) Other problems are tunneling of the order parameter in antiferromagnetic particles [3], and hopping of holes in a Hubbard model near half-filling [4].

The above calculations are best done using spin–coherent-state path integrals [5, 6]. The kinetic part of the action in such path integrals, which is variously known as the Wess-Zumino, Berry-phase, or Chern-Simons term, has a topological content [6], and gives rise to a phase in the imaginary-time transition amplitude. Very recently, Loss, DiVincenzo and Grinstein [7], and von Delft and Henley [8] have pointed out that in spin systems with high symmetry, the phases from symmetry-related paths connecting two classically degenerate minima can cancel each other exactly, yielding a vanishing tunnel splitting. The cancellations they find depend on the value of the spin, just as in the closely related phenomenon of the Haldane gap in one-dimensional antiferromagnets [9].

In this paper I wish to emphasize that the above cancellations, which I shall refer to as topological quenching, are unrelated to Kramers' degeneracy. I shall show this by means of a specific example for macroscopic quantum coherence (MQC), where the ground-state tunnel splitting vanishes even when Kramers' theorem is inapplicable. One can also understand

Kramers' degeneracy in terms of interfering paths, however, and this can obscure the distinction between the two effects. I will prove Kramers' theorem using path integrals, and then extend the proof to systems where a subset of the energy levels is systematically doubly degenerate. An example from ref. [8] is found to be precisely in this class.

The MQC example I consider provides, in fact, the main motivation for this paper. The system of interest is a small (~ 100 Å diameter), single-domain, ferromagnetic particle at a temperature well below the anisotropy gap. Spin waves are then frozen out, and the only interesting degree of freedom left is the orientation $\hat{\boldsymbol{M}}$ of the total magnetization $\boldsymbol{M}$. The Hamiltonian for the particle is then given by the magnetic anisotropy energy, and, since the individual moments move in unison under the specified conditions, we can write this in terms of a single large spin, J, proportional to $|\boldsymbol{M}|$. Keeping the leading-order terms in the anisotropy energy [10], we have

$$\mathscr{H} = k_1 J_z^2 + k_2 J_y^2 \,, \tag{1}$$

where $k_1 > k_2 > 0$ are proportional to the anisotropy coefficients, and we have taken the easy, medium, and hard axes as x, y, and z, respectively.

Since the Hamiltonian (1) has degenerate minima along the $\boldsymbol{x}$-axis, the spin can resonate between these two directions [1], providing a case of MQC. If J is half-integral, however, since eq. (1) is time-reversal invariant, by Kramers' theorem *all* eigenstates, and in particular the ground states, are doubly degenerate. This implies that the tunnel splitting, Δ, between the minima at $\hat{\boldsymbol{x}}$ and $-\hat{\boldsymbol{x}}$ vanishes [11, 12]. While this is a surprising result at first, it follows from a familiar theorem, and can be easily understood by noting that $\mathscr{H}$ in eq. (1) divides into two identical disjoint blocks in the J_z basis.

If we now apply a magnetic field along the hard axis ($\hat{\boldsymbol{z}}$) of our particle, the Hamiltonian becomes

$$\mathscr{H} = k_1 J_z^2 + k_2 J_y^2 - \gamma H J_z \,, \tag{2}$$

where γ is proportional to the g-factor. This is no longer time-reversal invariant, and Kramers' theorem is inapplicable. Nevertheless, we shall show that tunneling can still be forbidden in some cases. As H is increased, the topological phase difference between the relevant interfering trajectories changes from $2\pi J$ to 0, and the tunnel splitting oscillates, vanishing whenever the phase difference is an odd multiple of π. The effect is not confined to half-integer spins.

Let us first briefly review spin–coherent-state path integrals. (For details, see [5, 6].) By denoting a coherent state by $|\hat{\boldsymbol{n}}\rangle$, the imaginary-time propagator for spin J is given by

$$\langle \hat{\boldsymbol{n}}_2 | \exp[-\mathscr{H} T] | \hat{\boldsymbol{n}}_1 \rangle = \int [\mathrm{d}\hat{\boldsymbol{n}}] \exp[-S[\hat{\boldsymbol{n}}]\,, \tag{3}$$

where S, the Euclidean action, is given by

$$S = \int_{-T/2}^{T/2} [-iJ(1 - \cos\theta)\,\dot{\phi}(\tau) + E(\hat{\boldsymbol{n}})]\,\mathrm{d}\tau\,, \tag{4}$$

where (θ, ϕ) are the polar coordinates of $\hat{\boldsymbol{n}}$, $E(\hat{\boldsymbol{n}}) = \langle \hat{\boldsymbol{n}} | \mathscr{H} | \hat{\boldsymbol{n}} \rangle$, and the path integral (3) is over paths satisfying $\hat{\boldsymbol{n}}(-T/2) = \hat{\boldsymbol{n}}_1$, $\hat{\boldsymbol{n}}(T/2) = \hat{\boldsymbol{n}}_2$. The first term in eq. (4), which we write as $-iJA[\hat{\boldsymbol{n}}]$, has a simple geometrical interpretation [6], and is crucial in the ensuing discussion. For a closed loop, $A[\hat{\boldsymbol{n}}]$ is the area enclosed by the path on the unit sphere. This is only defined modulo 4π, but since $\exp[-i4\pi J] = 1$ for both integer and half-integer J, this ambiguity is physically irrelevant. For a path connecting $\hat{\boldsymbol{n}}_1 \neq \hat{\boldsymbol{n}}_2$, $A[\hat{\boldsymbol{n}}]$ is determined only up to a choice of gauge, but its variation is gauge independent, and clearly shows its geometrical meaning:

$$\delta A = \int \hat{\boldsymbol{n}} \cdot ((\mathrm{d}\hat{\boldsymbol{n}}/\mathrm{d}\tau) \times \delta\hat{\boldsymbol{n}})\,\mathrm{d}\tau . \tag{5}$$

This equation, and the formula $\hat{\boldsymbol{n}} \cdot (\mathrm{d}\hat{\boldsymbol{n}}/\mathrm{d}\tau) = 0$, can be used to obtain the classical equation of motion for $\hat{\boldsymbol{n}}$:

$$iJ\frac{\mathrm{d}\hat{\boldsymbol{n}}}{\mathrm{d}\tau} = -\hat{\boldsymbol{n}} \times \frac{\partial E}{\partial \hat{\boldsymbol{n}}}, \tag{6}$$

which is the imaginary-time equation for Larmor precession in the effective magnetic field, $\partial E(\hat{\boldsymbol{n}})/\partial\hat{\boldsymbol{n}}$. Note that if eq. (6) holds, $\mathrm{d}E/\mathrm{d}\tau = 0$, *i.e.* energy is conserved.

For large spin, and large time T, the path integral (3) can be evaluated using instanton or steepest-descent methods [6]. (See ref. [13] for examples.) The integral is dominated by the least-action paths, obtained by solving eq. (6) subject to the boundary conditions. Energy conservation forces the path to have complex coordinates in general, and the action will have both real and imaginary parts (denoted S_r and S_i), the latter of which can only arise from the real part of A: $S_\mathrm{i} = -iJA_\mathrm{r}$. Destructive interference can occur when there are multiple paths between $\hat{\boldsymbol{n}}_1$ and $\hat{\boldsymbol{n}}_2$, for which the real part of the action, S_r, and the fluctuation determinants (1), D, are equal because of symmetry, and when the topological phases add to give zero. For two equivalent paths this happens when J times the area enclosed by the paths is an odd multiple of π.

Let us now apply this formalism to the MQC Hamiltonian (2). For $H < H_\mathrm{c} = 2k_1J/\gamma$, $E(\hat{\boldsymbol{n}})$ has degenerate minima at $\theta = \theta_0$, $\phi = 0, \pi$, where $\cos\theta_0 = H/H_\mathrm{c} \equiv u_0$. (See fig. 1.) Writing $K_1 = k_1J^2$, $K_2 = k_2J^2$, and adding a constant, we can write

$$E(\theta, \phi) = K_1(\cos\theta - \cos\theta_0)^2 + K_2\sin^2\theta\sin^2\phi . \tag{7}$$

We can evaluate the classical action (and its imaginary part in particular) for paths connecting the minima without solving for the paths themselves. Writing $u = \cos\theta$, $\lambda = K_2/K_1$, energy conservation gives

$$u = \frac{u_0 + i\lambda^{1/2}\sin\phi(1 - u_0^2 - \lambda\sin^2\phi)^{1/2}}{(1 - \lambda\sin^2\phi)} . \tag{8}$$

It suffices to consider fields which are such that $u_0^2 < 1 - \lambda$. One can then take $\phi(\tau)$ to be entirely real, and the square root in eq. (8) is real too.

It is clear from symmetry that there are two instanton paths, $(\theta_\pm(\tau), \phi_\pm(\tau))$, which wind around the hard axis in opposite directions. (See fig. 1.) We will take $\phi_-(\tau) = -\phi_+(\tau)$, with $\phi_\pm(-\infty) = 0$, and $\phi_\pm(\infty) = \pm\pi$. The real part of A for these paths is given by

$$A_{\mathrm{r},\pm} = \int_0^{\pm\pi}\left(1 - \frac{u_0}{1 - \lambda\sin^2\phi}\right)\mathrm{d}\phi = \pm\pi(1 - u_0(1-\lambda)^{-1/2}) . \tag{9}$$

By adding the contributions from both paths, the tunnel splitting is found to be $D\exp[-S_\mathrm{r}]\,|\cos\Phi(H)|$, where $\Phi(H) = J(A_{\mathrm{r},+} - A_{\mathrm{r},-})/2$ (2). (The difference $(A_{\mathrm{r},+} - A_{\mathrm{r},-})$ is

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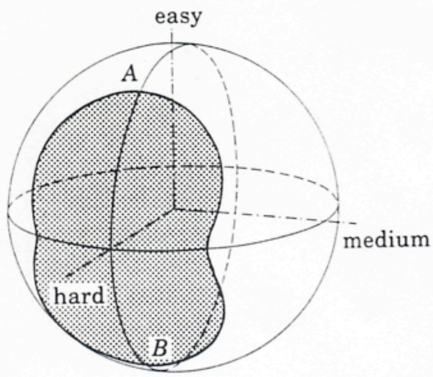
(1) This determinant is obtained by expanding the action $S[\hat{\boldsymbol{n}}]$ to second order in deviations from the classical path, and integrating over these deviations [11, 13].

(2) The integrals for S_r can also be done. We omit the answer as it is not relevant here.



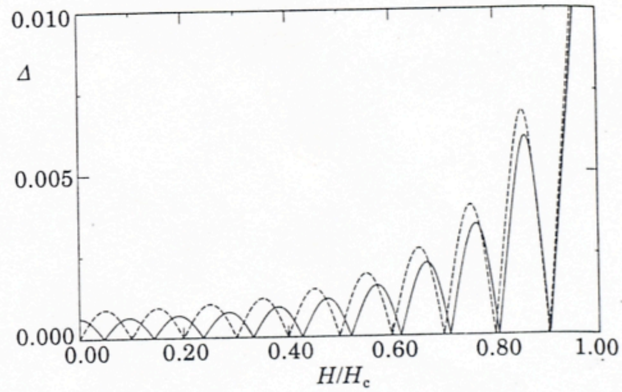


Fig. 1

Fig. 2

Fig. 1. – Unit sphere showing degenerate minima (A and B), joined by instanton paths (heavy lines). The hard axis (z-axis in the text) is rotated toward the reader for ease of visualization. The tunnel splitting is topologically quenched whenever the shaded area is $n\pi/J$, for odd n.

Fig. 2. – Numerically computed splitting, Δ, between the two lowest-energy states of the Hamiltonian (2) as a function of the applied field, with $K_1 = 1$, $\lambda = 0.1$. —— $J = 10$, - - - $J = 19/2$.

the shaded area in fig. 1.) The tunnel splitting is thus quenched whenever

$$H/H_c = (1-\lambda)^{1/2}(J - n - 1/2)/J\,, \tag{10}$$

where n is an integer. Including negative and zero values, there are $2J$ values of the field for which the splitting is quenched, whether J is an integer or half-integer. This last result will clearly hold for Hamiltonians more complicated than eq. (2) as long as they possess the necessary symmetry. This effect clearly has no analogue in the Josephson-junction systems [2b].

In fig. 2 we plot the numerically computed difference between the two lowest energies, *i.e.* the tunnel splitting, Δ, as a function of the field for the Hamiltonian (2), for $J = 19/2$ and $J = 10$. The oscillations are evident. The fields where Δ vanishes are almost, but not exactly, given by eq. (10). Further, when $\Delta = 0$, the excited states are not all doubly degenerate, showing again that topological quenching is unrelated to Kramers' degeneracy.

In addition to the problem of *coherence* between degenerate states, one may also consider macroscopic quantum *tunneling*, *i.e.* the decay out of a metastable state. (As emphasized by Leggett [2a], the two phenomena are physically very different, particularly from the viewpoint of experimental feasibility.) To the Hamiltonian of eq. (1) one could add a term describing a field along $-\hat{\boldsymbol{x}}$, creating a metastable state along $\hat{\boldsymbol{x}}$. The decay rate is computed by putting $\hat{\boldsymbol{n}}_1 = \hat{\boldsymbol{n}}_2 = \hat{\boldsymbol{x}}$ in eq. (3). The WKB exponent is given by the action for the so-called «bounce» paths, which start at $\hat{\boldsymbol{x}}$, touch a classical turning point, and return to $\hat{\boldsymbol{x}}$ [13]. (Again, there may be more than one path because of symmetry.) One can see that the topological phase from such a path cannot quench the decay rate as follows. It is always possible to choose one of the coordinates of this path, say $\theta(\tau)$, to be real: $\theta_i(\tau) = 0$. Then $\phi(\tau)$ will be complex, but the real part of the trajectory, (θ_r, ϕ_r), will go from $\hat{\boldsymbol{x}}$ to the turning point, and then retrace itself back to $\hat{\boldsymbol{x}}$. (This can be seen most easily by working to lowest order in $\varepsilon = (1 - H/H_{cr})$, where H_{cr} is the field at which the initial state becomes classically unstable. Examples of this procedure can be found in ref. [13] for many different symmetries. In each case the assertions made here can be seen to be true.) The area enclosed by the bounce, and the topological phase A_r, thus vanish for *each* bounce individually, and there is no quenching of the decay rate.

We now show how Kramers' theorem can be derived from spin path integrals for single-spin problems. The generalization to many particles with orbital degrees of freedom is straightforward. The key point is that if $\mathscr{H}$ is time-reversal invariant, then

$$\langle -\hat{\boldsymbol{n}}_0 \,|\exp[-\mathscr{H}T]|\hat{\boldsymbol{n}}_0\rangle = 0 \qquad \forall \hat{\boldsymbol{n}}_0, T, \tag{11}$$

if J is a half-integer. To see this, we first note that it suffices to consider only real paths in the path integral for eq. (11), and then note that we can pair *any* path $\hat{\boldsymbol{n}}(\tau)$ with $-\hat{\boldsymbol{n}}(-\tau)$. (Note also that both members of the pair tend to $\pm\hat{\boldsymbol{n}}_0$ at $\pm T/2$.) The potential energy term $E(\hat{\boldsymbol{n}})$ contributes equally to the action (4) for both paths, since time-reversal invariance is equivalent to $E(\hat{\boldsymbol{n}}) = E(-\hat{\boldsymbol{n}})$. To calculate the topological term, note simply that the two paths together divide the unit sphere into halves, so the area enclosed is 2π. Thus, the paths add exactly out of phase for half-integral J, proving eq. (11).

Let us now expand the coherent states in terms of (unnormalized) energy eigenstates $|\psi_E(\hat{\boldsymbol{n}})\rangle$ as

$$|\hat{\boldsymbol{n}}\rangle = \sum_E |\psi_E(\hat{\boldsymbol{n}})\rangle. \tag{12}$$

Let us define the time reversal operator Θ so that $\Theta|\hat{\boldsymbol{n}}\rangle = |-\hat{\boldsymbol{n}}\rangle$, and write $\Theta|\psi_E(\hat{\boldsymbol{n}})\rangle = |\bar{\psi}_E(\hat{\boldsymbol{n}})\rangle$ (the energy must be the same). The issue is whether $|\bar{\psi}_E(\hat{\boldsymbol{n}})\rangle \propto |\psi_E(\hat{\boldsymbol{n}})\rangle$. Writing $\hat{\boldsymbol{n}}$ for $\hat{\boldsymbol{n}}_0$, and substituting eq. (12) and its time-reversed counterpart in (11), we get

$$\sum_E \langle\bar{\psi}_E(\hat{\boldsymbol{n}})|\psi_E(\hat{\boldsymbol{n}})\rangle \exp[-ET] = 0. \tag{13}$$

Since this equation holds for all T, each term in the sum must vanish. Thus, either $|\psi_E(\hat{\boldsymbol{n}})\rangle$ and $|\bar{\psi}_E(\hat{\boldsymbol{n}})\rangle$ are distinct, or $|\psi_E(\hat{\boldsymbol{n}})\rangle = 0$. The latter cannot be true for all $\hat{\boldsymbol{n}}$, but since eq. (11) holds for all $\hat{\boldsymbol{n}}$, every energy state must have a degeneracy of even order.

A similar argument shows that levels must be paired in other situations. We require that there be two states $|\hat{\boldsymbol{n}}_1\rangle$ and $|\hat{\boldsymbol{n}}_2\rangle$ for which $\langle \boldsymbol{n}_1|\exp[-\mathscr{H}T]|\hat{\boldsymbol{n}}_2\rangle = 0$ for all T (and not just $T\to\infty$), and which are connected by a symmetry transformation Q which commutes with $\mathscr{H}$, *i.e.* $Q|\hat{\boldsymbol{n}}_1\rangle = |\hat{\boldsymbol{n}}_2\rangle$. (For time reversal, $Q = \Theta$, and $|\hat{\boldsymbol{n}}_2\rangle = |-\hat{\boldsymbol{n}}_1\rangle$.) The analogue of eq. (13) then holds, but only for isolated orientations, $\hat{\boldsymbol{n}}_1$ and $\hat{\boldsymbol{n}}_2$. The possibility that $\psi_E(\hat{\boldsymbol{n}}_1) = 0$ cannot now be excluded, so the correct theorem is that all energy states must either be doubly degenerate, or have zero-projection on $|\hat{\boldsymbol{n}}_1\rangle$ and $|\hat{\boldsymbol{n}}_2\rangle$.

An example discussed by von Delft and Henley [8] is precisely of this type. They consider a Hamiltonian with m-fold symmetry about the z-axis, and show that $\langle -\hat{z}|\exp[-\mathscr{H}T]|\hat{z}\rangle$ vanishes unless $2J$ is an integer multiple of m, as follows. In the path integral, *any* path $\hat{\boldsymbol{n}}(\tau)$ can be combined with $m-1$ other paths obtained by successive $2\pi/m$ rotations about the z-axis. The area between two successive paths is $4\pi/m$, and since S_r is the same for all paths by symmetry, the topological phase factors add to produce the necessary cancellation.

For $m = 3$, von Delft and Henley suggest an energy with trigonal symmetry:

$$E(\theta,\phi) = B_1\cos 3\phi\sin^3\theta + B_2\sin^2\theta. \tag{14}$$

This also has $\theta\to\pi-\theta$ symmetry, however, and this provides the necessary connection between $|\hat{z}\rangle$ and $|-\hat{z}\rangle$ for our theorem on pairing to apply. The energy (14) arises (up to a constant) from a Hamiltonian

$$\mathscr{H} = B_1'(J_+^3 + J_-^3) + B_2'(J_x^2 + J_y^2), \tag{15}$$

where $J_\pm = J_x \pm iJ_y$, and $B_i' \propto B_i$. By writing $\mathscr{H}$ in the J_z basis, it is easy to see that for $2J \neq km$, where k is an integer, about 2/3 of the states are indeed doubly degenerate, and the non-degenerate states are orthogonal to $|\pm\hat{z}\rangle = |j, \pm j\rangle$ (3). It would be interesting to know if this type of pairing arises in the study of crystal field splittings.

We conclude by returning to our magnetic MQC problem and making a few qualitative remarks on the role of dissipation, such as might be caused by eddy currents, phonons [14], or nuclear spins [15]. Dissipation generally increases the real part of the action, S_r, reducing the magnitude of the tunneling rate, and is one of the reasons why MQP, especially coherence, are so hard to see. In the present context we would expect it to diminish the effects of interference among symmetry-related paths, leading to (partial) *un*quenching of the tunnel splitting. A quantitative investigation of this effect is an open and interesting problem.

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(3) For eq. (15) one can show more than is implied by the theorem. There is systematic pairing even for $2J = km$, as $\mathscr{H}$ divides into three blocks for all J, two of which are identical, or paired. The states $|\pm\hat{z}\rangle$ belong to the paired blocks for $2J \neq km$, and to the unpaired block for $2J = km$.

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