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Alternating-current-hampered tunnelling of magnetization

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Abstract. Strong evidence is presented that an alternating magnetic field can only hamper the tunnelling of magnetization. It is surmised that this is a 'no-go' theorem, which is to be contrasted with the particle case where an ac field favours tunnelling. We employ a WKB formalism, study a mesoscopic spin in various anisotropies, and show how to apply the averaging method to the high-frequency regime so as to reduce the problem to one with a stationary field. The latter can be analysed straightforwardly and so can the adiabatic case. In addition, we put forward an exactly soluble case underlining the claim that hampering is quite a general phenomenon. Some exceptions related to resonance are explained as well.

1. Introduction

Tunnelling is one of the most pronounced manifestations of quantum mechanics, in that an object penetrates a region which is classically forbidden and, hence, inaccessible. There has been a long-lasting interest in the tunnelling of particles [1, 2] but the attention attracted by their spin counterparts is much more recent [3–10]. Generically, one studies a problem with a symmetry and a tunnelling term that lifts some, usually twofold, degeneracy. The eigenfunctions of the Hamiltonian H then respect this symmetry; they occur in, say, odd/even pairs, are localized in *both* minima of H, and the system performs a coherent oscillation between the two wells with a rate of the form $\tau^{-1} = \tau_0^{-1}\exp(-I/\hbar)$. The prefactor τ_0^{-1} is an attempt frequency, I is some action, and $\exp(-I/\hbar)$ may be interpreted as a tunnelling probability.

Here we are interested in the tunnelling of a giant spin with spin quantum number $S \gg 1$ under the influence of a time-dependent perturbation. The solution to this problem is of particular relevance to mesoscopic magnetic moments [6, 10] in an anisotropy field. In the stationary case, the difference between spins and particles is already remarkable. Whereas particles give rise to an action I which shows, typically, a square-root dependence upon the coupling constants, spins exhibit a *logarithmic* dependency [3, 4]. In a periodically driven system, particles commonly tunnel more quickly than under the influence of a constant potential [11–13]. The key question is now: How do *spins* behave? To answer this question we will study a few typical models in the context of the averaging method [14, 15], that allows a reduction to a time-independent problem which is more easily accessible to, e.g., a WKB analysis [3, 4]. One has to realize, however, that the WKB analysis is instrumental in solving a time-dependent problem such as the present one. We will also discuss an exact solution, underlining our conclusion that *in general, and in contrast to the particle case* [11–13], *a periodically driven spin is hampered when it is going to tunnel*. In order to prepare the ground for the ensuing calculations, we must now make a small detour.

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The type of equation which we have to study is of the form

$$\dot{x} = \epsilon f(t, x) \qquad f(t+T, x) = f(t, x) \tag{1}$$

with some 'small' parameter $\epsilon > 0$. The function $f(t, x)$ and its partial derivative with respect to x are uniformly bounded in x and t by a positive constant M and are periodic in t. Since ϵ is small as compared to $1/T$, x hardly changes during a period of duration T, and one may therefore average (1) so as to get the autonomous equation

$$\dot{y} = \epsilon f_0(y) \qquad f_0(y) = \frac{1}{T}\int_0^T \mathrm{d}t\ f(t, y) \tag{2}$$

where f_0 does not depend on time any more. The solution to (2) approximates $x(t)$ to order ϵ for times $t \leqslant 1/\epsilon$. The key to the proof [14, 15] which is behind what follows is defining

$$u_1(t, x) = \int_0^t \mathrm{d}t'\ [f(t', x) - f_0(x)]$$

noting that, whatever t is, one has $\sup|u_1(t, x)| \leqslant 2MT$, and studying the time evolution of z defined by $x(t) := z(t) + \epsilon u_1(t, z(t))$. A simple calculation shows that z satisfies (2)—up to an error $O(\epsilon^2)$. This is the *method of averaging* [14, 15], which dates back to Lagrange.

The three periodically driven spin systems which we will study are typical examples, and are characterized by the Hamiltonians

$$H_1 = -\gamma S_z^2 - \delta\cos(\omega t)S_z - \alpha S_x \tag{3}$$
$$H_2 = -\gamma S_z^2 - \alpha\cos(\omega t)S_x \tag{4}$$
$$H_3 = -\gamma S_z^2 - \alpha[\cos(\omega t)S_x + \sin(\omega t)S_y] \tag{5}$$

with α, γ, and δ positive. The anisotropy which produces the 'energy barrier' is represented by $-\gamma S_z^2$. It is assumed to dominate the other terms so that we arrive at a well-defined tunnelling problem where the spin tunnels along the S_z-axis. Phrased differently, $\alpha, \delta \ll \Gamma S$ where $\Gamma = \gamma\hbar$ has the dimension of frequency. In H_1 there is an oscillating magnetic field parallel to the anisotropy axis; in H_2 and H_3 it is orthogonal to the z-axis. The fields in H_1 and H_2 are linearly polarized, and that of H_3 is circularly polarized. As for H_1, the unperturbed case $\delta = 0$ represents coherent tunnelling in that the Hamiltonian has a symmetry: a rotation through π about the x-axis; H_2 has the very same symmetry. It is natural to compare the ac behaviour of H_1 with that for $\delta = 0$, since $\delta \neq 0$ in conjunction with $\omega \neq 0$ in general favours tunnelling of particles [12] ($S_z \leftrightarrow q$). If spins were similar to particles, kicking a spin periodically would have the same effect. It does not. For H_2 and H_3, the reference state is $\omega = 0$. Given an unperturbed level splitting $\Delta E = \hbar\omega_0$, we discern three cases: $\omega \gg \omega_0$, resonance $\omega \approx \omega_0$, and the adiabatic case $\omega \ll \omega_0$. For all three Hamiltonians the reference ('unperturbed') operator is $H_u = -\gamma S_z^2 - \alpha S_x$. Its ground-state level splitting was computed more than ten years ago:

$$\hbar\omega_0 = \alpha\left(\frac{\alpha}{\Gamma S}\right)^{2S-1} \qquad \text{for } S \gg 1 \tag{6}$$

(cf. equation (C.19) in [3]).

As for the context of the present paper, a few historical and practical remarks are in order. The problem of how a giant quantum spin tunnels was solved surprisingly late in the history of quantum mechanics. It was only in 1986 that two types of solution appeared. Enz and Schilling [5] solved a special case by mapping it onto a particle problem through the Villain transformation. The particle problem was then solved to high precision by means of a functional-integration technique. Van Hemmen and Sütő [3] started with the Schrödinger equation directly and developed a WKB formalism for quantum spins, which is used throughout what follows. It allows both for high-precision calculations of the level splitting and the tunnelling rate, and for a universal representation of these quantities, that does not depend on the detailed form of the Hamiltonian and (thus) is less precise but has been shown [4] to exactly agree with the spin-functional-integral results of Chudnovsky and Gunther [6]. Until now, however, the Hamiltonian describing a system was not allowed to be time dependent. Since it is a simple matter to put small magnetized particles, i.e., giant spins, in a homogeneous magnetic ac field, and the strength of an external field is at one's disposal, we thought it worthwhile to analyse the ac problem in some detail.

In section 2 we treat the case of a linearly polarized field in the three frequency regimes mentioned above. We will have recourse to the method of averaging at high frequencies, to a two-level approximation at resonance, and an adiabatic treatment for low frequencies. Circular polarization is to be discussed in section 3. The time evolution operator for H_3 can be obtained exactly.

2. Linear polarization

Our analysis is most easily performed in a spectral representation with S_z diagonal [3, 4]. In the context of the WKB formalism the *ansatz* for the wave function is $\psi = \exp[(\mathrm{i}/\hbar)S_0 + S_1]$. It is implicitly understood that we take the semiclassical limit $\hbar \to 0$ and $S \to \infty$ so that $\hbar S = \sigma$ is constant. Then S_1 is subdominant and can be dropped. For the time being, however, $\hbar$ is finite. Furthermore, in our spectral representation $S_x = a(s)(T_{\hbar} + T_{-\hbar})/2$ and $(T_{\pm\hbar} f)(s) = f(s \pm \hbar)$ while $a(s) = \sqrt{\sigma^2 - s^2}$ and s ranges through the set $\{m\hbar : -S \leqslant m \leqslant S\} \subseteq [-\sigma, \sigma]$. The time-dependent Schrödinger equation for H_1 now reads

$$\mathrm{i}\hbar \frac{\partial \psi(s,t)}{\partial t} = -[\gamma s^2 + \delta s \cos(\omega t)]\psi(s,t) - \frac{\alpha}{2} a(s)[\psi(s+\hbar) + \psi(s-\hbar)]. \tag{7}$$

2.1. The high-frequency regime

Let us assume that $\omega \gg \omega_0$ and use the interaction picture. This is equivalent to the substitution $\psi(s,t) = \tilde{\psi}(s,t)\exp\{(\mathrm{i}/\hbar)[\gamma s^2 t + \delta s\, \omega^{-1}\sin(\omega t)]\}$ and gives

$$\mathrm{i}\hbar \frac{\partial \tilde{\psi}(s,t)}{\partial t} = -\frac{\alpha}{2} a(s) \mathrm{e}^{\mathrm{i}\Gamma t}[\mathrm{e}^{\mathrm{i}\Phi(s,t)}\tilde{\psi}(s+\hbar,t) + \mathrm{e}^{-\mathrm{i}\Phi(s,t)}\tilde{\psi}(s-\hbar,t)] \tag{8}$$

where $\Gamma = \gamma\hbar$ and

$$\Phi(s,t) = 2\gamma s t + (\delta/\omega)\sin\omega t. \tag{9}$$

We imagine that the spin has to tunnel from $-\sigma$ to σ or conversely, and concentrate on the region where $s \approx 0$, so that (8) attains the form (1). If there is a transition, it takes place on the time-scale ω_0^{-1}. Therefore, and because $\omega \gg \omega_0$, we can average over the time evolution during a single period of the external field. Assuming $s \approx 0$ we average (8) over one period and find

$$\mathrm{i}\hbar \frac{\partial \tilde{\psi}_a(s,t)}{\partial t} = -\frac{\alpha_{\mathrm{eff}}}{2} \mathrm{e}^{\mathrm{i}\Gamma t}[\mathrm{e}^{2\mathrm{i}\gamma s t}\tilde{\psi}_a(s+\hbar,t) + \mathrm{e}^{-2\mathrm{i}\gamma s t}\tilde{\psi}_a(s-\hbar,t)] \tag{10}$$

where $\alpha_{\mathrm{eff}} = \alpha J_0(\delta/\omega)$. The Bessel function J_0 stems from

$$J_0(z) = (1/2\pi)\int_0^{2\pi} \mathrm{d}\vartheta \;\; \cos(z\sin\vartheta)$$

which is Bessel's integral [16, 17]. The 'averaged' function $\tilde{\psi}_a$ plays the role of y in (2) with ϵ corresponding to $\alpha a(0)/\hbar = \alpha S$, and obeys the same equation as a wave function in the time-independent case where $\delta = 0$; cf. (8) and (9). The only—but important—modification is that α has been replaced by α_{eff}; the latter reduces to α, if $\delta = 0$.

We can apply the averaging method, if two conditions hold. First, $\epsilon \ll 1/T$ as discussed after (1). In the present case this means that $\alpha S \ll \omega = 2\pi/T$ and implies [3] $\omega_0 \ll \omega$. Second, the argument of Φ in (8) should vary slowly with time: $\gamma|s|T \ll 1$. This is equivalent to $|s| \ll \hbar\omega/\Gamma$. As long as $\omega/\Gamma \gg 1$ the region where $s \approx 0$ has a finite width. Usually the spin has to tunnel from $-\sigma$ to σ and conversely (or the like). If it is hindered near $s = 0$, tunnelling is hampered or blocked.

The Bessel function J_0 has infinitely many zeros z_ν, the first being $z_1 = 2.405$. Furthermore, $|J_0(z)| \leqslant 1$, and its maximum, i.e., 1, is assumed at $z = 0$. So we see that a spin never tunnels faster than in the stationary case where $\delta = 0$ because $|\alpha_{\text{eff}}| \leqslant \alpha$. It can even get stuck, if $\delta/\omega := z$ corresponds to one of the zeros z_ν of the Bessel function J_0. This allows a simple but striking experimental check: the spin gets stuck at discrete frequencies whose ratios are determined by the Bessel zeros z_ν.

Turning to H_2, we perform the transformation $\psi := \tilde{\psi}\exp(\mathrm{i}\gamma s^2 t/\hbar)$, obtain

$$\mathrm{i}\hbar\frac{\partial\tilde{\psi}_a(s,t)}{\partial t} = -\frac{\alpha}{2}a(s)\mathrm{e}^{\mathrm{i}\Gamma t}\cos(\omega t)[\mathrm{e}^{2\mathrm{i}\gamma st}\tilde{\psi}_a(s+\hbar,t) + \mathrm{e}^{-2\mathrm{i}\gamma st}\tilde{\psi}_a(s-\hbar,t)] \tag{11}$$

and average this in the region where $s \approx 0$ so as to find $\alpha_{\text{eff}} = 0$. That is to say, tunnelling is blocked completely—in clear contrast to the case where $\omega = 0$.

2.2. The adiabatic case

The adiabatic case where $\omega \ll \omega_0$ can be handled straightforwardly. Because $H_1(t)$ changes very slowly on the time-scale ω_0^{-1} given by the original tunnelling time, we can consider it as a time-independent operator depending only on a formal parameter t, and write $H_1(t) = H_0(\delta\cos\omega t)$ where the latter is defined through equation (21) below. The discussion of section 3 leading to equation (31) is therefore relevant to this case also (recall that $|\delta\cos\omega t| \leqslant \delta \ll \Gamma S$), and we conclude that the tunnelling probability—say, in the time interval $(t - 2\pi\omega_0^{-1}, t + 2\pi\omega_0^{-1})$—suffers a small reduction proportional to $(\delta\cos\omega t/\Gamma S)^2$.

The tunnelling rate for H_2 is determined as follows [3]. We pick a certain time t, write $\alpha(t) = \alpha\cos(\omega t)$, and treat it as a constant as long as $\alpha(t)$ is outside a suitably chosen neighbourhood $\mathcal{N}(0)$ of zero† so that the 'effective' tunnelling time is much less than ω^{-1}. Keeping the proviso in mind that we stay outside $\mathcal{N}(0)$, we substitute the WKB *ansatz* $\psi \sim \exp(\mathrm{i}S_0/\hbar)$, and compute $S_0(s,t) = W(s) - Et$ in the semiclassical limit, as $\hbar \to 0$:

$$E = -\gamma s^2 - \alpha(t)a(s)\cos\left(\frac{\partial W}{\partial s}\right). \tag{12}$$

This we can solve for W. Now the tunnelling rate is $\tau^{-1} = \Delta E/\pi\hbar$ and $\Delta E \propto |\langle\phi_\ell|\phi_r\rangle|$ where ϕ_r and ϕ_ℓ are the WKB wave functions belonging to E which start either on the right or on the left of $s = 0$. We have $\arccos(Z) = \pm(1/\mathrm{i})\ \mathrm{arccosh}(Z)$ and $\mathrm{arccosh}(Z) \approx \ln(2Z)$

† In the adiabatic regime, $\omega \ll \omega_0$. However, the 'effective' tunnelling frequency $\omega_0(t)$ that one can compute from equation (6) by replacing α by $\alpha(t)$ becomes much smaller than ω as time goes on. That is to say, starting with $\omega \ll \omega_0(0)$, one passes from the adiabatic case at $t = 0$ to the high-frequency case, via resonance, as time proceeds. For H_2, this is the heart of the problem in performing the adiabatic approximation. The neighbourhood $\mathcal{N}(0)$ depends on ω.

for large Z. The upshot of the calculation is

$$\tau^{-1} = \tau_0^{-1} \exp\left[-2S \ln\left(\frac{2|E|}{|\alpha(t)|/\gamma\sigma}\right)\right]. \tag{13}$$

In passing we note that the logarithm in the exponential is *typical of spins* [3, 4]. It stems from the arccosh term. For the ground-state pair with $E := -\gamma\sigma^2$, we are left with $\tau^{-1} = \tau_0^{-1}[|\alpha(t)|/2\gamma\sigma]^{2S}$, which can be compared directly with (6). The order of magnitude has been confirmed fully by a numerically exact diagonalization of the Hamiltonian [3], which gives the level splitting $\Delta E = \pi\hbar/\tau$. If desired, one may average τ^{-1} over one period once $\mathcal{N}(0)$ is small enough and, by Wallis's formula [16], end up with an extra prefactor—an explicit expression that decreases as $1/\sqrt{\pi S}$ for S large and entails a reduction of the unperturbed tunnelling rate.

2.3. *Resonance*

The resonance case where $\omega \approx \omega_0$ is of some independent interest. Its treatment has been included here since a spin offers some subtleties which are not present in the standard argument [20]. Here we can invoke the two-level approximation [4], which is specific to the resonance condition. We start with H_1 and write $H_1 = H_u + V(t)$ where $H_u = -\gamma S_z^2 - \alpha S_x$ and $V(t) = -\delta\cos(\omega t)S_z$. We concentrate on the ground-state pair $\{\varphi_0^{(0)}, \varphi_1^{(0)}\}$ of H_u with $\Delta E = E_1^{(0)} - E_0^{(0)} \equiv \hbar\omega_{10}$ where $\omega_{10} = \omega_0$ is given by equation (6). Let $\psi_k^{(0)} = \varphi_k^{(0)} \exp(-\mathrm{i}E_k^{(0)}t/\hbar)$. The wave function can be expanded in the form

$$\psi(t) = \sum a_k(t)\psi_k^{(0)}(t). \tag{14}$$

Denoting differentiation with respect to time by a dot, we obtain $\mathrm{i}\hbar\dot{a}_m = \sum_k V_{mk}a_k$, the V_{mk} being the matrix elements with respect to the $\psi^{(0)}$s. In agreement with the two-level approximation we restrict k to the set $\{0, 1\}$. We have

$$V_{10} = -\delta\cos(\omega t)\,\mathrm{e}^{\mathrm{i}\omega_{10}t} \sum_{m=-S}^{S} \varphi_1^{(0)}(m)\, m\hbar\, \varphi_0^{(0)}(m) \tag{15}$$

while the diagonal elements V_{00} and V_{11} vanish since the $\varphi_k^{(0)}$ are odd or even; there is no harm in assuming them to be real. The sum in (15) is written as $\hbar v_{10}$. For the ground-state pair [3], $v_{10} \approx S$. Putting $C = \delta v_{10}/2$ and $\Omega = \omega - \omega_{10}$, we are left with only two equations:

$$\dot{a}_0 = \mathrm{i}C(\mathrm{e}^{\mathrm{i}\omega t} + \mathrm{e}^{-\mathrm{i}\omega t})\mathrm{e}^{-\mathrm{i}\omega_{10}t}a_1 \approx \mathrm{i}C\mathrm{e}^{\mathrm{i}\Omega t}a_1 \tag{16}$$

$$\dot{a}_1 = \mathrm{i}C(\mathrm{e}^{\mathrm{i}\omega t} + \mathrm{e}^{-\mathrm{i}\omega t})\mathrm{e}^{\mathrm{i}\omega_{10}t}a_0 \approx \mathrm{i}C\mathrm{e}^{-\mathrm{i}\Omega t}a_0. \tag{17}$$

The second equality is a consequence of the *rotating-wave approximation* [19], which entails dropping the fast-rotating terms. It respects the normalization of the wave function. Since $|\Omega| \ll \omega + \omega_{10}$ and the frequencies under consideration are rather high, the approximation is a good one. The above equations closely resemble but are not identical to those derived by Rabi [21] in the thirties.

Given a first-order differential equation, one has to specify initial conditions. The spin starts, say, on the left, in the neighbourhood of $m = -\hbar S$. Now for large S the eigenfunctions $\varphi_0^{(0)}$ and $\varphi_1^{(0)}$ of H_u have the remarkable property [3] that to a very high precision the functions $(\varphi_0^{(0)} \pm \varphi_1^{(0)})/\sqrt{2}$ are localized at $m > 0$ and $m < 0$, respectively; cf. also equations (26) below. We can therefore take $a_0(0) = a_1(0) = 1/\sqrt{2}$ as an initial condition and ask whether at a later time t we find $a_0(t) = -a_1(t) = 1/\sqrt{2}$ or, in other words, whether the spin is in the other well.

We first study the case of perfect resonance, i.e., where $\Omega = 0$. Then (16) and (17) can be solved directly. Let $\boldsymbol{a}$ denote the vector (a_0, a_1) and let σ_x be the first Pauli spin matrix. The solution is $a_0(t) = a_1(t) = \exp(\mathrm{i}Ct)/\sqrt{2}$, because $\boldsymbol{a}(t) = \exp(\mathrm{i}C\sigma_x t)\boldsymbol{a}(0)$ and the initial condition $(1,1)/\sqrt{2}$ is an eigenvector of σ_x with eigenvalue 1. So the wave function at time t is

$$\psi(t) = \frac{1}{\sqrt{2}} \mathrm{e}^{-\mathrm{i}(E_0^{(0)}/\hbar - C)t}[\varphi_0^{(0)} + \mathrm{e}^{-\mathrm{i}\omega_{10}t}\varphi_1^{(0)}] \tag{18}$$

and the spin oscillates with the unperturbed frequency ω_{10} between the two wells.

For non-zero Ω we proceed à la Rabi [21] and differentiate a_1 twice, use (16) as well, and get a second-order linear differential equation:

$$\ddot{a}_1 + \mathrm{i}\Omega\dot{a}_1 + C^2 a_1 = 0. \tag{19}$$

This can be solved by the simple *ansatz*

$$a_1(t) = A\exp(\mathrm{i}\mu_+ t) + B\exp(\mathrm{i}\mu_- t)$$

where the $\mu_\pm$ are the two roots of the equation $\mu^2 + \Omega\mu - C^2 = 0$. Near resonance we find $a_1 = a_0\exp(-\mathrm{i}\Omega t)$ and hence, due to (14),

$$\psi(t) = \mathrm{e}^{-\mathrm{i}E_0^{(0)}t/\hbar} a_0(t)[\varphi_0^{(0)} + \mathrm{e}^{-\mathrm{i}\omega t}\varphi_1^{(0)}] \tag{20}$$

as $\Omega + \omega_{10} = \omega$. That is, near resonance we find a narrow frequency window where the spin oscillates in phase with the external field. One might think that resonance also occurs, if, say, $\omega = \omega_{30}$. In principle this is correct, were it not that the corresponding matrix element v_{30} vanishes to high precision. That is, we need another kind of argument. The above reasoning hinges on the symmetry properties of the unperturbed eigenstates and, thus, does not apply to H_2.

3. Circular polarization: an exactly soluble case

The third case (5) of a field with circular polarization can be treated exactly. The solution to a time-dependent Schrödinger equation $H(t)\psi = \mathrm{i}\hbar\, \partial\psi/\partial t$ starting at time $t = t_0$ is the unitary group $U(t, t_0)$ satisfying $U(t, t')U(t', t_0) = U(t, t_0)$ with the initial condition $U(t_0, t_0) = \mathbf{1}$. In the case of $H = H_3(t)$, the time evolution operator $U(t, t_0)$ has a nice closed form [18]. Indeed, if we define

$$H_0(\omega) = -\gamma S_z^2 - \omega S_z - \alpha S_x \tag{21}$$

and write $H_0 = H_0(\omega)$, then

$$U(t, t_0) = \exp\left(-\frac{\mathrm{i}\omega}{\hbar}\, tS_z\right)\exp\left[-\frac{\mathrm{i}}{\hbar}(t - t_0)H_0\right]\exp\left(\frac{\mathrm{i}\omega}{\hbar}\, t_0 S_z\right). \tag{22}$$

To prove (22) we note that $U(t_0, t_0) = \mathbf{1}$ and via a direct computation

$$\mathrm{i}\hbar\, \partial U(t, t_0)/\partial t = H_3(t)U(t, t_0). \tag{23}$$

Let $|m\rangle$ and $|n\rangle$ be two eigenstates of S_z with eigenvalues $m\hbar$ and $n\hbar$, respectively. The quantity

$$P_{mn}(t) = |\langle m|U(t, 0)|n\rangle|^2 \tag{24}$$

is the transition probability for a spin starting in $|n\rangle$ at time $t = 0$ to be in state $|m\rangle$ at time t. Because of equation (22), P_{mn} assumes the simple form

$$P_{mn}(t) = |\langle m|\mathrm{e}^{-\mathrm{i}tH_0/\hbar}|n\rangle|^2 \tag{25}$$

so the transition probability is fully determined by $H_0(\omega)$. We are interested in transitions between $|n<0\rangle$ and $|m>0\rangle$, corresponding to a tunnelling situation.

Let us consider the case where $m=S$, $n=-S$ in some detail. We recall that for $\omega=0$

$$|S\rangle \approx \frac{1}{\sqrt{2}}\big(\varphi_0^{(0)}+\varphi_1^{(0)}\big)$$
$$|-S\rangle \approx \frac{1}{\sqrt{2}}\big(\varphi_0^{(0)}-\varphi_1^{(0)}\big). \tag{26}$$

This approximation is a good one as long as $\alpha \ll \Gamma S$. Here $\varphi_0^{(0)}$ and $\varphi_1^{(0)}$ are the lowest even and odd eigenstates of $H_0(\omega=0)$; cf. figure 8 in [3] or figure 7 in [4]. Using (26) we find

$$P_{S,-S}(t) \approx \frac{1}{4}|1-\mathrm{e}^{-\mathrm{i}t\omega_0}|^2 \tag{27}$$

and

$$\max_t P_{S,-S}(t) \approx P_{S,-S}(\pi\omega_0^{-1}) \approx 1. \tag{28}$$

For $0<\omega\ll\Gamma S$ we still have

$$|S\rangle \approx a\varphi + b\psi$$
$$|-S\rangle \approx b\varphi - a\psi \tag{29}$$

where φ and ψ are eigenstates of $H_0(\omega)$, tending respectively to $\varphi_0^{(0)}$ and $\varphi_1^{(0)}$ as $\omega\to 0$. However, φ and ψ have no even–odd symmetry:

$$\sum_{m>0}|\langle m|\varphi\rangle|^2 > \sum_{m<0}|\langle m|\varphi\rangle|^2 \tag{30}$$

the $<$ sign holding for ψ. We therefore have $a^2+b^2=1$ and $a>1/\sqrt{2}$, $a-1/\sqrt{2}\propto\omega/\Gamma S$. Equations (25) and (29) then yield

$$P_{S,-S}(t) \leqslant 4a^2(1-a^2) = 1-c(\omega/\Gamma S)^2 \tag{31}$$

with some $c>0$ of the order of unity. As ω increases, $P_{S,-S}(t)$ rapidly approaches zero since the $|\pm S\rangle$ get closer and closer to two eigenstates of $H_0(\omega)$, which are separated by an energy difference $\sim 2S\hbar\omega$.

The properties of P_{mn} for $m>0$ and $n<0$ such that $|n|\geqslant m$ and $(m+|n|)\Gamma\gg\alpha$ (in practice, when $m+|n|$ is of the order of S) resemble that of $P_{S,-S}$: $\max_t P_{mn}(t)$ as a function of ω has a local maximum close to 1 near the frequency $(|n|-m)\Gamma$ and decays rapidly once ω moves away from this value. This holds because there are level crossings in the spectrum of $-\gamma S_z^2-\omega S_z$ at $\omega=k\Gamma$ for all $1\leqslant k\leqslant S$: the eigenvalues belonging to the eigenvectors $|m\rangle$ and $|n\rangle$ coincide for all pairs (m,n) such that $k=|n|-m$. The degeneracy of the (m,n) pair is lifted in the $(m+|n|)$th order of the perturbation by αS_x. The condition $(m+|n|)\Gamma\gg\alpha$ implies that the other energy levels of $-\gamma S_z^2-k\Gamma S_z$ are far away, as measured in units of $\alpha\hbar$. Therefore, one obtains two approximate eigenvectors of $H_0(k\Gamma)$ as linear combinations of $|m\rangle$ and $|n\rangle$. Inverting this relation, we arrive at an expansion, analogous to (29), for $|m\rangle$ and $|n\rangle$. The asymmetry $a-b$ of the wave functions and the tiny level splitting can be computed from the matrix elements of $(\alpha S_x)^{(m+|n|)}$ sandwiched between $|m\rangle$ and $|n\rangle$. Now $a-b$ vanishes as $\alpha/[(m+|n|)\Gamma]$ approaches zero, and the level splitting yields a tunnelling frequency similar to (6).

The present result for H_3 fully confirms the approximate calculation for H_2 in the high-frequency regime. It is easy to imagine more complicated Hamiltonians, e.g., a circular polarization about an arbitrary axis, but the reader will easily verify that one can just repeat one of the above arguments—up to a simple modification.

We could take, for instance, the y-axis as the rotation axis and study

$$H_4 = -\gamma S_z^2 - \alpha[\cos(\omega t)S_z + \sin(\omega t)S_x]. \tag{32}$$

We then perform the very same transformation as the one from (7) to (8) and (9), and replace α in (8) by $\alpha\sin(\omega t)$ and δ in (9) by α. Averaging (8) in the region where $s \approx 0$ we arrive at a vanishing α_{eff}. Hence tunnelling of a mesoscopic spin is blocked.

4. Conclusion

In view of the available evidence we tentatively suggest that a periodically driven mesoscopic spin is (nearly) always hampered or even blocked once the frequency ω is large enough. There are two cases: a linearly and a circularly polarized field. If the ac field is linearly polarized and parallel to the anisotropy axis, as in (3), then there exists a narrow transparent frequency window around the unperturbed tunnelling frequency. Outside this window the spin is hindered both in the high-frequency regime and in the adiabatic case. If the oscillating field is orthogonal to the anisotropy axis, as in (4), the spin is blocked completely in the high-frequency domain and seriously hampered in the low-frequency (adiabatic) range. Of course one might object that the high-frequency behaviour is to be expected 'since' a rapidly changing external field in general hampers precession. Precession, however, is in the classically allowed region whereas tunnelling is not. So hampering of tunnelling is not physically evident. It is a big advantage of the WKB technique that it allows a direct mathematical analysis of an ac field.

A circularly polarized field with its rotation axis parallel to the anisotropy axis, as in (5), leads to easy tunnelling at the frequencies $\omega = \Gamma, 2\Gamma, \ldots, S\Gamma$ and a blocking of the spin in between and as ω increases beyond $S\Gamma$. This result is exact, at least as far as time evolution is concerned. In the case where the direction of rotation is orthogonal to the anisotropy axis, no exact solution is known (yet) and we have to invoke the averaging method so as to infer that tunnelling is blocked. Spins are apparently hampered by an ac field, once the frequency is high enough. At first sight this might contradict intuition, but one has to realize that the time evolution operator, though linear, has the Hamiltonian *in the exponent*. Hence averaging is non-trivial.

On the basis of the present data we cannot but formulate the hypothesis that in general an ac field hampers spin tunnelling. If so, the behaviour of spins is in clear contrast with particle tunnelling [12] where, except for in the resonance case, a periodic driving *always enhances* the tunnelling rate. This makes mesoscopic spins a fascinating subject in their own right.

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References

[1] Pauli W 1933 *Handbuch der Physik* vol XXIV/1, ed H Geiger and K Scheel (Berlin: Springer) p 260 (reprinted in vol V/1 in 1958)
There also exists an English translation:
Pauli W 1980 *General Principles of Quantum Mechanics* (Berlin: Springer) section 12
[2] Bohm D 1951 *Quantum Theory* (Englewood Cliffs, NJ: Prentice-Hall) (reprinted 1989 (New York: Dover)) See in particular ch 12.
[3] Van Hemmen J L and Sütő A 1986 *Europhys. Lett.* **1** 481
Van Hemmen J L and Sütő A 1986 *Physica* B **141** 37
[4] Van Hemmen J L and Sütő A 1995 *Quantum Tunnelling of Magnetization–QTM'94* ed L Gunther and B Barbara (Dordrecht: Kluwer)
[5] Enz M and Schilling R 1986 *J. Phys. C: Solid State Phys.* **19** 1765
Enz M and Schilling R 1986 *J. Phys. C: Solid State Phys.* **19** L711
[6] Chudnovsky E M and Gunther L 1988 *Phys. Rev. Lett.* **60** 661
[7] Barbara B, Sampaio L C, Wegrowe J E, Ratnam B A, Marchand A, Paulsen C, Novak M A, Tholence J L, Uehara M and Fruchard D 1993 *J. Appl. Phys.* **73** 6703
[8] Chudnovsky E M 1993 *J. Appl. Phys.* **73** 6697
[9] Stamp P C E, Chudnovsky E M, and Barbara B 1992 *Int. J. Mod. Phys.* B **6** 1355
[10] Gunther L and Barbara B (ed) 1995 *Quantum Tunnelling of Magnetization–QTM'94* (Dordrecht: Kluwer)
[11] Großmann F, Dittrich T, Jung P and Hänggi P 1991 *Phys. Rev. Lett.* **67** 516
Großmann F, Jung P, Dittrich T and Hänggi P 1991 *Z. Phys.* B **84** 315
Großmann F 1992 *Doctoral Dissertation* University of Augsburg
[12] Dittrich T, Großmann F, Jung P, Oehlschlägel B and Hänggi P 1993 *Physica* A **194** 173
[13] Kleber M 1994 *Phys. Rep.* **236** 331
[14] Bogoliubov N N and Mitropolsky Yu A 1961 *Asymptotic Methods in the Theory of Nonlinear Oscillations* (New York: Gordon and Breach)
[15] Sanders J A and Verhulst F 1985 *Averaging Methods in Nonlinear Dynamical Systems* (New York: Springer)
[16] Abramowitz M and Stegun I A (ed) 1965 *Handbook of Mathematical Functions* (New York: Dover) equations 9.1.21, 9.1.62, and 6.1.49
[17] Watson G N 1922 *A Treatise on the Theory of Bessel Functions* (Cambridge: Cambridge University Press) section 2.2
[18] Tip A 1983 *J. Phys. A: Math. Gen.* **16** 3237
[19] Allen L and Eberly J H 1975 *Optical Resonance and Two-Level Atoms* (New York: Wiley) and references quoted therein
[20] Landau L D and Lifshitz E M 1959 *Quantum Mechanics* (London: Pergamon) problem of section 40
[21] Rabi I I 1937 *Phys. Rev.* **51** 652