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Quantum tunnelling of magnetization in an alternating magnetic field: localization and chaos

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Abstract. Tunnelling of mesoscopic quantum spins, i.e. magnetization, in a time-periodic external field is studied analytically. Three independent mechanisms of localization or blocking of the magnetization are isolated, namely the crossing of Floquet eigenvalues, special frequencies, and the breakdown of a symmetry which would allow tunnelling between degenerate minima. Symmetry breakdown is discussed in detail in the adiabatic region where the external field changes slowly. For high frequencies, we take advantage of the averaging method and are able to confirm that AC hampering is a rather general property. We also numerically show that spin hampering persists in the classically chaotic region, even outside the domain of applicability of the averaging method.

1. Introduction and summary

During the last decade, macroscopic quantum tunnelling of the magnetization has been a subject of intensive research, both theoretically [1–8] and experimentally; see in particular [6]. More recently, the problem of tunnelling of a spin (of large spin quantum number $S \gg 1$) under the influence of a perturbation periodic *in time* has aroused special interest in connection with the behaviour of mesoscopic magnetic moments in an anisotropic field [9]. In [9] the following three typical examples of a periodically driven spin system have been studied:

$$\hat{H}_1 = -\gamma S_z^2 - \alpha S_x - \delta S_z \cos(\omega t) \quad (1)$$

$$\hat{H}_2 = -\gamma S_z^2 - \alpha S_x \cos(\omega t) \quad (2)$$

$$\hat{H}_3 = -\gamma S_z^2 - \alpha[S_x \cos(\omega t) + S_y \sin(\omega t)] \quad (3)$$

with α, γ, δ positive. Throughout what follows, operators always wear a hat whereas their classical counterparts do not. The anisotropy is represented by $-\gamma S_z^2$ and dominates the other terms, i.e. $\alpha, \delta \ll \gamma \hbar S$, so that the tunnelling condition is satisfied. Given an *unperturbed* level splitting

$$\Delta E = \hbar \omega_0 \quad (4)$$

which is obtained, for example, by the setting $\delta = 0$ in \hat{H}_1 or $\omega = 0$ in \hat{H}_2 and \hat{H}_3 , we distinguish three cases, as in [9]:

- $\omega \gg \omega_0$, high frequency;
- $\omega \approx \omega_0$, resonance;
- $\omega \ll \omega_0$, adiabatic case.

The resonance case was analytically dealt with in [9], and we have nothing to add to its conclusions. In this paper, we shall complement the analysis of [9] in the other two cases. It is natural to compare the AC behaviour of \hat{H}_1 with that of $\delta = 0$ since $\delta \neq 0$ in general favours tunnelling in the particle case. For \hat{H}_2 and \hat{H}_3 , the reference state is $\omega = 0$. As compared with the time-independent ‘reference’ Hamiltonians, an AC field often seems to slow down tunnelling. We call this phenomenon ‘hampering’.

Before proceeding, it may be well to face the question of how the above Hamiltonians came about and whether spin quantum tunnelling can be observed experimentally [10–12]. Tunnelling of molecular spins has been found in Mn_{12} acetate crystals, where each molecule carries a spin of fixed angular momentum $10\hbar$ and experiences a *constant* magnetic field \mathbf{H} . The corresponding Hamiltonian is given by

$$\hat{H} = -\gamma S_z^2 - g\mu_B \mathbf{S} \cdot \mathbf{H}.$$

Replacing a constant \mathbf{H} by $\mathbf{H}(t) = (h_x, 0, h_z \cos \omega t)$ one obtains the Hamiltonian \hat{H}_1 . The Hamiltonians \hat{H}_2 and \hat{H}_3 can be realized equally straightforwardly.

The theoretical problem of how a giant quantum spin tunnels was solved surprisingly late in the evolution of quantum mechanics. It was 1986 when two types of solution appeared. Enz and Schilling [2] 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-integral technique. Van Hemmen and Sütő [1] started with the Schrödinger equation itself and developed a WKB formalism for quantum spins, which is used throughout the following. It allows for both 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 [8] to exactly agree with spin functional-integral results of Chudnovsky and Gunther [5]. The early papers did not allow the Hamiltonian to be time dependent. Lifting this restriction, then, seems more than timely. A first ansatz *à la* (1)–(3) was formulated by van Hemmen and Sütő [9].

In section 2 we study localization properties properly. We are able to identify *three* independent sources of *spin localization*, i.e. of how a spin gets stuck. We also study the adiabatic case in detail for all three Hamiltonians, pointing out some very interesting qualitative differences between them. Section 3 is devoted to a numerical study of the tunnelling probability in the high-frequency case for Hamiltonians \hat{H}_1 and \hat{H}_2 in different regions of the (δ, ω) plane, corresponding to classically integrable and chaotic dynamics. One of our motivations in this section was, and is, the striking phenomenon of ‘chaotic quantum tunnelling’ as studied for the particle case in [13–15]. Section 4 summarizes our results.

2. Mechanisms of localization

A beautiful feature of particle systems is the existence of apparently three independent mechanisms of localization: Anderson localization, localization by Cantori, and localization through scars of special periodic orbits [16]. Spins behave very differently from particles [1], but three (independent) mechanisms of spin localization in an AC field are now apparent.

- Crossing of Floquet eigenvalues.
- Localization, for instance, for a special sequence of values of δ/ω in \hat{H}_1 as furnished by the *averaging method* [9]. For the moment it looks as if this kind of localization and crossing of Floquet eigenvalues are two independent processes—exceptions are allowed—but a proof is still missing.

- Breakdown of a symmetry which allowed a tunnelling between degenerate minima.

For example, let \hat{R} denote a rotation through π about the x -axis. This is a symmetry of \hat{H}_2 , broken in \hat{H}_1 by the term $-\delta S_z \cos(\omega t)$, which thus destroys quantum coherence. We shall analyse all this in detail in the adiabatic regime.

2.1. Level crossing

Returning to the level crossing, we note that this type of localization has already been shown in the particle case [17]. We first discuss the analogues of the symmetry operator first introduced by Breuer *et al* [24]. In so doing, we assume a spectral representation with S_z diagonal: $S_z|m\rangle = \hbar m|m\rangle$ where $-S \leq m \leq S$ and S is the spin quantum number. Let $\sigma = \hbar S$ and s range through the interval $[-\sigma, \sigma]$, that would contain the spectrum of S_z in the limit $S \rightarrow \infty$ with $\sigma = \hbar S$ fixed. As long as S is finite, s samples the allowed eigenvalues of S_z and $S_x = a(s)(T_{\hbar} + T_{-\hbar})/2$. Here $(T_{\pm\hbar}f)(s) = f(s \pm \hbar)$ defines a shift operator and $a(s) \equiv \sqrt{\sigma(\sigma + \hbar) - s^2}$. We define the symmetry \hat{V} by

$$\hat{V} = \hat{R} \hat{\tau}_{\frac{T}{2}} \tag{5}$$

where \hat{R} is the rotation through π about the x -axis introduced before; in the spectral representation, it acts as $(\hat{R}f)(s, t) = f(-s, t)$. Furthermore, $\hat{\tau}_{\frac{T}{2}}$ is the shift by half a period in the time variable:

$$(\hat{\tau}_{\frac{T}{2}}f)(s, t) = f\left(s, t + \frac{T}{2}\right). \tag{6}$$

The operator \hat{V} is unitary,

$$\hat{V}^* = \hat{\tau}_{\frac{T}{2}}^* \hat{R}^* = \hat{\tau}_{-\frac{T}{2}} \hat{V}^* = \hat{V}^{-1}. \tag{7}$$

Furthermore, \hat{V} commutes with $\hat{H}_1(t)$ for all times,

$$\begin{aligned} (\hat{V} \hat{H}_1(t) f)(s, t) &= -[\gamma S^2 + \delta S \cos(\omega t)] f\left(-s, t + \frac{T}{2}\right) \\ &\quad + \frac{1}{2} a(s) \left[f\left(-s + \hbar, t + \frac{T}{2}\right) + f\left(-s - \hbar, t + \frac{T}{2}\right) \right] \\ &= (\hat{H}_1 \hat{V} f)(s, t). \end{aligned} \tag{8}$$

The monodromy matrix $U(T)$ is defined as the unitary time evolution operator $U(t)$ evaluated one period later, i.e. at time $t = T$. The operator $U(t)$ being unitary, one can write $U(T) = \exp(iH_F T)$ for some Hermitian H_F , whose eigenvalues are called *Floquet* eigenvalues. They are defined mod $2\pi/T = \omega$. If we want to indicate which Hamiltonian is involved, we write $U_i(T)$ with $i = 1, 2, 3$ specifying one of the Hamiltonians listed in (1)–(3). Because of (8), \hat{V} also commutes with the time-evolution operator associated with $\hat{H}_1(t)$, and hence with the monodromy matrix,

$$[\hat{V}, U_1(T)] = 0. \tag{9}$$

Accordingly, Floquet eigenvalues *may cross* upon variation of a single real parameter (ω or δ , keeping the other ones fixed) as long as the eigenstates in the two branches that cross correspond to different eigenvalues of \hat{V} . In order to see this we use a von Neumann–Wigner argument [19]. The gist of their argument is ingenious but quite simple. Let us take a complex matrix whose elements depend on some real parameters. For instance, a Hamiltonian is, in general, a complex Hermitian matrix. It is obvious that this kind of

symmetry as well as ‘accidental’ degeneracies (which will be neglected) reduce the number of free parameters. Taking advantage of the spectral theorem one counts the remaining free real parameters of the matrix *with and without* a degenerate eigenvalue. Their difference D gives us the number of parameters that have to be varied in order to produce a level crossing. It is three for a complex Hermitian matrix and two for a real symmetric matrix.

We now show that D is one in the present case. To this end, we focus on the local behaviour of the eigenvalues $u_+(\vartheta)$ and $u_-(\vartheta)$ of $U_1(T, \vartheta) = U_1(T)$ where ϑ stands for the set of parameters δ, ω . Restricting the monodromy matrix to the degenerate subspace at ϑ we obtain a 2×2 matrix-valued function of ϑ .

$$\tilde{M}(\vartheta) = \begin{pmatrix} \langle \Psi | U_1(T, \vartheta) | \Psi \rangle & \langle \Psi | U_1(T, \vartheta) | \Phi \rangle \\ \langle \Phi | U_1(T, \vartheta) | \Psi \rangle & \langle \Phi | U_1(T, \vartheta) | \Phi \rangle \end{pmatrix}.$$

The eigenvalues of $\tilde{M}(\vartheta)$ are

$$u_{\pm}(\vartheta) = \frac{1}{2} [\langle \Psi | U_1(T, \vartheta) | \Psi \rangle + \langle \Phi | U_1(T, \vartheta) | \Phi \rangle] \pm [(\langle \Psi | U_1(T, \vartheta) | \Psi \rangle - \langle \Phi | U_1(T, \vartheta) | \Phi \rangle)^2 + |\langle \Psi | U_1(T, \vartheta) | \Phi \rangle|^2]^{1/2}.$$

Hence, equality of the eigenvalues, $u_+(\vartheta) = u_-(\vartheta)$, is achieved by imposing three real conditions:

$$\langle \Psi | U_1(T, \vartheta) | \Psi \rangle = \langle \Phi | U_1(T, \vartheta) | \Phi \rangle \quad (10)$$

$$\Re\{\langle \Psi | U_1(T, \vartheta) | \Phi \rangle\} = 0 \quad \Im\{\langle \Psi | U_1(T, \vartheta) | \Phi \rangle\} = 0. \quad (11)$$

The eigenvalues of U_1 are nondegenerate, and therefore are also eigenvalues of \hat{V} by (9). Suppose now that the two ‘crossing’ eigenvectors $|\Psi\rangle$ and $|\Phi\rangle$ correspond to *distinct* eigenvalues $\exp(i\phi_1)$ and $\exp(i\phi_2)$ of \hat{V} ; note that \hat{V} is unitary by (7). By (9) we find

$$\langle \Psi | U_1(T, \vartheta) | \Phi \rangle = \langle \Psi | \hat{V}^* U_1(T, \vartheta) \hat{V} | \Phi \rangle = \langle \Psi | U_1(T, \vartheta) | \Phi \rangle e^{i(\phi_1 - \phi_2)}.$$

By hypothesis, $\exp[i(\phi_1 - \phi_2)] \neq 1$, and therefore $\langle \Psi | U_1(T, \vartheta) | \Phi \rangle = 0$, which means that (11) is automatically satisfied and equality of the eigenvalues is achieved by imposing just one real condition, namely (10). Since $u_{\pm}(\vartheta) = \exp[-i\varepsilon_{\pm}(\vartheta)T]$, with $\varepsilon_{\pm}(\vartheta)$ defined modulo $2\pi/T = \omega$, it is thereby also expected that the set of crossings of Floquet eigenvalues $\varepsilon_{\pm}(\vartheta)$ have codimension 1. The same conclusions apply to \hat{H}_2 upon replacing \hat{V} by \hat{R} , which is a symmetry of \hat{H}_2 . For \hat{H}_3 , we define a different dynamical symmetry,

$$W = \hat{R}' \tau_{\frac{T}{2}} \quad (12)$$

where \hat{R}' denotes rotation through π about the z -axis. With this symmetry, the same conclusions hold for \hat{H}_3 . We note that in special cases the two distinct eigenvalues of \hat{V} are $+1$ and -1 [17]. However, \hat{V} does not have the property $\hat{V}^2 = \hat{V}$, and therefore the designation of ‘generalized parity’ may be misleading.

The same general arguments suggesting that crossings leads to localization, which have been discussed for the particle case in [17], are also applicable to our models. Accordingly, we refer to crossing as the first mechanism of localization.

2.2. Averaging method and special frequencies

The existence of ‘special frequencies’ has been proven for \hat{H}_1 by an application [9] of the averaging method to the WKB analysis of [1, 8]. The averaging method is a very useful technique [20] dating back to Lagrange (late 18th century). Special cases have frequently been rediscovered. For example, Kayanuma [21] explained a localization result of Großmann and Hänggi [22], that has a direct bearing upon the particle analogue of

the present case, by exploiting the two-level approximation. His high-frequency solution ($\omega \gg \Delta$) is a good illustration of the averaging technique, that is by no means restricted to the two-level atom. Neither is the argument of [9]. Assuming that the initial state is an eigenstate $|m\rangle$ of S_z , the following conditions are required for this type of localization [9]: $\alpha S \ll \omega$ and

$$\delta/\omega = z_\nu \quad \nu = 1, 2, \dots \tag{13}$$

where z_ν is one of the zeros of the zeroth-order Bessel function J_0 . Combined with the tunnelling condition $\delta, \alpha \ll \gamma S$, this implies that $\alpha S \ll \omega \ll \gamma S/z_\nu$. So the larger z_ν is, the narrower the region of parameter space is where the localization mechanism is active.

By the von Neumann–Wigner argument discussed above in the context of the high-frequency case, Floquet eigenvalues may cross along a curve in the (δ, ω) plane. When a two-level approximation holds, this curve coincides with (13), with $\nu = 1$; the arguments have been spelt out for the particle case in [21, 22, 25] and hold for our models with obvious modifications. Again by the same general reasoning as discussed in [25], the two-level approximation is not valid for ‘too small an ω ’, which also shows that the two mechanisms listed above, crossing and averaging, are independent. Since both the crossing mechanism and the averaging method are effective for $\omega \gg \omega_0$ (as for the level crossing, not exclusively), the third type of localization, which occurs in the adiabatic regime $\omega \ll \omega_0$ and is to be discussed next, is manifestly independent of the previous two.

2.3. Breakdown of quantum coherence

We now turn to the third mechanism of localization, namely a breakdown of a discrete symmetry such as \hat{R} . Following Leggett [7] we call this symmetry breaking ‘breakdown of quantum coherence’. Our main result is the localization of *low-lying* states, e.g. $|\pm S\rangle$, for the dynamics defined by \hat{H}_1 and \hat{H}_3 in the adiabatic regime. We also show that, when ω is ‘not too small’, *all* states $|\pm m\rangle$ with $m \in [-S, S]$, are effectively localized, if the dynamics is generated by \hat{H}_3 .

Let us consider \hat{H}_1 in the adiabatic regime first. If $\omega \ll \omega_0$, we may *fix* the value of t and study the tunnelling induced by the (quasi)static Hamiltonian

$$\hat{H}_1 = -\gamma S_z^2 - \alpha S_x - \lambda S_z \tag{14}$$

where $\lambda = \delta \sin(\omega t)$ is now a slowly varying function of t . We adopt a two-level approximation as in section 9 of [8]. In order to compute the splitting of the energy level $E_0 = -\gamma S^2$, we restrict the powers of

$$\hat{H} = \hat{H}_0 - \hat{V}$$

with

$$\begin{aligned} \hat{H}_0 &= -\gamma S_z^2 - \lambda S_z \\ \hat{V} &= \alpha S_x = \frac{\alpha}{2}(S_+ + S_-) \end{aligned}$$

to the subspace spanned by $|\pm S\rangle$, diagonalize them and consider \hat{V} to be small. The eigenvalues of the matrix H^k in this subspace can be written as the k th power of some numbers E_{1k} and E_{2k} , which can be considered as approximations of the true lowest two energies. Then we obtain

$$E_{2k}^k - E_{1k}^k = (E_{2k} - E_{1k})(E_{2k}^{k-1} + E_{2k}^{k-2}E_{1k} + \dots + E_{1k}^{k-1}) \approx (E_{2k} - E_{1k})kE_0^{k-1}$$

for which the level splitting $\Delta E = E_{2k} - E_{1k}$ can be computed.

In the space spanned by $|\pm S\rangle$, the lowest nonvanishing order k corresponds to $k = 2S$. To leading order $\alpha/(\gamma S) \ll 1$, we are thus led to diagonalizing the matrix

$$\tilde{M} = \begin{pmatrix} \langle S|\hat{H}_0^{2S}|S\rangle & \langle S|\hat{V}^{2S}|S\rangle \\ \langle -S|\hat{V}^{2S}|S\rangle & \langle -S|\hat{H}_0^{2S}|S\rangle \end{pmatrix}. \quad (15)$$

On the diagonal positions of \tilde{M} we have neglected matrix elements of the form $\langle S|S_+^{k_0}\hat{H}_0^{2S-2k_0}S_-^{k_0}|S\rangle$ as compared with $\langle S|\hat{H}_0^{2S}|S\rangle$ because of the condition $\alpha/(\gamma S) \ll 1$. There are, however, many such terms and we shall return to them at the end of this section. Assuming, for the moment, that (15) is a good approximation, we end up with

$$\langle \pm S|\hat{H}_0^{2S}|\pm S\rangle = (\gamma S^2 \pm \lambda S)^{2S} \approx (\gamma S^2)^{2S} \pm 2S^2\lambda(\gamma S^2)^{2S-1} = (\gamma S^2)^{2S} \left(1 \pm \frac{2\lambda}{\gamma}\right) \quad (16)$$

while

$$\langle S|\hat{V}^{2S}|S\rangle = \langle -S|\hat{V}^{2S}|S\rangle = \alpha^{2S}\langle S|S_+^{2S}|S\rangle = \alpha^{2S}(2S)! \quad (17)$$

The eigenvalues ε_{\pm} of \tilde{M} are

$$\begin{aligned} \varepsilon_{\pm} &= \frac{1}{2}(\text{tr } M) \pm \frac{1}{2}\sqrt{(\text{tr } M)^2 - 4 \det M} \\ &= (\gamma S^2)^{2S} \pm \sqrt{(\gamma S^2)^{4S} - (\gamma S^2)^{4S} \left[1 - \left(\frac{2\lambda}{\gamma}\right)^2\right] + \alpha^{4S}(2S)!^2} \\ &= (\gamma S^2)^{2S} \left\{ 1 \pm \sqrt{\left(\frac{2\lambda}{\gamma}\right)^2 + \left(\frac{\alpha}{\gamma S^2}\right)^{4S} (2S)!^2} \right\}. \end{aligned} \quad (18)$$

Let

$$\Delta \equiv \left(\frac{\alpha}{\gamma S^2}\right)^{4S} (2S)!^2 \approx \left(\frac{2\alpha S}{e\gamma S^2}\right)^{4S} = \left(\frac{2\alpha}{e\gamma S}\right)^{4S}. \quad (19)$$

By obtaining the approximate equality we have exploited Stirling's theorem [23]. The quantity Δ is very small due to the tunnelling condition. We thus find

$$\varepsilon_{\pm} \approx (\gamma S^2)^{2S} \left\{ 1 \pm \left(\frac{2|\lambda|}{\gamma}\right) \left[1 + \frac{1}{2} \left(\frac{\gamma}{2\lambda}\right)^2 \Delta\right] \right\}. \quad (20)$$

Suppose $\lambda > 0$. Then ε_+ corresponds to the ground state. The corresponding eigenvalue equation is

$$M \begin{pmatrix} a_1 \\ a_2 \end{pmatrix} = \varepsilon \begin{pmatrix} a_1 \\ a_2 \end{pmatrix}$$

leading to

$$(\gamma S^2)^{2S} \left(1 + \frac{2\lambda}{\gamma}\right) a_1 + \alpha^{2S}(2S)! a_2 = (\gamma S^2)^{2S} \left(1 + \frac{2|\lambda|}{\gamma}\right) \left[1 + \frac{1}{2} \left(\frac{\gamma}{2\lambda}\right)^2 \Delta\right].$$

Hence, we obtain for $\lambda > 0$ and using (19)

$$\begin{aligned} \frac{a_2}{a_1} &= \frac{\Delta}{8(2S)!} \left(\frac{\gamma S^2}{\alpha}\right)^{2S} \left(1 + \frac{2|\lambda|}{\gamma}\right) \left(\frac{\gamma}{\lambda}\right)^2 \\ &= \frac{1}{8(2S)!} \left(\frac{\alpha}{\gamma S^2}\right)^{2S} \left(1 + \frac{2|\lambda|}{\gamma}\right) \left(\frac{\gamma}{\lambda}\right)^2. \end{aligned} \quad (21)$$

Now again by Stirling's theorem [23]

$$\frac{1}{(2S)!} \left(\frac{\alpha}{\gamma S^2}\right)^{2S} \approx \left(\frac{2S\alpha}{e\gamma S^2}\right)^{2S} = \left(\frac{2\alpha}{e\gamma S}\right)^{2S}$$

so that a_2/a_1 is exponentially small as $\alpha/(\gamma S) \ll 1$ due to the tunnelling condition and, thus, the spin is to be localized. If $\lambda < 0$, then ε_- corresponds to the ground state and we find that a_1/a_2 is exponentially small; as expected, the spin 'gets stuck' in one of the wells and quantum coherence is broken.

Expression (18) is a key issue due to two reasons. First, it shows clearly that here we have to require

$$2\lambda \ll \gamma \tag{22}$$

since otherwise perturbation theory is senseless. Furthermore, it indicates how small λ must be in order for localization to break down. When both terms under the square root in (18) are of equal magnitude so that

$$\frac{2\lambda}{\gamma} \approx \left(\frac{\alpha}{\gamma S^2}\right)^{4S} (2S)!^2 \approx \left(\frac{2\alpha}{e\gamma S}\right)^{2S} . \tag{23}$$

Formulated as an understatement, λ has to be rather small.

We now return to approximation (15). We have estimated the diagonal terms $\langle \pm S | (\hat{H}_0 + \hat{V})^{2S} | \pm S \rangle$ by $\langle \pm S | \hat{H}_0^{2S} | \pm S \rangle$. We write symbolically

$$(\hat{H}_0 + \hat{V})^{2S} = \hat{H}_0^{2S} + \sum_{k \neq 2S} \{ \hat{H}_0^k \hat{V}^{2S-k} \}$$

where, in the last sum, all possible orderings of \hat{V} relative to \hat{H}_0 —and consistent with there being $(2S - k)$ operators \hat{V} , and k operators \hat{H}_0 —are included. The operator norm of \hat{H}_0^{2S} is

$$\|\hat{H}_0^{2S}\| = \sup_{\|\Phi\|=\|\Psi\|=1} |\langle \Phi | \hat{H}_0^{2S} | \Psi \rangle| = (\gamma S^2 + |\delta| S)^{2S} = \|\hat{H}_0\|^{2S}$$

and hence

$$\begin{aligned} |\langle \pm S | \sum_{k \neq 2S} \{ \hat{H}_0^k \hat{V}^{2S-k} \} | \pm S \rangle| &\leq \sum_{k \neq 2S} \|\hat{H}_0^k\| \|\hat{V}^{2S-k}\| \leq \sum_{k \neq 2S} \|\hat{H}_0^k\| \|\hat{V}\|^{2S-k} \\ &= \|\hat{H}_0^{2S}\| \frac{(\|\hat{H}_0\| + \|\hat{V}\|)^{2S} - \|\hat{H}_0\|^{2S}}{\|\hat{H}_0^{2S}\|} \\ &= \left[\left(1 + \frac{\|\hat{V}\|}{\|\hat{H}_0\|} \right)^{2S} - 1 \right] \|\hat{H}_0\|^{2S} \\ &\leq \exp \left(2S \frac{\|\hat{V}\|}{\|\hat{H}_0\|} \right) \|\hat{H}_0\|^{2S}. \end{aligned} \tag{24}$$

We have $\|\hat{V}\| = \alpha S/2$ and $\|\hat{H}_0\| = \gamma S^2 + |\delta| S$. If, then,

$$2S \frac{\|\hat{V}\|}{\|\hat{H}_0\|} \approx 2S \frac{\alpha S}{2\gamma S^2} = \frac{\alpha}{\gamma} \ll 1 \tag{25}$$

it may easily be checked from (24) that the conclusions using (15) are unaffected, the corrections being small.

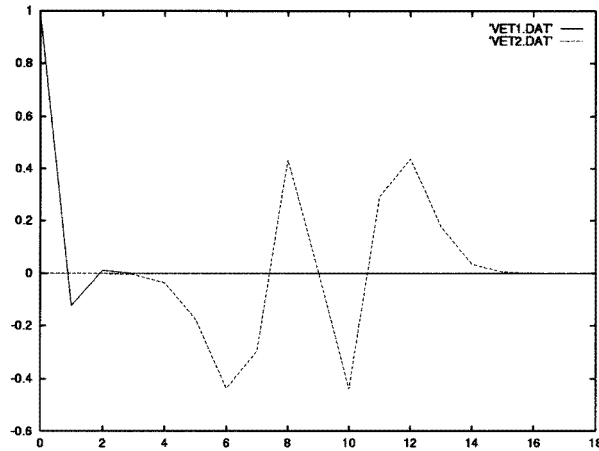


Figure 1. Eigenvectors of the Hamiltonian $\hat{H} = -\gamma S_z^2 - \lambda S_z + \alpha S_x$ with parameters $\gamma = 1$, $\alpha = 1$, and $\lambda = 0.01$. The vertical axes show the components of each eigenvector as a function of S_z , which is diagonal. In the legend, *vet1.dat* represents a ground-state eigenvector. It is localized. The same does not hold for *vet2.dat*, which represents an eigenvector that corresponds to an excited state.

The right-hand side of (23) is an extremely small number, of the order of ΔE given by (4). In fact, we know that tunnelling is restored in (14) for $\lambda \rightarrow 0$. Since tunnelling lifts the degeneracy of the eigenstates of

$$\tilde{H}_1(\lambda = 0) = -\gamma S_z^2 - \alpha S_x$$

for $\alpha \neq 0$, their projection operators are strongly analytic [27], whatever λ , since in general no crossing occurs. That is, once $\lambda\gamma$ is of the order of the level splitting for $\lambda = 0$, localization breaks down. The point is that the eigenstates of $\tilde{H}_1(\lambda = 0)$ are also eigenstates of \hat{R} and, thus, localized in *both* wells. This somewhat subtle feature is a pure quantum phenomenon due to tunnelling and should disappear in the classical limit $S \rightarrow \infty$ and $\hbar \rightarrow 0$ with $\hbar S = \sigma$ constant, say, $\sigma = 1$. It is, however, not obvious how to prove such a statement technically. For the sake of completeness this is done in appendix C.

The restriction to the *low-lying* states in the above result is *not* a technicality. If the state we start with is an excited state $|m\rangle$ for which $m \approx 0$, the perturbation $-\lambda S_z$ in (14) has little effect. Hence, it is plausible that tunnelling will persist. This is shown in figure 1.

We now consider \hat{H}_3 . It has been shown in [9] that the evolution operator $U_3(t, t_0)$ from t_0 to t may be written

$$U_3(t, t_0) = \exp\left(-\frac{i\omega}{\hbar} t S_z\right) \exp\left[\frac{i}{\hbar} (t - t_0) \hat{H}_0\right] \exp\left(\frac{i\omega}{\hbar} t S_z\right) \quad (26)$$

where

$$\hat{H}_0 = -\gamma S_z^2 - \alpha S_x - \omega S_z. \quad (27)$$

This result is exact. By (26), the tunnelling amplitude between eigenstates $|m\rangle$ and $| -m\rangle$ of S_z equals

$$|\langle -m | U_3(t, t_0) | m \rangle| = |\langle -m | e^{\frac{-i}{\hbar} (t-t_0) \hat{H}_0} | m \rangle| \quad (28)$$

and, therefore, \hat{H}_0 is the *exact* effective Hamiltonian for tunnelling—for all times!

Since ω in (27) plays the role of λ in (14), we may thus assert, by (22) and (23), that if

$$\left(\frac{2\alpha}{e\gamma S}\right)^{2S} \ll \frac{2\omega}{\gamma} \ll 1 \tag{29}$$

then there is *localization*. This concludes the analysis of \hat{H}_3 . Note, however, that due to (29) we have $\omega \gg \omega_0$. That is to say, we are still in the high-frequency region and nothing can be said about the behaviour under $\hat{H}_3(t)$ in the adiabatic regime.

We finally turn to \hat{H}_2 . A folklore dictum (for which, even in the atomic case, no proof exists; see [18] and references quoted therein) asserts that in the *adiabatic* limit the *average* of the transition rate over one period (T),

$$\langle \Gamma_{\text{DC}}^{\hat{H}_2} \rangle = \frac{1}{T} \int_0^T dt \Gamma_{\text{DC}}^{\hat{H}_2}(\alpha |\cos(\omega t)|) \tag{30}$$

is the leading term in a (presumably asymptotic) expansion of the transition (in our case, tunnelling) rate in powers of $\omega = 2\pi/T$. Here $\Gamma_{\text{DC}}^{\hat{H}_2}(\lambda)$ is the tunnelling rate for the *static* model

$$\hat{H}_2 = -\gamma S_z^2 - \lambda S_x \tag{31}$$

with (fixed) index λ . We denote the time average by angular brackets. In our case, with $c := |E|/(\alpha\sigma)$, $d := 2S$, and $|E| = \gamma\sigma^2$, we have to evaluate [1, 8]

$$\int_0^{\frac{\pi}{2}} dx \exp\left[-c \ln \frac{d}{\cos x}\right] = \int_0^{\frac{\pi}{2}} dx \left(\frac{\cos x}{d}\right)^c.$$

Wallis' formula [28] provides us with the estimate

$$\frac{2}{\pi} \int_0^{\frac{\pi}{2}} dx x \left(\frac{\sin}{\cos}\right)^{2n} (x) = \frac{\Gamma(n + \frac{1}{2})}{\sqrt{\pi}\Gamma(n + 1)} \approx \frac{1}{\sqrt{\pi n}} \left(1 - \frac{1}{8n} + \frac{1}{128n^2} + \dots\right).$$

We therefore obtain

$$\langle \Gamma_{\text{DC}}^{\hat{H}_2} \rangle \approx \Gamma_{\omega=0} \left(\frac{\alpha}{\gamma\sigma}\right)^{\frac{1}{2}} \tag{32}$$

where

$$\Gamma_{\omega=0} = \tau_0^{-1} \left(\frac{\alpha\sigma}{|E|}\right)^{2S} \tag{33}$$

is the dominant term in the tunnelling rate [1] with τ_0^{-1} as an attempt frequency. Equation (32) shows that tunnelling persists for \hat{H}_2 in the adiabatic regime (in contrast to \hat{H}_1), albeit at a slightly lower rate.

We have now completed our analysis of the localization properties of Hamiltonians \hat{H}_1 , \hat{H}_2 , and \hat{H}_3 . It includes both the adiabatic region $\omega \ll \omega_0$, and the region where blocking of the spin may be proved by the averaging method, which requires $\omega_0 \ll \omega$ and $\omega \gg \alpha S$; the latter overlaps the region where crossings occur. It is natural to investigate the complementary domain, where $\omega_0 \ll \omega \ll \alpha S$: Does localization hold here too? Answering this question will be the subject of section 3.

3. Chaotic quantum tunnelling

The tunnelling of particles was studied from a completely different point of view than ours in [13–15]. The presumably most striking result is due to Bohigas *et al* [14]. For a special quantum billiard, they found that the tunnelling rate is dramatically enhanced *at the classical chaos border*. Despite several efforts, a comprehensive theory of this phenomenon does not seem to exist.

It may well be that our models provide the simplest nontrivial example of classically chaotic dynamics. This is exemplified in appendix B by the calculation of the *stochasticity parameter* for \hat{H}_2 . We do so by using the Chirikov criterion of resonance overlap,

$$s_p = 2\sqrt{\frac{\alpha}{\gamma\omega}}. \quad (34)$$

That is, for $s_p < 1$ the classical system is regular whereas it is in the chaotic regime for $s_p > 1$. Note that s_p becomes large as $\omega \rightarrow 0$. There exists, however, a small region around $\omega = 0$ where integrability is restored and adiabatic invariants survive everywhere in phase space—except for a set of small Lebesgue measure, proportional to ω [29]. The other simplifying feature (in comparison with [16]) is the finite dimension of the Hilbert space, which eliminates the need of truncation.

We are going to study the tunnelling probability as a function of time for the Hamiltonians \hat{H}_1 and \hat{H}_2 , which we modify slightly for technical reasons related to the numerics. We now consider

$$\hat{H}_1 = -\frac{\gamma S_z^2}{S} + \alpha S_x + \delta S_z \sin(\omega t) \quad (35)$$

$$\hat{H}_2 = -\frac{\gamma S_z^2}{S} + \alpha (1 + \sin(\omega t)) S_x. \quad (36)$$

We first consider the classical dynamics for \hat{H}_1 . The classical Hamiltonian $h = h(q, p)$ is

$$h(S_z, \phi) = -\gamma S \cos^2 \theta + \alpha S \sin \theta \cos \phi + \delta S \cos \theta \sin \omega t \quad (37)$$

where $S_x = S \sin \theta \cos \phi$, $S_y = S \sin \theta \sin \phi$, and $S_z = S \cos \theta$. The canonically conjugate variables for this system [1] are $q = S_z$ and $p = -\phi$, and therefore the equations of motions are

$$\dot{\phi} = \frac{\partial h}{\partial S_z} = -2\gamma \cos \theta + \delta \sin(\omega t) - \alpha \coth \theta \cos \phi \quad (38)$$

and

$$\dot{S}_z = -\frac{\partial h}{\partial \phi} = \alpha S \sin \phi \sin \theta. \quad (39)$$

Figures 2–5 exhibit Poincaré sections for the classical version of the Hamiltonian (1) with fixed $\gamma \rightarrow \gamma/S$ and *increasing* δ , and show that in this way we enter the classically chaotic regime. The borderline of chaotic motion is given by $\delta = 0.5$, in agreement with the occurrence of the first avoided crossing.

The Poincaré sections are obtained by the stroboscopic map, as is usual in the case of a periodically driven Hamiltonian system. For the sake of comparison, $S = 9$ and the parameters α and γ equal those in [1]. The Floquet spectrum is also shown as a function of δ for $\omega = 0.5$.

As an important preliminary check, we have verified for \hat{H}_1 that we obtain the right limit behaviour as $\omega \rightarrow \infty$. By first-order perturbation theory, the Floquet eigenvalues

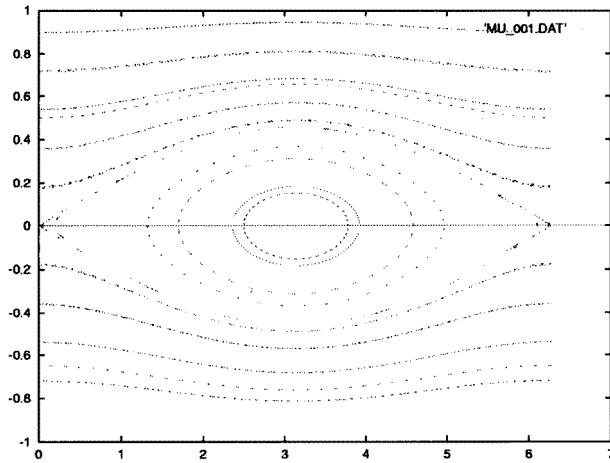


Figure 2. Poincaré section for the classical Hamiltonian $h = -\gamma S \cos^2 \theta + \alpha S \sin \theta \cos \phi + \delta S \cos \theta \sin \omega t$. Its quantum analogue is \hat{H}_1 . We have taken $\gamma = 1$, $\alpha = 1$, $S = 9$, $\omega = 0.5$, and $\delta = 0.001$. The vertical axis represents S_z/S and the horizontal axis represents ϕ . The above domain is inside the integrable region and corresponds to case A in figure 6.

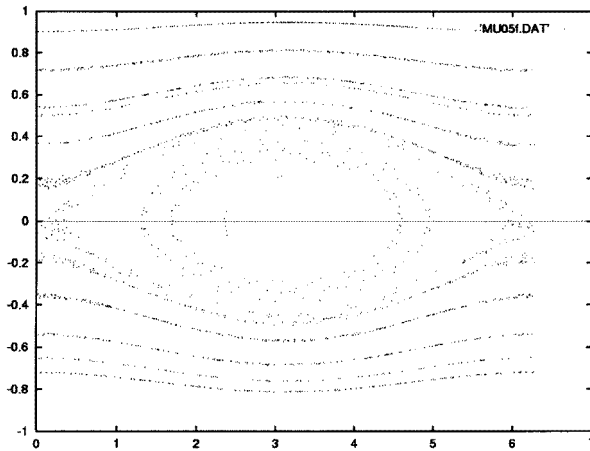


Figure 3. As in figure 2, except for $\delta = 0.5$. Here we are at the borderline between the integrable and chaotic region.

should agree with the values of [1] (after multiplication by $2\pi/\omega$ and reduction mod 2π —see table 1) to order δ/ω . This is exemplified by table 1 and is an important check of our numerical results.

In order to determine the Floquet spectrum, we have integrated the differential equations of appendix A, using unitarity violation of the monodromy matrix as an additional check on global errors.

The usual Runge–Kutta error estimate (see appendix A) is not very reliable, and therefore table 1 provides a crucial check on our numerical results. Note that, for $S = 9$, the tunnelling rate is of the order of 10^{-18} requiring a minimum of 20 meaningful digits and necessarily quadruple precision.

We now present our numerical result for the tunnelling probability as a function of time.

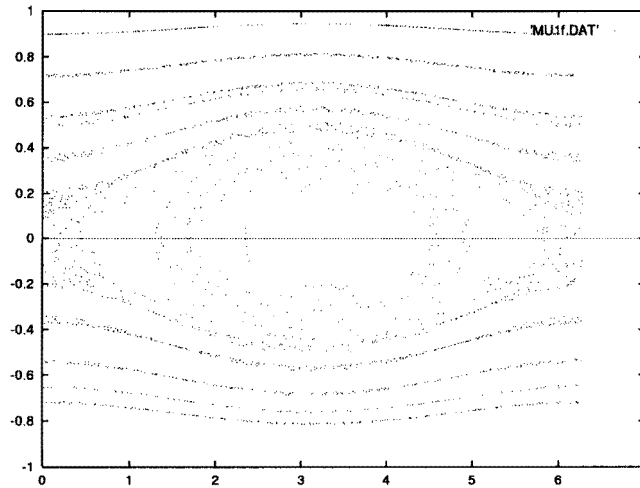


Figure 4. As in figure 2, except for $\delta = 1$. This is case B in figure 6. We are now in the chaotic regime.

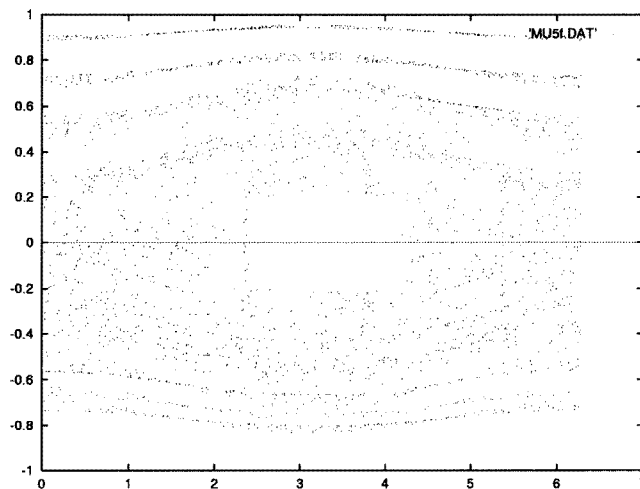


Figure 5. As in figure 2, except for $\delta = 5$. We are deeply inside the chaotic region. Note that our initial state at $S_z/S = \frac{6}{9} = 0.66$ does not lie in a regular region.

As in the autonomous case, the tunnelling probability does not reach the value 1; this would only occur in the limit $S \rightarrow \infty$.

In Figure 6 we display the tunnelling probability as a function of time in three cases: the static one ST (after van Hemmen and Sütő [1]), (A) the case $\omega = 0.5$ and $\delta = 0.001$, and (B) the classically chaotic region $\omega = 0.5$, $\delta = 1$. We have kept $\gamma = 9$ and $\alpha = 1$ throughout and started with the initial state $|m = 6\rangle$, which has a very small splitting ($\approx 10^{-7}$) in the static case. It is seen that close agreement exists between ST and A. In the transition from classically integrable to chaotic behaviour, namely (A) \rightarrow (B), which roughly corresponds to the variation in the Poincaré sections of figures 2–4, an opposite effect to the one apparently observed in [15] is verified: the tunnelling probability is reduced

Table 1. Check of the results for the model \hat{H}_1 of the static Hamiltonian. These eigenvalues were obtained with $\gamma = 9$, $\delta = 0.001$, $\omega = 500$, $\alpha = 1$. E_c are the eigenvalues obtained by diagonalization of the monodromy matrix which are of the form $u_n = e^{i\epsilon_n \frac{2\pi}{\omega}}$. So, to obtain the values above, one must calculate $i \log(u_n)$. These values must be multiplied by $\omega/2\pi$ and confronted with the values obtained for van Hemmen (E_{vH}) for the eigenvalues of the static Hamiltonian $\delta = 0$. β is the estimated precision.

E_c	$\times \frac{\omega}{2\pi}$ ($\omega = 500$)	E_{vH}	β
-9.220 291 881 760 114 850 200 550 89E - 2	79.57	7.337 283 483 003 323 805 035 273 80	10^{-30}
-3.980 741 518 036 502 834 887 348 88E - 2	79.57	3.167 779 460 356 240 263 315 887 5810	10^{-30}
8.429 476 322 849 272 559 398 102 54E - 3	79.57	-0.670 792 728 941 682 368 583 472 3931	10^{-30}
5.216 498 782 563 983 592 889 967 72E - 3	79.57	-4.151 156 421 101 217 941 323 506 9306	10^{-30}
8.774 637 245 986 085 926 460 979 72E - 2	79.57	-6.982 636 303 677 337 320 169 798 599	10^{-30}
0.136 552 359 392 034 079 030 707 79	79.57	-10.866 495 939 805 844 893 146 076 409	10^{-30}
0.128 325 790 112 240 690 849 646 04	79.57	-10.211 843 705 063 453 038 139 245 735	10^{-30}
0.211 805 495 502 701 898 332 715 72	79.57	-16.854 947 534 101 353 083 524 721 593	10^{-30}
0.211 962 963 232 154 197 519 280 41	79.57	-16.867 478 511 550 537 141 389 124 295	10^{-30}
0.321 507 652 906 522 877 484 110 21	79.57	-25.584 768 402 233 723 023 730 743 735	10^{-30}
0.321 508 257 797 453 050 819 715 56	79.57	-25.584 816 538 382 275 536 760 504 660	10^{-30}
0.457 937 623 528 463 812 541 466 68	79.57	-36.441 593 843 858 335 350 740 052 50	10^{-30}
0.457 937 622 177 687 020 445 729 70	79.57	-36.441 593 781 100 890 451 626 824 570	10^{-30}
0.620 169 469 642 367 619 500 283 36	79.57	-49.356 707 032 560 733 890 838 852 595	10^{-30}
0.620 169 393 454 196 292 292 656 06	79.57	-49.356 707 032 529 031 612 824 085 5	10^{-30}
1.024 425 673 990 868 650 911 511 6	79.57	-81.264 720 225 331 333 361 417 146 7	10^{-30}
0.804 886 347 272 543 971 539 195 27	79.57	-64.302 042 358 895 243 941 990 245 5	10^{-30}
0.804 882 757 462 375 096 355 065 20	79.57	-64.302 042 358 895 237 751 109 080 5	10^{-30}
1.024 429 341 102 735 535 170 714 5	79.57	-81.264 720 225 331 333 361 037 678 9	10^{-30}

from a finite value of the order of 10^{-7} to a blocking of the spin in the chaotic region (B,C)! This blocking of the spin is not rigorously accounted for by the averaging method of [9], because the condition $\omega \gg \alpha S$ does not hold here, but it is clear that even in this region the behaviour of spins differs qualitatively from the particle case.

There may be two independent reasons for the above surprising behaviour, which are different from both [15] and the more comprehensive theory of [30]. First, the classical transport mechanism, diffusion inside the chaotic region of phase space, depends on the existence of a symmetry-related doublet which remains in a regular region even after chaos becomes global. The anharmonic oscillator [13, 15] exhibits this behaviour but our model does not. See figure 5 where the initial state becomes immersed in the chaotic region. Secondly, we have not followed the behaviour of doublets of Floquet eigenvalues, as in [15]. In our model Hamiltonian \hat{H}_1 , the splitting of such doublets indeed grows with increasing δ (for fixed ω), i.e. when we gradually penetrate the chaotic region of parameters. This is, however, mostly an effect of the perturbation, which in our case is diagonal in the unperturbed operator ($-\gamma S_z^2$)—in contrast to the oscillator case of [15]. Instead we have dealt with a global quantity, the tunnelling probability, which involves a summation over several (intermediate) Floquet states. This is the most relevant measurable quantity, except in situations where only a slight deviation from the static case occurs.

This section was bound to be phenomenological because we were not able to treat the spin's behaviour analytically once it had entered the chaotic regime. The main result here is the numerical verification that the spin continues to be blocked in this region, reflecting a marked difference from the particle case, where acceleration of tunnelling by several orders of magnitude has been shown to occur [13–15].

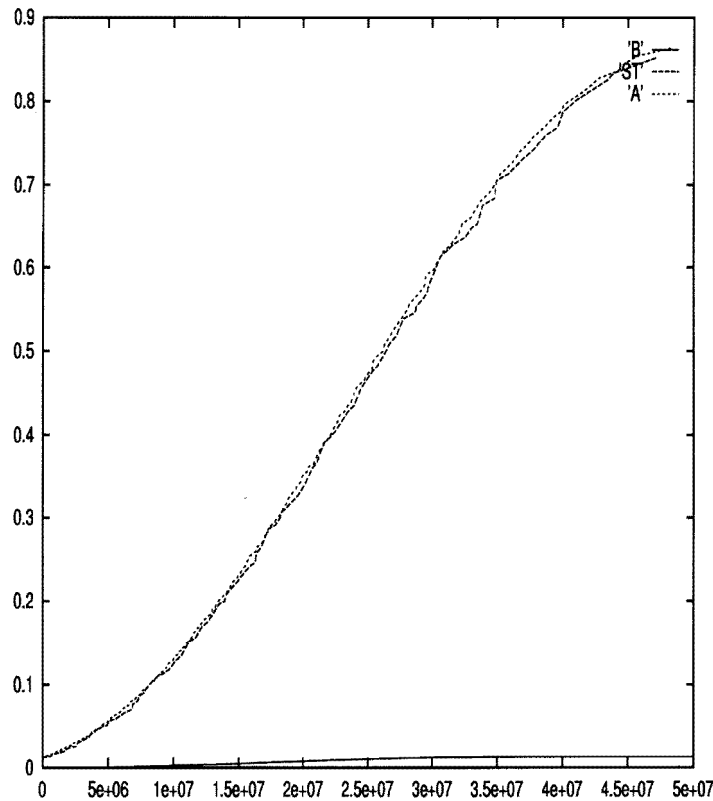


Figure 6. Transition probability for the quantum-mechanical Hamiltonian \hat{H}_1 as a function of time for the cases ST (static), A (integrable; cf figure 2) and B (chaotic; cf figure 4). There is good agreement between ST and A, and the hampering (blocking) of the spin in case B.

4. Conclusion

For $\omega \gg \omega_0$ and $\omega \gg \gamma$, the Hamiltonian \hat{H}_3 has *all* Floquet states localized either on the right or on the left. If (29) applies, the localization only holds for the ‘lowest’ states. For \hat{H}_2 as given by [2] there is always tunnelling. In the adiabatic regime, the rate is slightly lower than ω_0 . For \hat{H}_1 with $\omega \gg \omega_0$ and $\omega \gg \alpha S$, the spin is hampered. In the adiabatic regime, the *lowest* states are even localized.

The three mechanisms of localization of a quantum spin which we have analysed in the present paper, namely crossing of Floquet eigenvalues, averaging, and spatial symmetry breakdown, are omnipresent. So they seem to be of considerable conceptual importance, even more so since spin behaviour is markedly different from that of particles and a few fascinating open problems remain to be solved. For instance, do theoretical superstructures exist[†] behind Floquet-level crossing and averaging? Is there a precise semiclassical theory

[†] Grossmann and Hänggi [22] found a case where the two seem to coincide. Their spin has quantum number $S = \frac{1}{2}$, the opposite of what we have considered here, namely $S \gg \frac{1}{2}$. The evidence is purely numerical, provided by figure 1 of their paper, and stems from equation (27) of [31]. Equation (27) is an approximate identity which is valid for the high frequency case but whose derivation is missing. For spin $\frac{1}{2}$ we have $S_z^2/\hbar = \mathbb{I}$ the unit matrix, and thus $H_1 = -\gamma\hbar^2\mathbb{I} - \delta\cos(\omega t)S_z - \alpha S_x$. We now rotate through $\pi/2$ about the y -axis so as to interchange the x - and the z -axis and are left with Shirley’s Hamiltonian. The rest is plain averaging.

of blocking a spin in the chaotic regime? Can one extend the averaging method of [9] to larger domains? We are looking forward to the first experiments probing time-dependent spin quantum tunnelling in an alternating magnetic field. It may be that we will all have to face some puzzling surprises.

Acknowledgments

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Appendix A. Numerics

The Floquet eigenvalues ε_n and eigenfunctions Ψ_n may be defined [32] in terms of the evolution operator $U(t)$ evaluated at $t = T = 2\pi/\omega$ and starting at $t = 0$,

$$U\Psi_n = e^{-i\varepsilon_n T}\Psi_n. \tag{40}$$

In our case the Hilbert space is C^{2S+1} , and $-S \leq n \leq S$ while n is an integer. We solve the Schrödinger equation up to $t = T$

$$i\frac{\partial\Psi}{\partial t} = \hat{H}\Psi \tag{41}$$

in terms of the $(2S + 1)$ orthonormal eigenfunctions $|m\rangle$ of S_z :

$$S_z|m\rangle = m|m\rangle \quad m = -S, \dots, S \tag{42}$$

through the fourth-order Runge–Kutta method. That is, we take $\psi(0) = |m\rangle$ and thus find the columns of the matrix $U(T)$. The eigenvectors and eigenvalues follow as usual. The estimated error at each iteration is $|h|^5$ with $|h|$ being the integration time step. For $\omega \approx 1$, we have divided each period into $N_T = 4 \times 10^6$ parts, and a rough estimate of the precision is thus $[2\pi(2S + 1)/(\omega N_T)]^5 \times N_T \approx 10^{-17}$. However, using violation of unitarity of $U(T)$ as an *ad hoc* criterion:

$$|1 - |e^{-i\varepsilon_n T}|| > \beta$$

we arrived at† $\beta < 10^{-6}$, a much lower precision than 10^{-17} . This is probably due to an inaccuracy in the diagonalization process.

For ω small, it is important to sample the evolution during one period. We divided one period into N intervals with starting points n_i and $1 \leq i \leq N$, and find (by integration, as above) the vectors $\Psi^{n_i} = U(n_i, 0)|m_0\rangle$ where $|m_0\rangle$ is a fixed tunnelling state (with $m_0 = 6$). Furthermore, we evaluated Ψ^{n_i} using the Floquet eigenvalues and eigenvectors so as to find $\Psi(n_i + T)$. In the figures, we have kept only the *maxima* of the transition probability using this procedure for $\omega = 0.5$.

Appendix B. Estimating the stochasticity parameter

Let us consider the classical model

$$\hat{H}_2^{\text{cl}} = -\frac{\gamma}{S}S_z^2 - \alpha S_x \cos \omega t. \tag{43}$$

Defining the canonical variables $q = S_z$, and $p = -\phi$, we may write

$$S_x = \sqrt{S^2 - S_z^2} \cos \phi = \sqrt{S^2 - q^2} \cos p.$$

† For \hat{H}_2 , with the parameters $\gamma = 9$, $\alpha = 1$, $\delta = 1.0$, and $\omega = 0.5$, we have taken $N_T = 4 \times 10^6$.

We split $\hat{H}_2^{\text{cl}} = \hat{H}_0 + V(t)$ so that

$$\hat{H}_0 = -\frac{\gamma}{S} S_z^2$$

and

$$V(t) = -\alpha \sqrt{S^2 - S_z^2} \cos \phi \cos \omega t.$$

The condition for a resonance of order m is

$$m \frac{\partial \hat{H}_0}{\partial q} = \omega. \tag{44}$$

Equation (44) defines $q = q_m$, the angular momentum at the resonance, and $\omega = \Omega_m$, the corresponding frequency. We now transform to variables centred about the resonance,

$$J = q - q_m \tag{45}$$

$$\tilde{\phi} = \phi - \Omega_m t. \tag{46}$$

The generating function F_2 performing this canonical transformation is

$$F_2 = (J + q_m)(\phi - \omega t) = F_2(\phi, J)$$

with $\partial F_2 / \partial \phi = q$ and $\partial F_2 / \partial J = \tilde{\phi}$. The new Hamiltonian is

$$K(J, \tilde{\phi}, t) \approx \text{constant} + \frac{J^2}{2} \left(\frac{\partial^2 \hat{H}_0}{\partial q^2} \Big|_{q=q_m} \right) + \alpha \sqrt{S^2 - q_m^2} \left(\frac{e^{i(\Omega_m t + \tilde{\phi})} + e^{-i(\Omega_m t + \tilde{\phi})}}{2} \right). \tag{47}$$

In (47) we write $\cos(\omega t) = \frac{1}{2}(e^{i\omega t} + e^{-i\omega t})$ and keep only the resonant terms. We then find that only $m = \pm 1$ survives in (47). The upshot is, up to an irrelevant constant,

$$K = -\frac{\gamma J^2}{S} + \frac{\alpha S}{2} \cos \tilde{\phi} \sqrt{1 - \left(\frac{\omega}{2\gamma}\right)^2}. \tag{48}$$

Comparing (48) with the pendulum Hamiltonian $\frac{I^2}{2m} - mg \cos \tilde{\phi}$, we see that the width of an island in angular momentum space is

$$\Delta q = 4 \left(\frac{KS}{2\gamma} \right)^{\frac{1}{2}}$$

with

$$K = \frac{\alpha S}{2} \sqrt{1 - \left(\frac{\omega}{2\gamma}\right)^2}. \tag{49}$$

The spacing Δ' between two neighbouring resonances is

$$\Delta' = \frac{S\omega}{\gamma} = q|_{m=1} - q|_{m=-1} = \frac{\omega S}{2\gamma} - \left(-\frac{\omega S}{2\gamma}\right). \tag{50}$$

The Chirikov criterion [33] asserts that when $\Delta' = \Delta q$, the particle is able to wander about all of phase space, and global chaos sets in. In our case this gives, by (48) and (49),

$$\frac{\omega}{\gamma} = 2 \sqrt{\frac{\alpha}{\gamma}} \left[1 - \left(\frac{\omega}{2\gamma}\right)^2 \right]^{\frac{1}{4}} \approx 2 \sqrt{\frac{\alpha}{\gamma}}. \tag{51}$$

Hence, when the *stochasticity parameter*

$$s_p = 2 \sqrt{\frac{\alpha\gamma}{\omega}} \tag{52}$$

is of order one, there is a transition to global chaos (stochasticity). The factor of 2 in (52) is far from optimal. Equation (52) provides only an order of magnitude.

Appendix C. Classical limit

In the classical limit $\hbar \rightarrow 0$, $S \rightarrow \infty$ in conjunction with $\hbar S = 1\sigma$, say 1, the tunnelling condition becomes

$$\alpha \ll \hbar\gamma S = \gamma \quad (53)$$

since now

$$\hbar S = 1. \quad (54)$$

We rewrite \tilde{H}_1 as given by (14) in the correct-dimensional form,

$$\tilde{H}_1 = -\gamma\hbar^2 S_z^2 - \alpha\hbar S_x - \lambda\hbar S_z \quad (55)$$

where, now, S_x and S_z are dimensionless. Because of (54), \tilde{H}_1 may be written

$$\tilde{H}_1 = -\gamma \frac{S_z^2}{S^2} - \alpha \frac{S_x}{S} - \lambda \frac{S_z}{S}. \quad (56)$$

Let $\Omega_0(\lambda)$ denote the ground state of \tilde{H}_1 . Define the order parameter

$$m_s(\lambda) = \langle \Omega_0(\lambda) | \frac{S_z}{S} | \Omega_0(\lambda) \rangle. \quad (57)$$

If the ground state is a *tunnelling* state, it is (for $\alpha \rightarrow 0$ and $\lambda \rightarrow 0$) a linear combination of states $|+S\rangle$ and $|-S\rangle$ with *equal* coefficients (for $S \rightarrow \infty$). In the classical limit, it is thus natural to say that there is tunnelling, if

$$\lim_{\lambda \rightarrow 0} \lim_{S \rightarrow \infty} m_s(\lambda) = 0 \quad (58)$$

and no tunnelling (localization in one of the wells depending on whether $\lambda > 0$ or $\lambda < 0$), if

$$\lim_{\lambda \rightarrow 0} \lim_{S \rightarrow \infty} m_s(\lambda) \neq 0. \quad (59)$$

These notions make sense, with the same meaning as above, for finite quantum systems having S large. However, the double limit with *first* $S \rightarrow \infty$ and *then* $\lambda \rightarrow 0$ restores localization. That is to say, (58) holds, as we now prove.

Proposition. In the classical limit $\hbar \rightarrow 0$ and $S \rightarrow \infty$ with $\hbar S = \sigma$, say 1, localization holds under the condition $\lambda \ll \gamma\hbar S \equiv \Gamma$.

Proof. In the classical limit, the spins behave like classical rotors: $S_z = S \cos \theta$, $S_x = S \sin \theta \cos \phi$, $S_y = S \sin \theta \sin \phi$. Let us define

$$e_S^\pm(\theta, \phi) = \min_{\theta, \phi} g_S^\pm(\theta, \phi) \quad (60)$$

where

$$g_S^+(\theta, \phi) = -\gamma \left(\frac{S+1}{S} \right)^2 \cos^2 \theta - \alpha \left(\frac{S+1}{S} \right) \sin \theta \cos \phi - \lambda \left(\frac{S+1}{S} \right) \cos \theta \quad (61)$$

$$g_S^-(\theta, \phi) = -\gamma \cos^2 \theta - \alpha \sin \theta \cos \phi - \lambda \cos \theta. \quad (62)$$

Clearly, $g_S^-(\theta, \phi)$ is the classical rotor energy and g_S^+ is a slight modification of it, which arises naturally because of Lieb's interlacing inequalities [34]. Furthermore, (60) and (61) are the corresponding ground-state energies.

The following upper and lower bounds hold [34]:

$$\frac{e_S^-(\lambda - \varepsilon) - e_S^+(\lambda)}{\varepsilon} \leq \langle \Omega_0(\lambda) | \frac{S_z}{S} | \Omega_0(\lambda) \rangle \leq \frac{e_S^-(\lambda) - e_S^+(\lambda - \varepsilon)}{\varepsilon} \quad (63)$$

whatever $\varepsilon \geq 0$. Note that

$$g(\theta, \phi) = \lim_{S \rightarrow \infty} g_S^+(\theta, \phi) = \lim_{S \rightarrow \infty} g_S^-(\theta, \phi) \quad (64)$$

where

$$g(\theta, \phi) = -\gamma \cos^2 \theta - \alpha \sin(\theta) \cos(\phi) - \lambda \cos(\theta). \quad (65)$$

We also have

$$e(\lambda) = \lim_{S \rightarrow \infty} e_S^\pm = \min_{\theta, \phi} g(\theta, \phi). \quad (66)$$

Taking the limit $S \rightarrow \infty$ in (63) first and *next* the limit $\hbar \downarrow 0$, we obtain

$$\frac{d^-e(\lambda)}{d\lambda} \leq \lim_{S \rightarrow \infty} \langle \Omega_0(\lambda) | \frac{S_z}{S} | \Omega_0(\lambda) \rangle \leq \frac{d^+e(\lambda)}{d\lambda} \quad (67)$$

where $d^\pm e(\lambda)/d\lambda$ denote the right (resp. left) derivatives of $e(\lambda)$. If $\lambda \neq 0$, $g(\theta, \phi)$ has a *unique* minimum at $(\theta_0(\lambda), \phi_0(\lambda))$ and

$$\lim_{S \rightarrow \infty} m_S(\lambda) = \cos \theta_0(\lambda). \quad (68)$$

At $\lambda = 0$ and $\gamma = \alpha/2$ a simple bifurcation occurs with 'exchange of stability'. For $\alpha < 2\gamma$ and $\lambda = 0$, $g(\theta, \phi)$ possesses two minima,

$$\theta_0^- = \pi - \arcsin \frac{\alpha}{2\gamma} \quad \phi_0^- = 0 \quad (69)$$

and

$$\theta_0^+ = \arcsin \frac{\alpha}{2\gamma} \quad \phi_0^+ = 0. \quad (70)$$

Because of (68)–(70) we thus find

$$\lim_{\lambda \rightarrow 0^-} \lim_{S \rightarrow \infty} m_S(\lambda) = -\cos(\theta_0^-) = \sqrt{1 - \left(\frac{\alpha}{2\gamma}\right)^2} \quad (71)$$

whereas

$$\lim_{\lambda \rightarrow 0^+} \lim_{S \rightarrow \infty} m_S(\lambda) = -\cos(\theta_0^+) = -\sqrt{1 - \left(\frac{\alpha}{2\gamma}\right)^2}. \quad (72)$$

Therefore the ground state is localized in the classical limit and quantum coherence is broken, provided $\alpha < 2\gamma$.

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