Coherence in magnetic quantum tunneling

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Currently, spin tunneling at very low temperatures is assumed to proceed as an incoherent sequence of events that take place whenever a bias field \( h(t) \) that varies randomly with time \( t \) becomes sufficiently small, as in Landau-Zener transitions. We study the behavior of a suitably defined coherence time \( \tau_\phi \). Coherence effects become significant when \( \tau_\phi \lesssim \tau_h \), where \( \tau_h \) is the correlation time for \( h(t) \). The theory of tunneling of Prokof’ev and Stamp (PS), which rests on the assumption that \( \tau_\phi \lesssim \tau_h \), is extended beyond this constraint. It is shown, both analytically and numerically, that \( \tau_\phi \gtrsim \tau_h \) when \( \tau_h \delta h \lesssim h \), where \( \delta h \) is the rms deviation of \( h \). Equations that give \( \tau_\phi \) and the tunneling rate as a function of \( \tau_h \delta h \) both for \( \tau_h \delta h \gtrsim h \), where the theory of PS hold, and for \( \tau_h \delta h \lesssim h \), where it does not, are derived.

Magnetic quantum tunneling (MQT) of large spins has become a subject of much interest. Much of it stems from the expectation that MQT will contribute to our understanding of decoherence. Decoherence effects play an important role in the transition from quantum to classical physics and in quantum computing. Magnetic quantum tunneling (MQT) through thermally activated states is thought to proceed incoherently. However, at sufficiently low temperatures, tunneling must proceed though the ground state doublet. Two-state systems are often used to model tunneling under such conditions. Existing theory treats tunneling between these two states as an incoherent sequence of events that take place whenever the difference between the two-state energies becomes sufficiently small. It is in the spirit of Landau-Zener schemes that are often used in the theory of non-adiabatic transitions to estimate transfer rates between two states whose energies sometimes cross.

Here, I study coherence effects in MQT by means of the following two-state model,

\[
\mathcal{H} = \varepsilon \sigma_z + \Delta \sigma_x - h(t) \sigma_z, \tag{1}
\]

where \( \mathcal{H} \) is the Hamiltonian, \( \sigma_z \) and \( \sigma_x \) are Pauli spin matrices, \( \varepsilon \) is an energy that is constant in time, \( h(t) \) varies with time \( t \) following a stationary random Gaussian process, and \( \Delta \) stands for the tunneling frequency (\( h = 1 \), and \( k = 1 \), where \( k \) is Boltzmann’s constant, from here on). Two-state systems have been used to study MQT, but they have also been used to study decoherence in other systems, such as in (hypothetical) quantum computers. The heat bath is replaced by \( h(t) \) in this model. Consequently, the model is applicable only if \( \Delta \ll T \) and \( \varepsilon \ll T \), where \( T \) is the temperature. This is amply satisfied in present day experiments in MQT when no transverse field is applied.

The aim of this paper is to study within the model defined above the behavior of the coherence time \( \tau_\phi \) (defined below) and of the tunneling rate \( \Gamma \) as a function of \( \tau_h \delta h \), where \( \delta h \) is the rms deviation of \( h(t) \) and \( \tau_h \) is its correlation time. Unless stated otherwise, \( \tau_h \Delta \ll 1, \Delta \ll \delta h \), and \( \Delta \ll (\tau_h \delta h) \delta h \) are assumed to hold everywhere. It is shown that decoherence effects are partially averaged out when \( \tau_h \delta h \lesssim 1 \). I derive, and support with numerical results, that if \( \tau_h \delta h \lesssim 1 \), then \( \tau_\phi/\tau_h \approx 1/(\tau_h \delta h)^2 \), instead of \( \tau_\phi \approx 1/\delta h \) that is shown to hold if \( \tau_h \delta h \gtrsim 1 \). Furthermore, instead of the relation,

\[
\Gamma \approx \sqrt{\frac{\pi}{2}} \frac{\Delta^2}{\delta h} \exp \left[ -\frac{\varepsilon^2}{2(\delta h)^2} \right], \tag{2}
\]

derived below for the tunneling rate under conditions of full incoherence (that is, for \( \tau_\phi \lesssim \tau_h \)), which is the underlying assumption in the spin tunneling theory of Prokof’ev and Stamp, I derive the relation

\[
\Gamma \approx \frac{\Delta^2 \tau_\phi}{1 + \tilde{\tau}_\phi \tau_h^2}, \tag{3}
\]

where \( \tilde{\tau}_\phi^{-1} = 2\tau_h(\delta h)^2 \), is derived under the condition that \( \tau_h \delta h \lesssim 1 \). Note that \( \tilde{\tau}_\phi \approx \tau_\phi \) if \( \tau_h \delta h \lesssim 1 \). Finally, concluding remarks about the likelihood that \( \tau_h \delta h \lesssim 1 \) is realized in Fe\(_8\) and Mn\(_{12}\) are made.

In order to define a measure of coherence, consider the probability \( p_\sigma(t) \) that the system evolve in time \( t \) from an initial state \( |\psi\rangle \) into a final state \( |\sigma\rangle \). We can then write,

\[
p_\sigma(t + \tau) = \sum_{\sigma', \sigma''} w(\sigma, \sigma', \sigma''; \tau) |\sigma\rangle \langle \psi(t)|\psi(t)\rangle \langle \sigma|\sigma''\rangle, \tag{4}
\]

where \( w(\sigma, \sigma', \sigma''; \tau) = \langle \sigma''|U(t_1)|\sigma|\sigma|U(t_1)|\sigma'\rangle \), \( U(t) \equiv \mathcal{T} \exp[-i \int_0^t \mathcal{H}(t) dt_1] \) for a given \( h(t) \) in Eq. (1), and \( \mathcal{T} \) is a time ordering operator. Averaging Eq. (4) over different \( h(t) \) time sequences, then when \( \tau_h \) for
$h(t)$ fulfills $\tau_h \ll \tau, t$, its right hand side breaks up into sums of \textit{products of two terms}, $\langle w(\sigma, \sigma', \sigma''; \tau) \rangle_h$ and $\langle \sigma'|\psi(t)\rangle\langle \psi(t)|\sigma'' \rangle_h$, where $\langle \rangle_h$ stands for average over all different histories of $h(t)$. We are interested here in establishing the physical conditions under which $\langle w(\sigma, \sigma', \sigma''; \tau) \rangle_h \rightarrow \delta_{\sigma,\sigma'}W_{\sigma,\sigma'}(\tau)$ ensues, because this leads to

$$p_{\sigma}(t + \tau) = \sum_{\sigma'} W_{\sigma,\sigma'}(\tau)p_{\sigma'}(t).$$

(5)

No interference terms from Eq. (4) appear in this equation. Thus, probabilities, rather than probability amplitudes, for alternative paths to a final state are summed. Its applicability is, therefore, tantamount to incoherence. Solutions to Eq. (6) exhibit no oscillations. For any two state system, they relax to the final state exponentially. Consider the off-diagonal quantity,

$$q(\sigma, \sigma', \sigma''; \tau) = \frac{\langle w(\sigma, \sigma', \sigma''; \tau) \rangle_h}{(p_1p_2)^{1/2}}$$

(6)

for $\sigma' \neq \sigma''$, where $p_1 = \langle |\sigma''|U^\dagger(|\{h\}, \tau)|\sigma\rangle^2_h$, and $p_2 = \langle |\sigma'|U(|\{h\}, \tau)|\sigma'\rangle^2_h$. As shown above, $q = 0$ leads to Eq. (4), and therefore to incoherence. On the other hand, the system evolves in time in a pure state if there is no averaging over different $h(t)$ time evolutions. No decoherence occurs then, and it follows from the definition of $q$ that $| q | = 1$ then. Quantity $q$ provides, therefore, an appropriate measure of coherence between the two alternative paths $\sigma'$ and $\sigma''$. This is in analogy to an interference experiment that may be performed to test the degree of coherence between the parts of a particle-wave that go through two slits placed at $\sigma'$ and $\sigma''$. Let the coherence time be defined as the smallest time $\tau_\phi$ such that, say, $| q(\tau) | < 1/2$ for all $\tau > \tau_\phi$. (Below, $q$ is independent of $\sigma, \sigma', \sigma''$ throughout. Accordingly, these variables of $q$ are omitted from here on.)

Numerical results are obtained for the system’s time evolution for a given $h(t)$ time sequence by iterating a finite difference Schrödinger’s equation,

$$\psi(t + Dt) = \{1 - iDt\mathcal{H}(t) - [Dt\mathcal{H}(t)]^2/2\}\psi(t),$$

(7)

where $\mathcal{H}$ is defined in Eq. (4), $\psi$ is a two-component state function, and $Dt$ is a suitably small time interval ($Dt \lesssim 10^{-6}/\Delta$ keeps the normalization condition satisfied within 1 part out of 1000 throughout the reported simulations).

The values of $h(t)$ are assumed to follow from a stationary random Gaussian process, with a distribution for $h$ centered on 0, and $h(t)$ is correlated over time according to $\langle h(t)h(0) \rangle_t = (\delta h)^2 \exp(-t/\tau_h)$. This is accomplished with Langevin’s (finite difference) equation, $h(t + Dt) = h(t) - Dt[h(t)/\tau_h + f(t)]$, where $f(t)$ is assigned an independent random number at each value of $t$. All values of $\tau_h$ and $Dt$ used fulfill $Dt \lesssim \tau_h/10$. The values of $h$ obtained follow the prescribed behavior for it if $\langle f \rangle = 0$ and $\langle f(t)f(t') \rangle = \delta_{t,t'}(2 - Dt/\tau_h)(\delta h)^2/(\tau_hDt)$, where $\delta_{t,t'}$ is the Kronecker delta function. The correction term $Dt/\tau_h$ comes about from applying the fluctuation dissipation theorem to the finite difference Langevin equation used here.

Numerical results obtained for the tunneling probability and for $q$ are shown in Fig. 1 for $\delta h/\Delta = 50$ and $\tau_h\Delta = 10^{-4}$. Note that even though $\delta h$ is 50 times larger than $\Delta$, the tunneling probability undergoes oscillations, and, correspondingly, $q \ll 1$ only after a time that is much larger than $\Delta^{-1}$. As shown below, the high degree of coherence exhibited in Fig. 1 occurs when $\tau_h(\delta h)^2 \lesssim \Delta$. It may be realized in MQT experiments by enlarging $\Delta$ through the application of an external field, as in Refs. [1, 4] where $\Delta \approx 30K$ is several orders of magnitude larger than the linewidth [see Eq. (6) below].

![FIG. 1. The probability $p$ that tunneling occurs in time $t$ and the off-diagonal quantity $q$ are shown versus $t\Delta$. The data points shown follow from an average over $4 \times 10^3$ time evolutions with different realizations of $h(t)$, with a vanishing time average value of $h$, $\delta h/\Delta = 50$, and $\tau_h\Delta = 10^{-4}$.](chart)

This extreme condition is worth considering, because it serves to illustrate how decoherence effects can be drastically suppressed. Quite simply, rapid fluctuations in $h$ are almost completely averaged out. More specifically, they are scaled down by the factor $\sqrt{\tau_\phi}/\tau_h$ in time $\tau_\phi$. Thus, $\delta h/\Delta = 50$ is scaled down by 100 in Fig. 1.

Numerical results that illustrate the effects of partially averaging decoherence out are exhibited in Fig. 2.
\[ P = \frac{(1/2) \Delta^2 t / \langle (\delta h)^2 \rangle}{t \Delta} \]

FIG. 2. Tunneling probability \( p \) versus time. The curves shown follow from an average over \( 4 \times 10^3 \) time evolutions with different realizations of \( h(t) \), \( \varepsilon = 0 \), \( \delta h / \Delta = 10^3 \), and \( \tau_h \Delta = 10^{-5} \). From data points obtained for \( q \) (not shown), the value of time where \( q = 1/2 \), that is \( T_\phi \), can be obtained. It turns out that \( \tau_\phi \approx 0.07/\Delta \). The full line is for \( \tau_\phi \); the dotted line is for the prediction that follows for \( p \) assuming incoherence for times shorter than \( \tau_\phi \); the dashed-dotted line is for \( (\Delta t)^2 \), that is fully coherent evolution of \( p \); finally, the long dashed line is for the asymptotic evolution after decoherence sets in.

To start the derivation of the tunneling rates, consider the solution to Schrödinger’s equation, \( \psi(t) = U_0(\{ h \}, t) U_I(\{ h \}, t) \psi(0) \), where \( U_0(\{ h \}, t) = \exp \left[ -i \int_0^t \mathcal{H}_0(\tau) d\tau \right] \), \( \mathcal{H}_0(\tau) = [ \varepsilon - h(\tau)] \sigma_z \), \( U_I(\{ h \}, t) = T \exp \left[ -i \int_0^t \mathcal{V}_I(\tau) d\tau \right] \), \( \mathcal{V}_I(\tau) = U_0^\dagger(\{ h \}, t) V U_0(\{ h \}, t) \), and \( V = \Delta \sigma_z \). The probability amplitude \( a(t) \) that tunneling take place in time \( t \), is that a transition from state \( \sigma_z = -1 \) to state \( \sigma_z = +1 \) occurs, is

\[ |a(t)| = \left| \int_0^t dt_1 \langle 1 | \Delta \sigma_z e^{-2i \int_0^{t_1} \mathcal{H}_0(\tau) d\tau} | -1 \rangle \right|. \tag{8} \]

in first order perturbation theory, where use has been made of the relation \( \exp(A \sigma_z) \sigma_x = \sigma_x \sigma_z \exp(-A \sigma_z) \) for any \( c \)-number \( A \). Now, since \( \mathcal{H}_0(\tau)| \pm 1 \rangle = \pm [\varepsilon - h(\tau)] | \pm 1 \rangle \), it follows that \( \int_0^t \mathcal{H}_0(\tau) d\tau \) can be replaced in Eq. \( \text{[8]} \) by \( [\varepsilon - H(t_1)] t_1 \), where \( H(t) = \int_0^t \mathcal{H}(\tau) d\tau \).

In order to get some feeling for the physics behind the results to be derived, a rough argument that leads to semiquantitative relations for the coherence time and the tunneling rate \( \Gamma \) is given first. For shortness, the argument is restricted to the condition \( \varepsilon = 0 \), but the derivation itself of Eqs. \( \text{[2]} \) and \( \text{[3]} \) is not. It follows from Eq. \( \text{[2]} \), and the definitions of \( q \) and of \( \tau_\phi \) that, in general,

\[ \tau_\phi \approx 1 / \delta H(\tau_\phi), \tag{9} \]

where \( \delta H(\tau_\phi) \) is the rms value of \( H(\tau_\phi) \). Furthermore, \( \delta H(\tau_\phi) \approx \delta h \) if \( \tau_h \delta h \gtrsim 1 \). Therefore, if \( \tau_h \delta h \gtrsim 1 \). There is then complete incoherence, that is, \( \tau_\phi \lesssim \tau_h \). The scheme of Prokof’ev and Stamp, of an incoherent sum of tunneling probability increments that occur whenever \( h(t) \) becomes comparable to \( \Delta \), is then justified. If, on the other hand, \( \tau_h \delta h \lesssim 1 \), then \( \delta H(\tau_\phi) \approx \delta h / \sqrt{\tau_\phi / \tau_h} \), since \( \tau_\phi / \tau_h \) is, in a rough sense, the number of times that \( h \) fluctuates independently during time \( \tau_\phi \). Therefore, substitution into Eq. \( \text{[2]} \) gives

\[ \tau_\phi / \tau_h \approx 1 / (\tau_h \delta h)^2 \tag{11} \]

if \( \tau_h \delta h \lesssim 1 \). This equation exhibits explicitly how averaging out of fluctuations makes \( \tau_\phi \) larger than \( \tau_h \) if \( \tau_h \delta h \lesssim 1 \). Data points obtained numerically for \( \tau_\phi \) are shown in Fig. 3. Note that Eq. \( \text{[11]} \) also implies that coherence times become larger than tunneling times when \( \tau_h \delta h^2 \lesssim \Delta \).

The tunneling rate \( \Gamma \) is estimated next for \( \varepsilon = 0 \) and \( \tau_h \delta h \lesssim 1 \). It follows from Eq. \( \text{[8]} \) that the probability \( p(t, H(t)) \) for a given \( H(t) \) that a spin has tunneled at time \( t \) is \( p(t, H(t)) \approx \Delta^2 t^2 \) for \( H(t) \lesssim 1 \). It may seem strange that a spin under a large field tunnels as fast as one for which \( H = 0 \) even though the line shape is known to be a Lorentzian function, but note that \( \Delta^2 (\Delta^2 + H^2)^{-3/2} \sin^2(\Omega t) \), where \( \Omega^2 = H^2 + \Delta^2 \), is approximately \( \Delta^2 t^2 \) for \( \Omega t \lesssim 1 \), which is independent of \( H \). Averaging over all \( h(t) \) sequences leads to \( p(\tau_\phi) \approx \Delta^2 \tau_\phi^2 \).

Equation \( \text{[8]} \) gives \( p(t) \approx p(\tau_\phi) \tau_\phi / \tau_\phi \) for \( t \gtrsim \tau_\phi \), since

\[ \tau_\phi \approx 1 / \delta h, \tag{10} \]
\[ \tau_h \lesssim \tau_\phi \] under the assumed condition \( \tau_\phi \delta h \lesssim 1 \). It follows then that, \( \Gamma \sim \Delta^2/|\tau_h(\delta h)|^2 \) for \( \varepsilon = 0 \).

The derivation of Eqs. (2), (3), (11), and (12), from which we digressed below Eq. (8) is now resumed. It is assumed that \( h(t) \) follows a stationary random Gaussian process, centered on 0 [a non-zero value simply redefines \( \varepsilon \) in Eq. (1)], and \( h(t) \) is correlated over time, with a correlation time \( \tau_h \) defined by \( \tau_h [h(t)^2] = \int_0^\infty \langle h(t)^2 \rangle dt \delta h(t') \), where \( \langle \ldots \rangle_\delta \) stands for an average over all \( h(t) \) histories. The probability,

\[
P(t) = \Delta^2 \int_0^t dt_2 \int_0^{t_2} dt_1 \langle e^{2iF(t_1,t_2)} + cc \rangle, \tag{12}
\]

follows straightforwardly from Eq. (8) and the statement following it. Now, \( \int_0^{t_2} h(\tau) d\tau \) follows a Gaussian distribution since \( h(t) \) follows, by assumption, from a stationary random Gaussian process. Therefore, after averaging over all histories of \( h(t) \), Eq. (12) becomes

\[
P(t) = \Delta^2 \int_0^t dt_2 \int_0^t dt_1 e^{-2F(t_1,t_2)} \cos [2\varepsilon(t_2 - t_1)], \tag{13}
\]

where \( F(t_1, t_2) = \int_0^{t_2} dt_1 \int_0^{t_1} dt_2 \langle h(\tau_1) h(\tau_2) \rangle_\delta \). Now, \( F(t_1, t_2) \approx 2(\delta h)^2 \tau_h |t_2 - t_1| \) if \( \tau_h \ll |t_2 - t_1| \), and a bit of reflection shows that this relation applies if \( \tau_\phi \delta h \ll 1 \) and \( t \gg 1/|\tau_h(\delta h)|^2 \) (that is, if \( t \gg \tau_\phi \)), which leads to Eq. (3). On the other hand, \( F(t_1, t_2) \approx (\delta h)^2 |t_2 - t_1|^2 \) if \( \tau_h \gg |t_2 - t_1| \), and this relation is applicable if \( \tau_\phi \delta h \gg 1 \) and \( t \gg 1/(\tau_\phi \delta h) \) (that is, if \( t \gg \tau_\phi \)), which leads to Eq. (8). Similarly,

\[
q = \pm \frac{i\Delta}{\sqrt{P(t)}} \int_0^t dt_1 e^{2i\varepsilon t_1} e^{-2F(0,t_1)}, \tag{14}
\]

follows from Eq. (3) and the assumption of a stationary random Gaussian \( h(t) \). By the same arguments that led to Eqs. (2), and (8) above, equations (14) and (12) follow straightforwardly, independently of \( \varepsilon \).

Numerical results obtained for \( \Gamma \) and for \( \tau_\phi \) are shown in Fig. 3. There is good agreement with Eqs. (3), (11), (12), and (14).

In conclusion, coherence effects have been shown to set in when the coherence time \( \tau_\phi \) is longer than the correlation time \( \tau_h \) for \( h(t) \), and that this comes about because decoherence effects that arise from \( h(t) \) partially average out when \( \tau_h \lesssim 1/\delta h \). Coherence times are then larger than \( \tau_h \). Equations (2) and (3) which give \( \tau_\phi \) and the tunneling rate as a function of \( \tau_h \delta h \) both, for \( \tau_h \delta h \gtrsim h \), where the theory of Prokof’ev and Stamps holds, and for \( \tau_h \delta h \lesssim h \), where their theory does not hold, are derived.

The condition \( \tau_h \delta h \lesssim 1 \), which gives \( \tau_\phi \gtrsim \tau_h \), would be fulfilled in spin tunneling systems in which nuclear spin motion is driven by other atoms in the system with which hyperfine interaction strengths \( \delta^* \) are larger than \( \delta \) with the spins of interest if \( \tau_h^{-1} \sim \delta^* \). There seems to be some indirect experimental evidence that \( \tau_\phi \gtrsim \tau_h \), or, equivalently, that \( \tau_h \delta h \lesssim 1 \) in Fe₈ and in Mn₁₂ clusters. Resolution of the tunneling pair of energy eigenstates has been reported in Fe₈ and in Mn₁₂ under large transverse fields, both at 680 MHz. This implies that \( \tau_\phi \gtrsim 10^{-9} \) seconds in both cases. This is to be compared with the value of \( 1/\delta h \) that \( \tau_\phi \) would equal to [see, Eqs. (8) and (13)] if \( \tau_h \delta h \gg 1 \) were fulfilled. Now, \( \delta h \approx 0.02K \) for Fe₈ (as follows from measured hyperfine fields of \( 10^{-3} \)T reported by Wernsdorfer, \( g = 2 \), and \( S = 10 \), that is, \( \delta h \approx 0.5 \times 10^{-8} \)s. For Mn₁₂, \( \delta h \approx 10^{-2}T \), (see Ref. 8), from which \( 1/\delta h \approx 0.5 \times 10^{-8} \)s follows. It follows that, in fact, \( \tau_h \delta h \gtrsim 1 \) is not fulfilled, neither for Fe₈ nor for Mn₁₂, suggesting that tunneling in Fe₈ and in Mn₁₂ proceeds with some degree of coherence, even when no external transverse field is applied.

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