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 [2]. The phase of the tunneling amplitude depends on a topological phase [3].

In this Letter we point out that when there are several different symmetry-related tunneling paths connecting the same degenerate classical ground states, the topological phase can lead to destructive quantum interference between their 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 also present 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 non-zero if $s$ is integer.

For extended quantum spin systems in one and two dimensions, qualitative differences between integer and half-odd-integer spin $s$ have been reported previously [4]. In these papers, quantum interference between topologically distinct paths of a continuous unit vector field $\Omega(x,t)$ leads to interesting $s$-dependencies. Our examples show that similar effects occur for much simpler systems that involve only a small number of individual spins.

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 [2]:

$$U_{fi} \equiv \langle f|e^{-\hat{H}t/\hbar}|i\rangle = \int \mathcal{D}\Omega e^{-S/\hbar}.$$  \hspace{1cm} (0.1)

where $S = \int dt \mathcal{L}$ 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

$$\mathcal{L} = -i\hbar s\dot{\phi}(1 - \cos \theta) + \mathcal{H}(\phi, \theta).$$  \hspace{1cm} (0.2)
The coordinates \((\phi, \theta)\) label the coherent spin state \(|\phi, \theta\rangle\) for a particle with spin \(s\), and may be associated with a unit vector \(n\) in the \((\phi, \theta)\equiv \Omega\) direction. The dot on \(\dot{\phi}\) means \(\partial_{\tau}\). The “semi-classical” Hamiltonian is the expectation value \(H \equiv \langle \phi, \theta|\hat{H}|\phi, \theta\rangle\) of the operator \(\hat{H}\).

For simplicity, we shall consider only the case where \(|i\rangle\) and \(|f\rangle\) are “classically degenerate ground states” (in the sense that \(\langle i|\hat{H}|i\rangle = \langle f|\hat{H}|f\rangle\) are the smallest possible expectation values of \(\hat{H}\)), and are separated by an energy barrier.

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

\[
U_{fi} = \sum_l N^{(l)} e^{-S_o^{(l)}/\hbar} \equiv \sum_l U^{(l)}_{fi}, \tag{0.3}
\]

where \(N^{(l)} = \int D(\Omega - \Omega^{(l)}) e^{-\delta^2 S^{(l)} + \delta^3 S^{(l)}...}/\hbar\).

Here \(S_o^{(l)}\) 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. \((\overline{\phi}^{(l)}(\tau), \overline{\theta}^{(l)}(\tau))\).

The index \(l\) allows for the possibility of several different symmetry-related tunneling paths.

The prefactors \(N^{(l)}\) 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 \(S_o^{(l)}\) can have non-zero 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 \(E_o = H(\Omega^{(l)})\) 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 \(S_o^{(l)}\) is completely determined by the first term of \(\mathcal{L}\). This term has a well-known geometrical interpretation, 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

\[
A = \int d\tau \dot{\phi}(1 - \cos \theta), \tag{0.5}
\]
modulo $4\pi$, depending on which of the two oriented areas (the “inside” or the “outside” of the closed path) one considers.

Now consider two (of the possibly many) tunneling paths, say $\Omega^{(l)}(\tau)$ and $\Omega^{(l')}\left(\tau\right)$, 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. (0.3) are equal, i.e. that

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

(0.6)

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 $\text{Re}[S_o^{(l)}]$’s) and the local fluctuations around the tunneling paths (which determine the $\mathcal{N}^{(l)}$’s), are identical. Then $U_{fi}^{(l)}$ and $U_{fi}^{(l')}$ differ at most by a phase, namely

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

$$- \left. \int d\tau \text{Re}[\phi^{(l')}] \left( 1 - \text{Re}[\cos \theta^{(l')}\left(\tau\right)] \right) \right].$$  

(0.7)

Terms such as $\text{Re}[\ ]\text{Im}[\ ]$ do not appear due to eq. (0.6). We have assumed that $\text{Im}[\phi^{(l)}]\text{Im}[\cos \theta^{(l)}] = 0$ (same for $l \leftrightarrow l'$), for the following reason: The conserved energy condition, $H(\Omega^{(l)}(\tau)) = 0$, can be solved to find, for example, $\theta^{(l)}$ in terms of $\phi^{(l)}$. The dependent variable $\theta^{(l)}$ will be a complex function of the independent variable $\phi^{(l)}$, which can be taken to be real, so that $\text{Im}[\phi^{(l)}] = 0$. A similar argument works if one chooses to write $\phi^{(l)}$ in terms of $\theta^{(l)}$ [5].

Now, let $A_{\mathcal{W}}$ be either one of the two oriented areas on the unit sphere bounded by the loop formed by the two paths $\text{Re}[\Omega^{(l)}]$ and $\text{Re}[\Omega^{(l')}]$. Then, by eq. (0.5), the relative phase eq. (0.7) reduces to

$$(S_o^{(l)} - S_o^{(l')})/\hbar = -isA_{\mathcal{W}}.$$  

(0.8)

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

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If \( sA_{\mu\nu} \) is an odd multiple of \( \pi \), the amplitudes for the two paths interfere destructively, so that \( U_{fi}^{(l)} + U_{fi}^{(l')} = 0 \). In the simplest case where \( \Omega^{(l)} \) and \( \Omega^{(l')} \) 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 \( \Omega^{(l)} \) explicitly, as long as symmetry arguments can be invoked to assert that the relations (0.6) hold. It may happen that symmetry ensures that the prefactors \( N^{(l)} \) 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 complications 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 \( A_{\mu\nu} \) 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 (saying 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 \( \phi \). Suppose that \( |i\rangle = |\theta = 0\rangle \) and \( |f\rangle = |\theta = \pi\rangle \) correspond to the two degenerate classical ground state configurations. Clearly, if \( \Omega^{(0)} = (\phi^{(0)}, \theta^{(0)}) \) is a tunneling path from \( |i\rangle \) to \( |f\rangle \), so are the paths \( \Omega^{(l)} = (\phi^{(0)} + 2\pi l/m, \theta^{(0)}) \), for \( l = 0, \ldots, m - 1 \) (see fig. [1]). By symmetry, all \( \text{Re}[S^{(l)}_0] \) are equal to each other, as are all \( N^{(l)} \). Furthermore,

\[
(S^{(0)}_0 - S^{(l)}_0)/\hbar = -isA_{\mu\nu} = -is4\pi l/m ,
\]

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:

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

where the $\delta$-function is non-zero only if $2s$ is an integer multiple of $m$.

A realization of the above scenario, with $n = 3$, is afforded by a Hamiltonian with cubic anisotropy and a small additional uniaxial anisotropy along the [111] direction (thus 3-fold symmetry around [111]):

$$H = J_1\left(n_x^4 + n_y^4 + n_z^4 - \frac{1}{3}\right) + J_2\left(1 - (n \cdot d)^2\right), \quad (0.10)$$

where $n_x, n_y$ and $n_z$ are the direction cosines of the unit vector $n$ in the $(\phi, \theta)$ direction, and $J_1 \gg J_2 > 0$. The unit vectors $d \equiv \frac{1}{\sqrt{3}}(1,1,1)$ and $-d$ 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) [7]. The Hamiltonian is taken to be

$$\mathcal{H} = s^2J\sum_{\langle ij \rangle} \mathbf{n}_i \cdot \mathbf{n}_j, \quad (J > 0), \quad (0.11)$$

where $\mathbf{s}_i \equiv \langle \phi_i, \theta_i | \mathbf{\hat{s}}_i | \phi_i, \theta_i \rangle$ is the “classical” spin (see fig. 3a). Any configuration in which the spins on each triangle minimize their energy by assuming a coplanar configuration, with relative angles of 120°, (see fig. 3b), is a classical ground state. Therefore there are macroscopically many degenerate classical ground states. Generally fluctuations, both quantum and thermal, 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 [7, 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 “weathervane mode” (see fig. 3): the six spins of an ABABAB-hexagon in one maximally coplanar ground state, $|i\rangle$, rotate synchronously by 180° around the $z$-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. Due to the above-mentioned spin wave selection effects, there is a “coplanarity barrier”, say $J_b(\phi)$, to this type of motion, which, by reflection symmetry in the reference plane, obeys $J_b(\phi) = J_b(-\phi)$.

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

$$
\mathcal{L} = \sum_{j=1}^{6} -i\hbar s\dot{\phi}_j (1 - \cos \theta_j) + J_b \sum_{j=1}^{6} \sin^2 \frac{1}{2} (\phi_j + \phi_{j+1} + \pi) \\
+ s^2 J \sum_{j=1}^{6} [n_j \cdot n_{j+1} + n_j \cdot z + n_{j+1} \cdot z + \frac{3}{2}]. \tag{0.12}
$$

The index $j$ is defined modulo 6. We take $J \gg J_b > 0$. The $s^2 J n_j \cdot z$ terms (interaction with C-spins) and the phenomenological $\sin^2 \phi$ coplanarity barrier [8] 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 [10] that the hexagon tunneling problem can be mapped onto a simple model problem, defined by the Lagrangian

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

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 [10]):

Due to 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 $j = 1, \ldots, 6$. Reflection symmetry ensures that $\text{Re}[S_j^+] = \text{Re}[S_j^-]$ and $\mathcal{N}^+ = \mathcal{N}^-$. Along both tunneling paths, every $\bar{\phi}_j^\pm$ is purely real.
To satisfy $H = 0$ during the tunneling event, each $\theta_j$ 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 $\text{Re}[\cos \theta_j] = -\frac{1}{2} j = 1, \ldots, 6$.

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 $\text{Re}[\varphi_j]$ (or $\text{Re}[\varphi_j]$) changing from 0 to $\pi$ (or $-\pi$) for $(+)$- or $(-)$-paths. For each spin, the area enclosed between the $(+)$- and $(-)$-paths is thus equal to $\pi$, giving for the six spins a total phase difference of $6\pi$ between the amplitudes for an $(+)$- or $(-)$-event. It follows that the total tunneling amplitude becomes

$$U_{fi} = N e^{-S_0/h} (1 + e^{i6\pi s})$$

$$= \begin{cases} 
2N e^{-S_0/h} & \text{if } s \text{ is integer}, \\
0 & \text{if } s \text{ is half-odd-integer}.
\end{cases} \quad (0.13)$$

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) [10]. 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 imply that 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 [4, a]. It would therefore be of interest to find integer-spin realizations of the Kagomé lattice antiferromagnet, since $s$ is half-odd-integer for the two experimental realizations of the Kagomé lattice that have been proposed: the magnetoplumbite $SrCr_{8-x}Ga_{4+x}O_{19}$ ($s = \frac{3}{2}$) and the second layer of $He^3$ atoms on a graphite substrate ($s = \frac{1}{2}$) [11].

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 $(0, 1)$ [12].

The vanishing of tunneling amplitudes obviously implies an exact ground state degen-
eracy 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 am-
plitude between the two classical ground states, $n = \pm z$ along the two tunneling paths
$\bar{\theta}^\pm : 0 \to \pi$, $\bar{\phi}^+ = 0$, $\bar{\phi}^- = \pi$, 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
two-fold Kramers degeneracy for half-odd-integer $s$. It would be interesting to investigate
more generally under what circumstances vanishing semi-classical tunneling amplitudes also
imply exact quantum-mechanical degeneracies for all eigenstates.

In conclusion, we have shown that the topological phase factor occurring in spin tun-
neling amplitudes can have quite striking effects. 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 in-
terpreted 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 ver-
sus half-odd-integer $s$ effects can result from the competition between “order from disorder”
selection and tunneling effects.

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REFERENCES

[1] (a) E. M. Chudnovsky and L. Gunther, Phys. Rev. Lett. 60, 661 (1988); (b) A. Auerbach, B. E. Larson, Phys. Rev. Lett. 66, 2262 (1991); (c) G.-H. Kim and A. Garg, preprint; (d) D. D. Aschwalom et. al., Phys. Rev. Lett. 68, 3092, (1992).

[2] (a) J. R. Klauder, Phys.Rev. D 19, 2349 (1979); J. R. Klauder, in Coherent States, Applications in Physics and Mathematical Physics, ed. J. R. Klauder and B.-S. Skakerstam, World Scient. Publ. (1985); (b) E. Fradkin and M. Stone, Phys. Rev. B 38, 77215 (1988).

[3] This relative phase may be thought of as a Berry phase [4, b].

[4] (a) F. D. M. Haldane, Phys. Lett. A 93, 464 (1983); F. D. M. Haldane, J. Appl. Phys. 57, 3359 (1985); I. Affleck, Nucl. Phys. B 265, 409 (1985); (b) F. D. M. Haldane, Phys. Rev. Lett. 61, 1029 (1988).

[5] Examples of this procedure may be found in [1, c].

[6] Note that due to three reflection planes through this axis there are actually two groups of three tunneling paths each, related by reflection symmetry. The preceding arguments hold for each group separately.

[7] (a) A. B. Harris, C. Kallin, A. J. Berlinsky, Phys. Rev. B 45, 2899 (1992); (b) I. Ritchey, P. Chandra, P. Coleman, preprint; (c) S. Sachdev, Phys. Rev. B 45, 12377 (1992); (d) A. Chubukov, preprint.

[8] For a different class of tunneling events, a $|\sin \phi|$ coplanarity barrier has been found by [7, b]. However, our argument does not depend on the precise form of the barrier, as long as $J_b(\phi) = J_b(-\phi)$.

[9] E. P. Chan and C. L. Henley, unpublished (1991).

[10] J. von Delft and C. L. Henley, to be submitted for publication.

[11] C. Broholm et. al., Phys. Rev. Lett. 65, 3173 (1990); V. Elser, Phys. Rev. Lett. 62,
2405 (1990).

[12] D. Poilblanc et. al., Phys. Rev. B 43 10970 (1991).
FIGURES

FIG. 1. Three equivalent tunneling paths for a Hamiltonian with 3-fold rotation symmetry on a "wrapped-open" unit sphere.

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

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