

Destructive Quantum Interference in Spin Tunneling Problems

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In some spin tunneling problems there are several different but symmetry-related tunneling paths that connect the same initial and final configurations. The topological phase factors of the corresponding tunneling amplitudes can lead to destructive interference between the different paths, so that the total tunneling amplitude is zero. In the study of tunneling between different ground-state configurations of the *kagomé*-lattice quantum Heisenberg antiferromagnet, this occurs when the spin s is half odd integer.

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The problem of calculating the rate at which a quantum spin system tunnels between its different low-energy states has been of interest in various different contexts [1]. The tunneling amplitude is usually calculated by setting up a coherent-spin-state path integral and analytically continuing to imaginary time ($t \rightarrow -i\tau$); the leading contribution can be found using the method of steepest descent. The phase of the tunneling amplitude depends on a topological phase, variously called a Berry phase or a Wess-Zumino phase.

As was first pointed out by Haldane in his work on extended quantum spin systems in one and two dimensions, this phase can give rise to qualitative differences between systems with integer and half-odd-integer spin s , due to quantum interference between topologically distinct paths of a continuous unit vector field $\Omega(\mathbf{x}, t)$ [2].

In this Letter, we point out that destructive quantum interference between different tunneling paths can also occur in somewhat simpler contexts, for systems that involve only a small number of individual spins. When there are several different symmetry-related tunneling paths connecting two degenerate classical ground states of a spin system, the topological phase can lead to destructive quantum interference between the respective tunneling amplitudes, and hence to a total tunneling amplitude that is exactly zero. The occurrence of such a cancellation has a geometric interpretation and depends on the value of the spin s . We illustrate this with two examples: The first involves a Hamiltonian with an m -fold symmetry axis. The second concerns tunneling amplitudes between different degenerate ground-state configurations of a *kagomé*-lattice quantum Heisenberg antiferromagnet; these amplitudes are zero if s is half odd integer, but nonzero if s is integer.

The tunneling amplitude: In tunneling problems, the customary object of study is the imaginary time transition amplitude from an initial state $|i\rangle$ to a final state $|f\rangle$. For a spin system, this can be written as a coherent-spin-state path integral [3]:

$$U_{fi} \equiv \langle f | e^{-\hat{\mathcal{H}}t/\hbar} | i \rangle = \int \mathcal{D}\Omega e^{-\mathcal{S}/\hbar}, \quad (1)$$

where $\mathcal{S} = \int d\tau \mathcal{L}$ is the Euclidean action and $\mathcal{D}\Omega$ is the measure of the path integral. For the special case of a single spin, the Euclidean Lagrangian is

$$\mathcal{L} = -i\hbar s \dot{\phi}(1 - \cos\theta) + \mathcal{H}(\phi, \theta). \quad (2)$$

The coordinates (ϕ, θ) label the coherent spin state $|\phi, \theta\rangle$ for a particle with spin s , and may be associated with a unit vector \mathbf{n} in the $(\phi, \theta) \equiv \Omega$ direction. The origin of the topological first term, sometimes called a Wess-Zumino term, is clearly explained in [3(b)]. The dot on $\dot{\phi}$ means ∂_τ . The "semiclassical" Hamiltonian is the expectation value $\mathcal{H} \equiv \langle \phi, \theta | \hat{\mathcal{H}} | \phi, \theta \rangle$ of the operator $\hat{\mathcal{H}}$. For simplicity, we shall consider only the case where $|i\rangle$ and $|f\rangle$ are "classically degenerate ground states" (in the sense that $\langle i | \hat{\mathcal{H}} | i \rangle = \langle f | \hat{\mathcal{H}} | f \rangle$ are the smallest possible expectation values of $\hat{\mathcal{H}}$), and are separated by an energy barrier.

The above path integral can be evaluated by the method of steepest descent:

$$U_{fi} = \sum_l \mathcal{N}^{(l)} e^{-\mathcal{S}_0^{(l)}/\hbar} \equiv \sum_l U_{fi}^{(l)}, \quad (3)$$

where

$$\mathcal{N}^{(l)} = \int \mathcal{D}(\Omega - \bar{\Omega}^{(l)}) e^{-(\delta^2 \mathcal{S}^{(l)} + \delta^3 \mathcal{S}^{(l)\dots})/\hbar}. \quad (4)$$

Here $\mathcal{S}_0^{(l)}$ is the action evaluated along the l th "tunneling path," which is a solution to the Lagrangian equations of motion and will be denoted by overlined variables, e.g., $(\bar{\phi}^{(l)}(\tau), \bar{\theta}^{(l)}(\tau))$. The index l allows for the possibility of several different symmetry-related tunneling paths. The prefactors $\mathcal{N}^{(l)}$ measure the effects of fluctuations around the l th tunneling path. Tunneling problems are characterized by the fact that the coordinates in general acquire imaginary parts along the tunneling path (else it is not possible to satisfy the requirement that the Hamiltonian be a conserved quantity along the path). Consequently, the various $\mathcal{S}_0^{(l)}$ can have nonzero imaginary parts. Quantum interference, and possibly complete cancellation (so that $U_{fi} = 0$), can thus occur between the amplitudes of the different paths.

Geometric interpretation of phase: The conserved energy $E_0 = \mathcal{H}(\bar{\Omega}^{(l)})$ along the tunneling paths that connect the degenerate states $|i\rangle$ and $|f\rangle$ may be set equal to zero without loss of generality. Hence $S_0^{(l)}$ is completely determined by the topological term in \mathcal{L} . This term has a well-known geometrical interpretation [3(b)], which we now discuss.

Let $\Omega(\tau) = (\phi(\tau), \theta(\tau))$ be a purely real, *closed*, non-self-intersecting, smooth path in spin space. The area on the unit sphere enclosed by this path is given by

$$A = \int d\tau \dot{\phi}(1 - \cos\theta), \quad (5)$$

modulo 4π , depending on which of the two oriented areas (the “inside” or the “outside” of the closed path) one

$$(S_0^{(l)} - S_0^{(l')})/\hbar = -is \left[\int d\tau \text{Re}[\dot{\bar{\phi}}^{(l)}](1 - \text{Re}[\cos\bar{\theta}^{(l)}]) - \int d\tau \text{Re}[\dot{\bar{\phi}}^{(l')}] (1 - \text{Re}[\cos\bar{\theta}^{(l')}]) \right]. \quad (7)$$

Terms such as $\text{Re}[\text{Im}[\]]$ do not appear due to Eq. (6). We have assumed that $\text{Im}[\dot{\bar{\phi}}^{(l)}]\text{Im}[\cos\bar{\theta}^{(l)}] = 0$ (same for $l \leftrightarrow l'$), for the following reason: The conserved energy condition, $\mathcal{H}(\bar{\Omega}^{(l)}(\tau)) = 0$, can be solved to find, for example, $\bar{\theta}^{(l)}$ in terms of $\bar{\phi}^{(l)}$. The dependent variable $\bar{\theta}^{(l)}$ will be a complex function of the independent variable $\bar{\phi}^{(l)}$, which can be taken to be real, so that $\text{Im}[\dot{\bar{\phi}}^{(l)}] = 0$. A similar argument works if one chooses to write $\bar{\theta}^{(l)}$ in terms of $\bar{\phi}^{(l)}$ [1(c)].

Now, let $A_{ll'}$ be either one of the two oriented areas on the unit sphere bounded by the loop formed by the two paths $\text{Re}[\bar{\Omega}^{(l)}]$ and $\text{Re}[\bar{\Omega}^{(l')}]$. Then, by Eq. (5), the relative phase Eq. (7), which is sometimes called a Berry phase [2(b)], reduces to

$$(S_0^{(l)} - S_0^{(l')})/\hbar = -isA_{ll'}. \quad (8)$$

The 4π ambiguity in $A_{ll'}$ is irrelevant, since $\exp(-i4\pi s) = 1$ for any spin s .

If $sA_{ll'}$ is an odd multiple of π , the amplitudes for the two paths interfere destructively, so that $U_{fi}^{(l)} + U_{fi}^{(l')} = 0$. In the simplest case where $\bar{\Omega}^{(l)}$ and $\bar{\Omega}^{(l')}$ are the only two tunneling paths, this means that the *total* amplitude U_{fi} is zero. Note that this result does not depend on the detailed dynamics of the tunneling motion. In particular, it is not necessary to know the imaginary parts of the tunneling paths $\bar{\Omega}^{(l)}$ explicitly, as long as symmetry arguments can be invoked to assert that the relations (6) hold. It may happen that symmetry ensures that the prefactors $\mathcal{N}^{(l)}$ are equal to *all* orders of the steepest descent method, which is an expansion in powers of $1/s$ (and not merely to the lowest order that is usually employed). In this case the cancellation, if it happens, is exact to all orders in $1/s$.

Clearly, the occurrence of destructive interference depends crucially on the value of s . This could create com-

considerations.

Now consider two (of the possibly many) tunneling paths, say $\bar{\Omega}^{(l)}(\tau)$ and $\bar{\Omega}^{(l')}(\tau)$, not necessarily purely real, connecting $|i\rangle$ and $|f\rangle$. Suppose that some symmetry of the Hamiltonian ensures that the absolute values of the two tunneling amplitudes $U_{fi}^{(l)}$ and $U_{fi}^{(l')}$ of Eq. (3) are equal, i.e., that

$$\text{Re}[S_0^{(l)}] = \text{Re}[S_0^{(l')}], \quad \text{and} \quad \mathcal{N}^{(l)} = \mathcal{N}^{(l')}. \quad (6)$$

Intuitively speaking, this situation arises when the local neighborhoods of the two tunneling paths are, for symmetry reasons, identical for the two paths, so that the local shapes and sizes of the barriers (which determine the $\text{Re}[S_0^{(l)}]$) and the local fluctuations around the tunneling paths (which determine the $\mathcal{N}^{(l)}$), are identical. Then $U_{fi}^{(l)}$ and $U_{fi}^{(l')}$ differ at most by a phase, namely,

lications in the analysis of the tunneling behavior of, for example, a small ferromagnetic particle with effective spin ns (as has been considered in [1(c)]), where n is the number of spins in the particle, since the effective spin is then n dependent.

Turning on an external magnetic field can in general affect the occurrence of destructive interference by changing the initial conditions (i.e., $|i\rangle$ and $|f\rangle$) and the area $A_{ll'}$ enclosed between different tunneling paths, and by destroying the symmetry that ensures that the absolute values of the amplitudes for all tunneling paths are the same.

We now present two examples where destructive interference does occur.

Hamiltonian with m -fold symmetry axis: Consider a single spin s . Suppose that the Hamiltonian has an easy axis (say the z axis, $\theta = 0$), around which it has m -fold rotational symmetry: $\mathcal{H}(\phi, \theta) \equiv \mathcal{H}(\phi + 2\pi/m, \theta)$ for all ϕ . Suppose that $|i\rangle = |\theta = 0\rangle$ and $|f\rangle = |\theta = \pi\rangle$ correspond to the two degenerate classical ground-state configurations. Clearly, if $\bar{\Omega}^{(0)} = (\bar{\phi}^{(0)}, \bar{\theta}^{(0)})$ is a tunneling path from $|i\rangle$ to $|f\rangle$, so are the paths $\bar{\Omega}^{(l)} = (\bar{\phi}^{(0)} + 2\pi l/m, \bar{\theta}^{(0)})$, for $l = 0, \dots, m-1$ (see Fig. 1). By symmetry, all $\text{Re}[S_0^{(l)}]$ are equal to each other, as are all $\mathcal{N}^{(l)}$. Furthermore,

$$(S_0^{(0)} - S_0^{(l)})/\hbar = -isA_{0l} = -is4\pi l/m, \quad (9)$$

because the area on the unit sphere enclosed between the real parts of any two neighboring paths is, by symmetry, necessarily equal to $4\pi/m$. The total amplitude is thus:

$$U_{fi} = \mathcal{N}^{(0)} e^{-S_0^{(0)}/\hbar} \sum_{l=0}^{m-1} e^{i4\pi ls/m} = \delta_{2s, km} m \mathcal{N}^{(0)} e^{-S_0^{(0)}/\hbar},$$

where the δ function is nonzero only if $2s$ is an integer

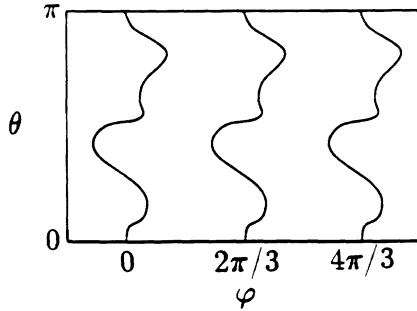


FIG. 1. Three equivalent tunneling paths for a Hamiltonian with threefold rotation symmetry on a “wrapped-open” unit sphere.

multiple of m .

A realization of the above scenario, with $m = 3$, is afforded by a Hamiltonian with the symmetry of a trigonal prism, i.e., threefold rotational symmetry around the \mathbf{z} axis and reflection symmetry in the x - y plane. An explicit example would be

$$\mathcal{H} = J_1 \cos 3\phi \sin^3 \theta + J_2 \sin^2 \theta, \quad (10)$$

with $0 < J_1 \leq J_2$. The unit vectors \mathbf{z} and $-\mathbf{z}$ define the two classical ground-state configurations.

Spin tunneling in the kagomé lattice: Our second example concerns spin tunneling events in a 2D quantum Heisenberg nearest-neighbor antiferromagnet on a *kagomé* lattice (Fig. 2) [4]. The Hamiltonian is taken to be

$$\mathcal{H} = s^2 J \sum_{\langle i,j \rangle} \mathbf{n}_i \cdot \mathbf{n}_j \quad (J > 0), \quad (11)$$

where $s\mathbf{n}_i \equiv \langle \phi_i, \theta_i | \hat{\mathbf{s}}_i | \phi_i, \theta_i \rangle$ is the “classical” spin [see Fig. 3(a)]. Any configuration in which the spins on *each* triangle minimize their energy by assuming a coplanar configuration, with relative angles of 120° [see Fig. 3(b)], is a classical ground state. Therefore there are macroscopically many degenerate classical ground states. Generally, both quantum and thermal fluctuations are expected to lift some of the degeneracies, thereby inducing magnetic ordering by selecting particular configurations (“order from disorder”). For example, spin wave expansions about various ground-state configurations have shown that maximally coplanar configurations, in which *all* spins in the lattice lie in the same plane (let this “reference plane” define $\phi = 0$ and $\phi = \pi$), have lower zero-point energies than any other configurations [4(b),(d)].

On the other hand, tunneling between different degenerate ground-state configurations competes with “order from disorder” selection effects, because it tends to drive the system into a superposition of degenerate states, rather than selecting a particular one. As the simplest example of a tunneling event on the *kagomé* lattice, we consider the so-called “weather vane mode” (see Fig. 2): the six spins of an *ABABAB* hexagon in one maximally

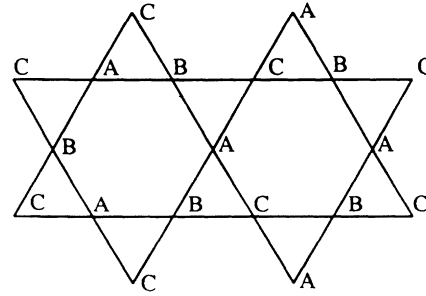


FIG. 2. An *ABABAB* hexagon of spins (on the left) in a coplanar ground-state configuration.

coplanar ground state, $|i\rangle$, rotate synchronously by 180° around the \mathbf{z} axis (defined by the C spins), to end up as a *BABABA* hexagon in another maximally coplanar ground state, $|f\rangle$, while all other spins remain fixed. Because of the above-mentioned spin wave selection effects, there is a “coplanarity barrier,” say $J_b f(\phi)$, to this type of motion; the barrier shape function $f(\phi)$ obeys $f(\phi) = f(-\phi)$, by reflection symmetry in the reference plane.

To study the hexagon tunneling event, we consider the following Euclidean Lagrangian:

$$\mathcal{L} = \sum_{j=1}^6 -i\hbar s \dot{\phi}_j (1 - \cos \theta_j) + J_b f(\phi_{av}) + s^2 J \sum_{j=1}^6 [\mathbf{n}_j \cdot \mathbf{n}_{j+1} + \mathbf{n}_j \cdot \mathbf{z} + \mathbf{n}_{j+1} \cdot \mathbf{z} + \frac{3}{2}]. \quad (12)$$

The index j is defined modulo 6, and $\phi_{av} \equiv \frac{1}{6} \sum_{j=1}^6 (\phi_j - \phi_j(0))$. We take $J \gg J_b > 0$. The $s^2 J \mathbf{n}_j \cdot \mathbf{z}$ terms (interaction with C spins) and the phenomenological $f(\phi_{av})$ coplanarity barrier are assumed to be the only ways the other spins in the lattice, which are assumed to remain fixed, influence the six spins on the hexagon.

To minimize the cost of the dominant $s^2 J$ term, the hexagon spins are expected to rotate collectively, maintaining mutual near coplanarity. Indeed, it can be shown [5] that the hexagon tunneling problem can be mapped onto a simple model problem, defined by the Lagrangian

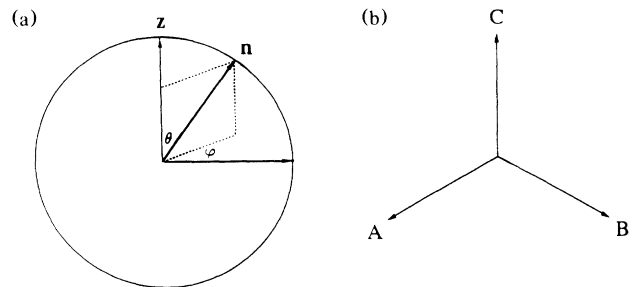


FIG. 3. (a) The unit vector $\mathbf{n} = (\phi, \theta)$. (b) Type *A*, *B*, and *C* spins on a triangle of the *kagomé* lattice.

$$\mathcal{L} = -i\hbar 6s\dot{\phi}(1 - \cos\theta) + 12s^2 J(\cos\theta - \frac{1}{2})^2 + J_b \sin^2\phi,$$

involving only a single (collective) spin degree of freedom with an effective spin of $6s$. For present purposes, however, the following observations suffice (discussed in detail in [5]): Because of the reflection symmetry about the reference plane, there are two possible tunneling paths, to be denoted by $(\bar{\phi}^\pm, \bar{\theta}^\pm)$; they differ from each other only in the *direction* of the $\bar{\phi}$ rotations and satisfy $\bar{\phi}_j^-(\tau) = -\bar{\phi}_j^+(\tau)$, for $j = 1, \dots, 6$. Reflection symmetry ensures that $\text{Re}[S_0^+] = \text{Re}[S_0^-]$ and $\mathcal{N}^+ = \mathcal{N}^-$. Along both tunneling paths, every $\bar{\phi}_j^\pm$ is purely real. To satisfy $\mathcal{H} = 0$ during the tunneling event, each θ_j develops a time-dependent imaginary part (which vanishes in $|i\rangle$ and $|f\rangle$), but $\text{Re}[\cos\theta_j]$ maintains the value it has in $|i\rangle$ and $|f\rangle$, namely, $\text{Re}[\cos\theta_j] = -\frac{1}{2}$, $j = 1, \dots, 6$.

Thus, for each of the six spins, the real part of the tunneling path is a contour of constant $\text{Re}[\cos\theta_j] = -\frac{1}{2}$, with $\text{Re}[\bar{\phi}_j^+]$ (or $\text{Re}[\bar{\phi}_j^-]$) changing from 0 to π (or $-\pi$) for (+) or (-) paths. For each spin, the area enclosed between the (+) and (-) paths is thus equal to π , giving for the six spins a total phase difference of $i6\pi s$ between the amplitudes for a (+) or (-) event. It follows that the total tunneling amplitude becomes

$$U_{fi} = \mathcal{N}^+ e^{-S_0^+/\hbar} (1 + e^{i6\pi s}) \\ = \begin{cases} 2\mathcal{N}^+ e^{-S_0^+/\hbar} & \text{if } s \text{ is integer,} \\ 0 & \text{if } s \text{ is half odd integer.} \end{cases} \quad (13)$$

Similarly, consider any larger closed "loop" of alternating A and B spins within a ground-state configuration. It can be proven that any such loop contains $4n+2$ spins (n is some integer) [5]. Again one can study the tunneling between two configurations that only differ by $\phi_l \rightarrow \phi_l + \pi$ (i.e., $A \leftrightarrow B$) for each spin on the loop. The relevant phase between (+) and (-) paths will be $i\pi(4n+2)s$, and for half-odd-integer s , destructive interference again occurs.

The above results have interesting consequences for the ground state of the *kagomé* antiferromagnet [5]: In that subset of parameter space where "order from disorder" selection effects and the competing tunneling effects that favor more disorder are more or less equally important, one might expect interesting integer versus half-odd-integer s effects, reminiscent of those found in 1D antiferromagnetic spin chains [2(a)].

Integer versus half-odd-integer s effects might also make their appearance in exact diagonalization studies of finite-size systems with discrete degeneracies, in the analysis of which spin tunneling methods should be very useful. The simplest realization of such a discrete degeneracy is the J_1 - J_2 square lattice antiferromagnet for $J_2 > J_1/2$, which shows a discrete degeneracy between antiferromagnetic ordering vectors $(1, 0)$ and $(0, 1)$ [6].

The vanishing of tunneling amplitudes obviously implies an exact ground-state degeneracy in the semi-

classical picture. Sometimes, this degeneracy can be shown to exist for *all* eigenstates of the system, on a purely quantum-mechanical level. For example, consider the following toy model for a quantum spin (independently suggested to us by V. Elser): $\mathcal{H} = -\hat{S}_z^2 - a\hat{S}_x^2$. Here $a \ll 1$, and \hat{S}_z and \hat{S}_x are spin operators. The tunneling amplitude between the two classical ground states, $\mathbf{n} = \pm z$ along the two tunneling paths $\bar{\theta}^\pm : 0 \rightarrow \pi$, $\bar{\phi}^+ = 0$, $\bar{\phi}^- = \pi$, is zero for half-odd-integer s (the phase difference is $2\pi s$). On the other hand, \mathcal{H} has time-reversal symmetry and hence all its eigenstates display a twofold Kramers degeneracy for half-odd-integer s . It would be interesting to investigate more generally under what circumstances vanishing semiclassical tunneling amplitudes also imply exact quantum-mechanical degeneracies for all eigenstates.

In conclusion, we have shown that the topological phase factor occurring in spin tunneling amplitudes can have quite striking effects in simple systems involving only a few individual spins. If there are several symmetry-related tunneling paths whose individual tunneling amplitudes have the same absolute value, their topological phases can lead to destructive quantum interference between the paths and a total tunneling amplitude that is zero. The conditions under which this occurs can be interpreted geometrically in terms of the areas on the unit sphere enclosed between the real parts of the various tunneling paths. In quantum antiferromagnets, interesting integer versus half-odd-integer s effects can result from the competition between "order from disorder" selection and tunneling effects.

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