Macroscopic magnetization tunneling and coherence: Calculation of tunneling-rate prefactors

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The tunneling of the total magnetization of a small ($\sim$100-Å-diameter) magnetic particle out of metastable easy directions or between degenerate easy directions is studied. The previously known Wentzel-Kramers-Brillouin exponents for the tunneling rates for these processes are supplemented by calculations of the prefactors, for various forms of the magnetocrystalline anisotropy. The calculations are done using spin-coherent-state path integrals. The formalism for evaluating fluctuation determinants for such path integrals is developed.

I. INTRODUCTION

A. Statement of the problem

Consider a spherical ferromagnetic particle of radius $\sim$50 Å. Such a small particle is typically a single domain, and at low temperatures (mK) the individual moments can be expected to rotate in unison in response to externally applied fields. The magnitude M_0 of the total magnetization is then fixed, and its orientation $\hat{\mathbf{M}}$ is the sole dynamical variable. The magnetocrystalline anisotropy creates easy and hard directions for $\hat{\mathbf{M}}$, and it is interesting to ask if one can see tunneling out of metastable easy directions in the presence of external fields, or resonance between equivalent easy directions. The observation of these phenomena would provide new instances of macroscopic quantum tunneling (MQT), or coherence (MQC), respectively, in the sense that one would then have evidence for the superposition of states that differed in the behavior of a macroscopic number of particles—10^5-10^6 magnetic moments in this case.[1-3] There is no evidence to date for MQC, and the only one for MQT is from experiments on current biased Josephson junctions.[4,5]

The possibility of macroscopic quantum phenomena in ferromagnetic particles has been pointed out by Chudnovsky and Gunther,[6] who showed, somewhat surprisingly, that with typical material parameters, the rate for MQT in ideally isolated particles could be made as high as 10^6-10^8 sec^{-1}. Since dissipation often reduces tunneling rates substantially, we investigated the effect of the magnetoelastic coupling between the magnetization and elastic waves.[7,8] We found, also surprisingly, that this is an extremely weak effect; the Wentzel-Kramers-Brillouin (WKB) exponent is increased by a relative amount $10^{-4}-10^{-6}$ in magnitude. This makes the experimental search for MQT in magnetic particles very interesting.

Chudnovsky and Gunther[6] calculated the exponential factors in the WKB rates for a few examples of MQC and MQT, choosing in each case the simplest possible form of the magnetic anisotropy energy. Subsequently, we calculated these factors for MQT for all the major crystal symmetries.[9]

In this paper, we complete the above calculations by finding the prefactors in the WKB tunneling rates. To be more specific, we note that quite generally, the tunneling rate, Γ for MQT or the splitting for MQC is given by an expression of the type

$$\Gamma = p_0 \omega_p (S_{\rm cl}/2\pi)^{1/2} e^{-S_{\rm cl}} , \tag{1.1}$$

where ω_p is the small-angle precession or oscillation frequency in the well, and $S_{\rm cl}$ is the WKB exponent. The notation comes from the fact that in standard instanton methods, $S_{\rm cl}$ is the classical action for the instanton (in units of $\hbar$). Usually, $S_{\rm cl} = O(E_b/\hbar\omega_p)$, where E_b is the barrier height. The dimensionless prefactor p_0 can often be of order 10 or so, and is therefore relevant to experiments. Our calculations may also be of some wider interest, since they involve spin-coherent-state path integrals in such a way that they cannot be obviously reduced to path integrals for motion in one dimension. The calculation of prefactors for one-dimensional problems is quite old,[10-13] but we are unaware of similar calculations for spin-coherent-state path integrals.

The plan of our paper is as follows. We describe the general setup for MQT and MQC below. In Sec. II, we formulate the problem of calculating an element of the density matrix for a single, large spin degree of freedom in general terms, without assuming a specific form of the anisotropy energy. We derive general formulas for the prefactor and the exponent in the WKB tunneling rate. Our approach follows that of Refs. 11–13 quite closely. In Sec. III we apply these formulas to MQT for biaxial, tetragonal and cubic crystal symmetries, and in Sec. IV, to two examples of MQC.

B. General setup for MQT and MQC

The general configuration for MQT that we shall consider consists of a spherical particle with its magnetization pointing along an easy axis, which we denote $\hat{\mathbf{z}}$, and an external field $\mathbf{H}$ applied opposite to $\hat{\mathbf{z}}$. The crystal symmetry of the magnetic material determines the form of the magnetocrystalline anisotropy energy density $E_a(\hat{\mathbf{M}})$, and this in turn determines the easy axis.[14] Since $E_a(\hat{\mathbf{M}}) = E_a(-\hat{\mathbf{M}})$ for general $\hat{\mathbf{M}}$ by time-reversal symmetry, there are at least two equivalent easy axes, $\hat{\mathbf{z}}$ and $-\hat{\mathbf{z}}$ when $\mathbf{H}=0$. We denote the magnitude of the field at which $\hat{\mathbf{z}}$ is rendered *classically* unstable by H_c. The possibility now arises that for $H < H_c$, the particle will tunnel out of the metastable direction $\hat{\mathbf{z}}$. All our calculations will be done to leading order in $\epsilon \equiv (1-H/H_c)$, since it turns out that to get an appreciable rate ϵ must be of order 0.01–0.001. This corresponds to an escape angle of a few degrees.

The general configuration for MQC consists of a similar particle, with the applied field being either zero, or along a special direction that makes equal angles with two or more zero-field easy directions. As long as the applied field is not too large, the total-energy *density*,

$$E(\theta,\phi)=E_a(\hat{\mathbf{M}})-\mathbf{M}\cdot\mathbf{H} \ , \tag{1.2}$$

has at least two degenerate minima, and the object is to study resonance between them.

II. INSTANTON CALCULATIONS FOR SPIN-COHERENT-STATE PATH INTEGRALS

In order to treat the magnetization quantum mechanically, we regard the total magnetic moment of the particle as equivalent to a large spin of magnitude J, given by

$$J=M_0 v_0/\hbar\gamma \ , \tag{2.1}$$

where $M_0=|\mathbf{M}|$, v_0 is the volume of the particle, and $\gamma=g\mu_B/\hbar$. We shall take the Hamiltonian for this spin to be such that

$$\langle \hat{\mathbf{n}}|H|\hat{\mathbf{n}}\rangle = v_0 E(\theta,\phi) \ , \tag{2.2}$$

where $|\hat{\mathbf{n}}\rangle$ is a spin-coherent state along a general direction $\hat{\mathbf{n}}$ with polar coordinates θ,ϕ, and $E(\theta,\phi)$ is the total-energy density (1.2). This choice guarantees the correct semiclassical dynamics for the spin.

In order to calculate the tunneling rate for either MQT or MQC, we consider matrix elements of the following type:

$$\langle \hat{\mathbf{n}}_1|e^{-HT}|\hat{\mathbf{n}}_2\rangle \ , \tag{2.3}$$

where $\hat{\mathbf{n}}_1=\hat{\mathbf{n}}_2$ is the metastable direction in the case of MQT, and $\hat{\mathbf{n}}_1$ and $\hat{\mathbf{n}}_2$ are two of the energetically degenerate—possibly the same—directions in the case of MQC. The calculations for MQT and MQC are very similar, so we will discuss only the former explicitly. In the limit that $T\to\infty$, we expect that

$$\langle \hat{\mathbf{n}}_1|e^{-HT}|\hat{\mathbf{n}}_1\rangle \to |\langle \hat{\mathbf{n}}_1|\psi_0\rangle|^2 e^{-E_0 T} \ , \tag{2.4}$$

where $|\psi_0\rangle$ is the wave function for the particle to have $\hat{\mathbf{M}}\simeq M_0\hat{\mathbf{n}}_1$, and E_0 is the corresponding energy. Since the state $|\psi_0\rangle$ is unstable, E_0 will have an imaginary part, which is related to the decay rate Γ by the usual formula,

$$\Gamma=-2\,\mathrm{Im}E_0 \ . \tag{2.5}$$

We can therefore obtain Γ if we can calculate the matrix element (2.3) in the limit $T\to\infty$. The tunneling matrix element for MQC can be found by a similar procedure.

A matrix element such as (2.3) is given by the spin-coherent-state path integral,

$$\mathcal{N}\int [d\hat{\mathbf{n}}]\exp\{-S[\hat{\mathbf{n}}(\tau)]\} \ , \tag{2.6}$$

where $\mathcal{N}$ is a normalization factor, and $S[\hat{\mathbf{n}}(\tau)]$ is the dimensionless Euclidean action

$$S[\hat{\mathbf{n}}(\tau)]=\int[-iJ\cos\theta\,\dot{\phi}+\hbar^{-1}v_0E(\theta,\phi)]d\tau . \tag{2.7}$$

The paths appearing in Eq. (2.6) are fixed at the end points $\tau=\pm T/2$ and we will change the notation slightly and write the general boundary conditions as $\hat{\mathbf{n}}(\pm T/2)=\hat{\mathbf{n}}_\pm$.

We now use standard instanton methods to evaluate Eq. (2.6). Such a calculation consists of two major steps. The first is to find the classical, or least-action path. This is usually relatively easy and gives the WKB exponent. The second step is to expand the action to second order in the fluctuations about the classical path, and evaluate the determinant (the so-called Van Vleck determinant) of the resulting quadratic form. This is much harder, and gives the less important prefactor. It is the second step that requires extra effort for calculating spin-coherent-state integrals, but we shall see that the first also has some interesting features.

To execute the first step, we must find the path (or paths) $\bar{\theta}(\tau),\bar{\phi}(\tau)$, that minimizes $S[\hat{\mathbf{n}}(\tau)]$, with boundary conditions $\bar{\theta}(\pm T/2)=\theta_\pm$ and $\bar{\phi}(\pm T/2)=\phi_\pm$. This path satisfies the equations of motion

$$\begin{aligned} i\hbar J\sin\bar{\theta}\,\dot{\bar{\theta}} &= v_0 E_\phi(\bar{\theta},\bar{\phi}) \ , \\ i\hbar J\sin\bar{\theta}\,\dot{\bar{\phi}} &= -v_0 E_\theta(\bar{\theta},\bar{\phi}) \ , \end{aligned} \tag{2.8}$$

where $E_\phi=\partial E/\partial\phi$ and $E_\theta=\partial E/\partial\theta$. An immediate problem is that these equations form a second-order system, but we must satisfy *four* boundary conditions, so that the problem is overdetermined, and a solution does not in general exist. The resolution of this difficulty was given by Klauder some time ago,[15,16] although the present authors only learned of it recently. It seems worthwhile to discuss this point, since none of the other papers on this subject (Refs. 6–9) have done so.[17]

Klauder showed that the action (2.7) could be replaced by S_η, given by

$$S_\eta[\hat{\mathbf{n}}(\tau)]=S[\hat{\mathbf{n}}(\tau)]+\frac{i}{4}\int d\tau\, J\eta(\dot{\theta}^2+\sin^2\theta\,\dot{\phi}^2) \ , \tag{2.9}$$

with the proviso that the limit $\eta\to 0$ be taken after doing the path integration. This has the effect of making the Euler-Lagrange equations a fourth-order system, allowing a solution to exist. The classical path develops boundary layers at $\tau=\pm T/2$ whose width is of order η. Outside the boundary layers the equation of motion is well given by Eq. (2.8). In the layer at $-T/2$, the path evolves rapidly from θ_-,ϕ_- to new values, which we denote by $\bar{\theta}_-,\bar{\phi}_-$. It then evolves by Eqs. (2.8) to values $\bar{\theta}_+,\bar{\phi}_+$ at $\tau=(T/2)-O(\eta)$. The final boundary layer connects these values to θ_+ and ϕ_+. Klauder showed that the contributions due to the term of order η in Eq. (2.9) could be dropped, and we could revert, in effect, to the original expression (2.6), provided we used the modified boundary conditions $\bar{\theta}(\pm T/2)=\bar{\theta}_\pm$ and $\bar{\phi}(\pm T/2)=\bar{\phi}_\pm$. The boundary layers provide two constraints among $\bar{\theta}_\pm,\bar{\phi}_\pm$, ensuring solvability of Eq. (2.8).

We can exploit these considerations for MQT calculations in the following way. We choose $\hat{\mathbf{n}}_1=\hat{\mathbf{z}}$. As usual, $\hat{\mathbf{z}}$ is a coordinate singularity of the spherical polar coordinates and ϕ is undetermined there. Since the classical trajectory for the matrix element (2.3) now begins and ends at $\hat{\mathbf{z}}$, we can take $\bar{\theta}_\pm=0$, and let $\bar{\phi}_\pm$ take on whatever value Eqs. (2.8) dictate, leaving it up to the boundary layers to connect to arbitrarily specified $\phi_\pm$. In this way we ensure that our answer for $\langle\hat{\mathbf{z}}|e^{-HT}|\hat{\mathbf{z}}\rangle$ will be smoothly connected to $\langle\hat{\mathbf{z}}'|e^{-HT}|\hat{\mathbf{z}}''\rangle$, where $\hat{\mathbf{z}}'$ and $\hat{\mathbf{z}}''$ are directions infinitesimally close to $\hat{\mathbf{z}}$, for which $\phi_\pm$ is well specified.

The second major step is to evaluate the Van Vleck determinant for small fluctuations about the classical path. We write

$$\theta(\tau)=\bar{\theta}(\tau)+\theta_1(\tau),\quad \phi(\tau)=\bar{\phi}(\tau)+\phi_1(\tau)\ , \tag{2.10}$$

and evaluate the action to second order in θ_1 and ϕ_1. Writing $S[\hat{\mathbf{n}}(\tau)]=S_{\rm cl}+\delta^2 S$, we have

$$\delta^2 S=-iJ\int\frac{d}{d\tau}\left[\sin\bar{\theta}\,\theta_1\right]\phi_1\,d\tau+\frac{i}{2}J\int\cos\bar{\theta}\,\dot{\bar{\phi}}\theta_1^2\,d\tau$$
$$+\frac{v_0}{2}\int(E_{\theta\theta}\theta_1^2+2E_{\theta\phi}\theta_1\phi_1+E_{\phi\phi}\phi_1^2)d\tau\ . \tag{2.11}$$

We have already done an integration by parts and used the fact that θ_1 and ϕ_1 vanish at the boundaries. The partial derivatives $E_{\theta\theta}$, $E_{\theta\phi}$, etc., are evaluated at the classical path. We complete the square for ϕ_1, and effect the Gaussian integration over ϕ_1.[18] This integration produces a determinantal factor that can be viewed as modifying the normalization factor $\mathcal{N}$. We ignore this effect for the moment, and focus on the term needed to complete the square. This term, along with the θ_1^2 terms in Eq. (2.11) leads to the following remaining action for θ_1:

$$I(\theta_1)=\int(A\dot{\theta}_1^2+B\theta_1\dot{\theta}_1+\tilde{C}\theta_1^2)d\tau\ . \tag{2.12}$$

Here,

$$A=\hbar J^2\sin^2\bar{\theta}/2v_0E_{\phi\phi}\ ,$$
$$B=iJ(\sin\bar{\theta}\,E_{\theta\phi}-\cos\bar{\theta}\,E_\phi)/E_{\phi\phi}\ , \tag{2.13}$$
$$\tilde{C}=v_0[E_{\phi\phi}(E_{\theta\theta}-\cot\bar{\theta}\,E_\theta)-(E_{\theta\phi}-\cot\bar{\theta}\,E_\phi)^2]/2\hbar E_{\phi\phi}\ .$$

We now transform the B term by writing $\theta_1\dot{\theta}_1=\frac{1}{2}d(\theta_1^2)/d\tau$, and integrating by parts. This gives

$$I(\theta_1)=\int(A\dot{\theta}_1^2+C\theta_1^2)d\tau\ , \tag{2.14}$$

with

$$C=\tilde{C}-iJ\frac{d}{d\tau}[(\sin\bar{\theta}\,E_{\theta\phi}-\cos\bar{\theta}\,E_\phi)/2E_{\phi\phi}]\ . \tag{2.15}$$

We shall see from the final answer for Γ that it is not necessary to find B or C explicitly.

We next turn to the normalization factor for the remaining path integral over θ_1. To do this, we write (2.6) as the formal limit of a discrete-time multiple integral. In other words, we write

$$\mathcal{N}\int[d\hat{\mathbf{n}}]=\lim_{n\to\infty}\left[\frac{2J+1}{4\pi}\right]^n\int d\hat{\mathbf{n}}_1\,d\hat{\mathbf{n}}_2\cdots d\hat{\mathbf{n}}_n\ , \tag{2.16}$$

where $\hat{\mathbf{n}}_k=\hat{\mathbf{n}}(-T/2+k\eta)$, and $\eta=T/(n+1)$ is the width of the time slices. Further,

$$d\hat{\mathbf{n}}_k=\sin\theta_k\,d\theta_k\,d\phi_k\simeq\sin\bar{\theta}_k\,d\theta_{1,k}\,d\phi_{1,k}\ . \tag{2.17}$$

We do not need to write the discretized version of $\delta^2 S$ explicitly. In addition to generating contributions to the B and C terms in the discretized version of Eq. (2.12), the Gaussian integration over $\phi_{1,k}$ will yield a factor of

$$[2\pi/\eta v_0E_{\phi\phi}(\bar{\theta}_k,\bar{\phi}_k)]^{1/2}\ ,$$

and we can write

$$\langle\hat{\mathbf{z}}|e^{-HT}|\hat{\mathbf{z}}\rangle=\mathcal{N}'e^{-S_{\rm cl}}\int[d\theta_1]e^{-I[\theta_1(\tau)]}\ , \tag{2.18}$$

where

$$\mathcal{N}'=\lim_{n\to\infty}\prod_{k=1}^{n}\left[\frac{2J+1}{2}\right][2\pi\eta v_0E_{\phi\phi}(\bar{\theta}_k,\bar{\phi}_k)]^{-1/2}\sin\bar{\theta}_k\ , \tag{2.19}$$

and $I[\theta_1(\tau)]$ is given by Eq. (2.14).

We now cast the θ_1 path integral into the standard form for a one-dimensional potential problem. We first note that in the limit of large J,

$$\mathcal{N}'=\lim_{n\to\infty}\prod_{k=1}^{n}(A_k/\pi\eta)^{1/2}\ , \tag{2.20}$$

where $A_k\equiv A(\bar{\theta}_k,\bar{\phi}_k)$. [We also define $C_k\equiv C(\bar{\theta}_k,\bar{\phi}_k)$.] Next, we change to a new time variable s defined by

$$ds=d\tau/2A(\bar{\theta}(\tau),\bar{\phi}(\tau))\ . \tag{2.21}$$

Then, in terms of discretized variables, our matrix element becomes

$$e^{-S_{\rm cl}}\lim_{n\to\infty}\left[\prod_{k=1}^{n}\int\frac{d\theta_{1,k}}{\sqrt{2\pi\Delta_k}}\right]\exp-\sum_{k=1}^{n}\left[\frac{1}{2\Delta_k}(\theta_{1,k}-\theta_{1,k-1})^2+2\Delta_k A_kC_k\theta_{1,k}^2\right]\ , \tag{2.22}$$

where $\theta_{1,0}=0$, and Δ_k, the width of the kth time slice in s, is given by

$$\Delta_k=(s_k-s_{k-1})=\eta/2A_k\ . \tag{2.23}$$

Equation (2.23) is precisely the standard path integral for one-dimensional motion, with Feynman's measure. Introducing a normalization constant N so that the measure agrees with Callan and Coleman,[12,13]—see Eq. (2.29) below—and writing

$$V(s)=4A[\bar{\theta}(\tau),\bar{\phi}(\tau)]C[\bar{\theta}(\tau),\bar{\phi}(\tau)] , \tag{2.24}$$

we finally have

$$\langle z|e^{-HT}|\hat{\mathbf{z}}\rangle \simeq e^{-S_{\rm cl}} N \int [d\theta_1] \exp -\frac{1}{2}\int \left[\left[\frac{d\theta_1}{ds}\right]^2 + V(s)\theta_1^2 \right] ds . \tag{2.25}$$

We now follow Refs. 12 and 13 to find the determinant of the quadratic form in Eq. (2.25). This form is diagonalized by the eigenfunctions $u_n(s)$ defined by

$$-\frac{d^2u_n}{ds^2}+V(s)u_n=\lambda_n u_n ,$$
$$u_n(\pm T/2)=0 . \tag{2.26}$$

Since the u_n's form a complete orthonormal set,

$$\int u_n(s)u_m(s)ds=\delta_{nm} , \tag{2.27}$$

we expand θ_1 as

$$\theta_1(s)=\sum_n c_n u_n(s) . \tag{2.28}$$

The normalization constant N is chosen so that in terms of the expansion coefficients c_n the measure $[d\theta_1]$ is defined as

$$[d\theta_1]=\prod_n (2\pi)^{-1/2}dc_n . \tag{2.29}$$

As usual there exists a zero mode corresponding to a translation of the center of the instanton, and a negative eigenvalue in the MQT case, which leads to the imaginary part of E_0. The only novel point is the handling of the zero mode, which we now discuss. It is apparent that $\delta^2 S$ vanishes for $(\theta_1,\phi_1)=(\dot{\bar{\theta}},\dot{\bar{\phi}})$. To see that $I(\theta_1)$ vanishes for $\theta_1=\dot{\bar{\theta}}$, let us write $\delta^2 S$ using a generalized operator notation as

$$\delta^2 S=(\theta_1 \;\; \phi_1)\begin{bmatrix} L_{\theta\theta} & L_{\theta\phi} \\ L^{\dagger}_{\theta\phi} & L_{\phi\phi}\end{bmatrix}\begin{bmatrix}\theta_1 \\ \phi_1\end{bmatrix} . \tag{2.30}$$

If we complete and remove the ϕ_1 square, we get $I(\theta_1)$ in the same notation:

$$I(\theta_1)=\theta_1(L_{\theta\theta}-L_{\theta\phi}L^{-1}_{\phi\phi}L^{\dagger}_{\theta\phi})\theta_1 . \tag{2.31}$$

Since $\delta^2 S(\dot{\bar{\theta}},\dot{\bar{\phi}})=0$, we have

$$L_{\theta\theta}\dot{\bar{\theta}}+L_{\theta\phi}\dot{\bar{\phi}}=0 ,$$
$$L^{\dagger}_{\theta\phi}\dot{\bar{\theta}}+L_{\phi\phi}\dot{\bar{\phi}}=0 . \tag{2.32}$$

Eliminating $\dot{\bar{\phi}}$ from these equations, we get

$$(L_{\theta\theta}-L_{\theta\phi}L^{-1}_{\phi\phi}L^{\dagger}_{\theta\phi})\dot{\bar{\theta}}=0 , \tag{2.33}$$

i.e., $I(\dot{\bar{\theta}})=0$.

Let us write the zero mode eigenfunction u_1 as

$$u_1(s)=S_n^{-1/2}\frac{d\bar{\theta}}{d\tau}(s) \tag{2.34}$$

where S_n is fixed by the normalization condition (2.27). We thus get

$$S_n=\int \left[\frac{d\bar{\theta}}{d\tau}\right]^2 ds=\int_{-T/2}^{T/2}\frac{1}{2A}\left[\frac{d\bar{\theta}}{d\tau}\right]^2 d\tau . \tag{2.35}$$

We shall see that it is not necessary to calculate S_n explicitly.

To perform the integration over c_1, we note that the change induced in θ_1 by a small change in the center of the instanton is

$$d\theta_1=\frac{d\bar{\theta}}{d\tau}d\tau . \tag{2.36}$$

Equating this to the change induced by a change in c_1,

$$d\theta_1=u_1(s)dc_1=S_n^{-1/2}\frac{d\bar{\theta}}{d\tau}dc_1 , \tag{2.37}$$

we get

$$(2\pi)^{-1/2}dc_1=(S_n/2\pi)^{1/2}d\tau . \tag{2.38}$$

So the factor from the zero mode to be included in the one instanton contribution to the matrix element (2.25) is $(S_n/2\pi)^{1/2}T$.

The calculation now follows Refs. 12 and 13 once again. All the arguments about summing multi-instanton configurations go through as before, and we get for the decay rate

$$\Gamma=-2\,{\rm Im}E_0$$
$$=k_s\left[\frac{S_n}{2\pi}\right]^{1/2} e^{-S_{\rm cl}}\left|\frac{\det'[-\partial_s^2+V(s)]}{\det(-\partial_s^2+\mu^2)}\right|^{-1/2} . \tag{2.39}$$

Here, k_s is the number of equivalent escape directions, i.e., the number of classical paths with the same action $S_{\rm cl}$, the prime on the det indicates that the zero eigenvalue is to be omitted, and μ^2 is given by

$$\mu^2=V(\pm\infty)=4AC|_{\tau=\pm\infty} . \tag{2.40}$$

The final point concerns the evaluation of the ratio of determinants. As explained in Ref. (13), this can be gotten from the asymptotic behavior of the zero mode. It follows from the differential equation (2.26) for $u_1(s)$ that we must have

$$u_1(s)\approx aS_n^{-1/2}e^{-\mu s}, \text{ as } s\rightarrow\infty , \tag{2.41}$$

where we have written the normalization factor $S_n^{-1/2}$ from Eq. (2.34) explicitly. In other words,

$$d\bar{\theta}/d\tau \approx a e^{-\mu s} \text{ as } s \to \infty \ . \tag{2.42}$$

Then

$$\frac{\det'[-\partial_s^2 + V(s)]}{\det(-\partial_s^2+\mu^2)} = -\frac{S_n}{2a^2\mu} \ . \tag{2.43}$$

(The minus sign reflects the one negative eigenvalue.) We thus get

$$\Gamma = k_s |a| (\mu/\pi)^{1/2} e^{-S_{\rm cl}} \ . \tag{2.44}$$

The quantities a and μ that appear in this formula can be obtained without calculating most of the intermediate quantities introduced above. All that is necessary is to differentiate the classical path to get $d\bar{\theta}/d\tau$, convert from τ to s, and read off a and μ by comparison with Eq. (2.42). It is not even necessary to integrate Eq. (2.21) to get the general relation between τ and s. Only the asymptotic relation is needed, and this is often very much easier to get.

In performing the Gaussian integral over ϕ_1, we have implicitly assumed that

$$E_{\phi\phi} > 0 \ . \tag{2.45}$$

This also ensures that $A > 0$, and that s is a monotonically increasing function of τ, which in turn makes the path integral in Eq. (2.25) well defined.

Condition (2.45) need not always be satisfied. If it is not, we can ask if it is possible to integrate out θ_1. Using the equations of motion [Eq. (2.8)], the condition on the positivity of the coefficient of θ_1^2 in Eq. (2.11) can be written as

$$E_{\theta\theta} - \cot\bar{\theta}\, E_\theta > 0 \ . \tag{2.46}$$

If this holds, we can repeat the above analysis and end up with a one-dimensional path integral over ϕ_1. The equivalent of Eq. (2.14), the reduced action for ϕ_1, is

$$I(\phi_1) = \int (A'\dot{\phi}_1^2 + C'\phi_1^2) d\tau \ , \tag{2.47}$$

with

$$\begin{aligned} A' &= \hbar J^2 \sin^2\bar{\theta}/2v_0(E_{\theta\theta} - \cot\bar{\theta}\, E_\theta) \ , \\ C' &= (v_0/2\hbar)[E_{\phi\phi} - E_{\theta\phi}^2/(E_{\theta\theta} - \cot\bar{\theta}\, E_\theta)] \\ &\quad + iJ\frac{d}{d\tau}[E_{\theta\phi}\sin\bar{\theta}/(E_{\theta\theta} - \cot\bar{\theta}\, E_\theta)] \ . \end{aligned} \tag{2.48}$$

The determinant of the operator in Eq. (2.47) is evaluated using techniques already described, and we will not discuss it any further.

For all the cases we have considered, one of the two conditions (2.45) and (2.46) always holds, and so we can always integrate out θ_1 or ϕ_1. Although this procedure is somewhat *ad hoc* and it is probably possible and rewarding to formulate the evaluation of the Van Vleck determinant in general terms, we shall not do so here. Instead we turn directly to applications of this formalism.

III. TUNNELING RATE FOR VARIOUS CRYSTAL SYMMETRIES

In this section we shall apply the formalism of the previous section to calculate the MQT rate for various crystal symmetries.[19] We have presented the values of the classical action for many of these in Ref. 9, and we shall adhere to our earlier notation as far as possible.

A. Biaxial symmetry

Let the easy axis be $\hat{\mathbf{z}}$, and the hard axis be $\hat{\mathbf{x}}$. In the presence of an external field $\mathbf{H}$ antiparallel to $\hat{\mathbf{z}}$, the energy $E(\theta,\phi)$ [Eq. (1.2)] is given in this case by

$$E(\theta,\phi) = (K_1 + K_2\sin^2\phi)\sin^2\theta + M_0 H\cos\theta \ . \tag{3.1}$$

where the anisotropy coefficients K_1 and K_2 are both positive. We assume that the higher anisotropy coefficients are negligible. The coercive field $H_c = 2K_1/M_0$, and in the limit of small $\epsilon = (1 - H/H_c)$, we get

$$E(\theta,\phi) = K_1\epsilon\theta^2 + K_2\theta^2\sin^2\phi - \frac{K_1}{4}\theta^4 + \cdots \ , \tag{3.2}$$

where we have subtracted a constant term.

There are two degenerate classical paths:

$$\begin{aligned} \bar{\theta}(\tau) &= 2\sqrt{\epsilon}\,\mathrm{sech}(\omega_p\tau) \ , \\ \bar{\phi}(\tau) &= -i\left[\frac{K_1\epsilon}{K_2}\right]^{1/2}\tanh(\omega_p\tau) + n\pi \ , \end{aligned} \tag{3.3}$$

where $n=0$ or 1, and $\omega_p = (2v_0/\hbar J)(K_1K_2\epsilon)^{1/2}$. The corresponding classical action is

$$S_{\rm cl} = (8/3)\epsilon^{3/2} J (K_1/K_2)^{1/2} \ . \tag{3.4}$$

It suffices to consider the path with $n=0$ in Eq. (3.2). As required by energy conservation,

$$\bar{\phi}^2 = -K_1(\epsilon - \bar{\theta}^2/4)/K_2 \ . \tag{3.5}$$

Note that $E_{\phi\phi} \simeq 2K_2\theta^2 > 0$, so we can integrate out ϕ_1. It is easy to show that

$$\begin{aligned} A(\tau) &= \hbar J^2/4v_0K_2 \ , \\ C(\tau) &= v_0K_1\epsilon[1 - 6\,\mathrm{sech}^2(\omega_p\tau)]/\hbar \ . \end{aligned} \tag{3.6}$$

Since $A(\tau)$ is a constant, s and τ are simply related by a change of scale. [We give the quantity $C(\tau)$ so that readers can verify that $V(\infty) = \mu^2$.] It is a simple matter to show that

$$\frac{d\bar{\theta}}{d\tau} \approx -4\epsilon^{1/2}\omega_p \exp[-J(K_1\epsilon/K_2)^{1/2}s] \ . \tag{3.7}$$

Thus, $|a| = 4\epsilon^{1/2}\omega_p$, and $\mu = J(K_1\epsilon/K_2)^{1/2}$. Substituting in the general formula (2.44), and using $k_s = 2$, Eq. (3.4) for $S_{\rm cl}$, and the equation just above it for ω_p, we get the result[20]

$$\Gamma = 2\sqrt{12}\,\omega_p (S_{\rm cl}/2\pi)^{1/2} e^{-S_{\rm cl}} . \tag{3.8}$$

B. Tetragonal symmetry (Ref. 19)

We once again take the easy axis to be $\hat{\mathbf{z}}$. The energy $E(\theta,\phi)$ is given by

$$E(\theta,\phi) = K_1 \sin^2\theta + [K_2 - K_2' \cos(4\phi)] \sin^4\theta + M_0 H \cos\theta . \tag{3.9}$$

Again, $K_1 > 0$, $H_c = 2K_1/M_0$, and in the small-ϵ limit, we get (up to a constant)

$$E(\theta,\phi) = \epsilon K_1 \theta^2 - \tilde{K}\theta^4 - K_2'\theta^4 \cos(4\phi) . \tag{3.10}$$

We have defined

$$\tilde{K} = (K_1 - 4K_2)/4 , \tag{3.11}$$

and we can always choose $K_2' > 0$. We also assume that $(K_2' + \tilde{K}) > 0$. The exit point [the point where $E(\theta,\phi) = 0$] is then found to be

$$\theta_0^2 = K_1 \epsilon / (K_2' + \tilde{K}) . \tag{3.12}$$

[If $(K_2' + \tilde{K}) < 0$, the exit angle is nonzero even when $\epsilon = 0$.] We also define

$$K_\alpha = [\tilde{K}^2 - (K_2')^2]^{1/2} , \tag{3.13}$$

and assume that this is real; if not, the correct answers can be obtained by analytic continuation.

The classical equations of motion (2.8) are tedious to integrate, and the final answer is

$$\begin{aligned} \bar{\theta}^2(\tau) &= K_1\epsilon / [\tilde{K} + K_2' \cosh(4\omega_p \tau)] , \\ \bar{\phi}(\tau) &= -i\omega_p \tau . \end{aligned} \tag{3.14}$$

where $\omega_p = \epsilon\gamma H_c$. (We have not bothered to state the symmetry-related values of ϕ.) The classical action is found to be

$$\begin{aligned} S_{\rm cl} &= \frac{iJ}{2} \int_{-\infty}^{\infty} \bar{\theta}^2(\tau) \dot{\bar{\phi}}\, d\tau = (J\omega_p/2) \int_{-\infty}^{\infty} \bar{\theta}^2(\tau)\, d\tau \\ &= J(K_1\epsilon/4K_\alpha) \ln[(\tilde{K} + K_\alpha)/K_2'] . \end{aligned} \tag{3.15}$$

As $\tau \to \infty$, we have

$$\dot{\bar{\theta}} \approx -2\omega_p (2K_1\epsilon/K_2')^{1/2} e^{-2\omega_p \tau} . \tag{3.16}$$

Once again $E_{\phi\phi} > 0$ and we can integrate out ϕ_1. We find that,

$$A(\tau) = (\hbar J^2/32 v_0)(\epsilon K_1 - \tilde{K}\bar{\theta}^2)^{-1} , \tag{3.17}$$

so that the asymptotic relation between s and τ involves the τ integral of $\bar{\theta}^2$. This can be read off from Eq. (3.15), and after some algebraic manipulations we get

$$2\omega_p \tau \approx \frac{J}{4} s + \frac{\tilde{K}}{2K_\alpha} \ln\left[\frac{\tilde{K} + K_\alpha}{K_2'}\right] . \tag{3.18}$$

We can now read off μ and a by combining Eqs. (3.16) and (3.18). Substituting the results in Eq. (2.45) and using $k_s = 4$, we finally get

$$\Gamma = 8\omega_p \left[\frac{JK_1\epsilon}{2\pi K_2'}\right]^{1/2} \left[\frac{K_2'}{\tilde{K} + K_\alpha}\right]^{\tilde{K}/2K_\alpha} e^{-S_{\rm cl}} . \tag{3.19}$$

It is not particularly illuminating to write out the prefactor p_0 [see Eq. (1.1)], although it can be seen that it is dimensionless and independent of v_0.

C. Cubic symmetry

The energy $E(\theta,\phi)$ is given in this case by

$$E(\theta,\phi) = K_1(\alpha_x^2\alpha_y^2 + \alpha_y^2\alpha_z^2 + \alpha_z^2\alpha_x^2) + M_0 H \cos\theta , \tag{3.20}$$

where α_x, α_y, and α_z are the direction cosines of $\mathbf{M}$. There are two cases to consider: $K_1 > 0$ in which case the easy axis is [100], and $K_1 < 0$ in which case the easy axis is [111].

If the easy axis is [100], $H_c = 2K_1/M_0$, and to leading order in ϵ, we have

$$E(\theta,\phi) = K_1\epsilon\theta^2 - (9/8)K_1\theta^4 - (1/8)K_1\theta^4 \cos(4\phi) . \tag{3.21}$$

This is of the same form as the tetragonal case Eq. (3.10) with $\tilde{K} = 9K_1/8$, $K_2' = K_1/8$, and $K_\alpha = 5^{1/2}K_1/2$. Substituting these values in Eqs. (3.15) and (3.19), we get

$$\begin{aligned} S_{\rm cl} &= J\epsilon 5^{-1/2} \ln(2 + \sqrt{5}) , \\ \Gamma &= 6.588\,\omega_p (S_{\rm cl}/2\pi)^{1/2} e^{-S_{\rm cl}} , \end{aligned} \tag{3.22}$$

and we have chosen to write the answer in the form (1.1).

If the easy axis is [111], $H_c = 4|K_1|/3M_0$, and to leading order in ϵ, we have

$$E(\theta,\phi) = \frac{2}{3}\epsilon|K_1|\theta^2 - \frac{\sqrt{2}}{3}|K_1|\theta^3 \cos(3\phi) . \tag{3.23}$$

The classical path is given by

$$\begin{aligned} \bar{\theta}(\tau) &= \sqrt{2\epsilon}\, \mathrm{sech}(3\omega_p \tau) , \\ \bar{\phi}(\tau) &= -i\omega_p \tau , \end{aligned} \tag{3.24}$$

where, now, $\omega_p = 2\epsilon|K_1|/3J$, and we have again omitted symmetry related paths. Once more, $E_{\phi\phi} > 0$, so ϕ_1 can be integrated out. The quantity $A(\tau) = \hbar J^2/12 v_0|K_1|\epsilon$, which is again a constant, so that the relation between s and τ is very simple. It is easy to do the rest of the calculation, and we finally get

$$\begin{aligned} \Gamma &= 18\sqrt{2}\,\omega_p (S_{\rm cl}/2\pi)^{1/2} e^{-S_{\rm cl}} , \\ S_{\rm cl} &= 2J\epsilon^2/3 . \end{aligned} \tag{3.25}$$

IV. MACROSCOPIC QUANTUM COHERENCE

In this section we shall apply our formalism to two examples of macroscopic quantum coherence. The simplest way to obtain degenerate easy directions is to not apply any external field. If the crystal has biaxial symmetry, the easy directions are 180° apart, and the tunnel splitting is small, since the angle through which the magnetization must tunnel is large. We can increase the splitting by applying a field at right angles to the easy axis, as this moves the easy directions toward one another. This problem is solved in Sec. IV A. A system where one may get large splittings without applying any field is one with easy-plane hexagonal symmetry. The rotational symmetry in the basal plane will be broken by a sixth-order anisotropy term that is likely to be small, and hence lead to large tunneling matrix elements between the six easy directions in the basal plane, i.e, large splittings.[21] We study this in Sec. IV B.

A. Uniaxial symmetry

We consider a uniaxial system with an easy axis $\hat{\mathbf{z}}$, and apply a field $\mathbf{H}$ along $\hat{\mathbf{x}}$. Ignoring anisotropy in the basal plane, the energy can then be written as

$$\begin{aligned} E(\theta,\phi) &= K\sin^2\theta - HM_0\sin\theta\cos\phi + H^2M_0^2/4K \\ &= K(\sin\theta-\sin\theta_0)^2 + 2K\sin\theta_0\sin\theta(1-\cos\phi)\,, \end{aligned} \tag{4.1}$$

where $\sin\theta_0 = HM_0/2K$, and it is assumed that $H < H_c = 2K/M_0$. The easy directions are $\hat{\mathbf{n}}_1$, $\hat{\mathbf{n}}_2$, with polar coordinates $\theta=\theta_0$, $\pi-\theta_0$, and $\phi=0$. We have chosen $E=0$ along these directions.

Energy conservation gives the following relation for the classical path:

$$\sin^2(\bar{\phi}/2) = -(\sin\bar{\theta}-\sin\theta_0)^2/4\sin\bar{\theta}\sin\theta_0\,, \tag{4.2}$$

and combining this with the equations of motion, the instanton that goes from θ_0 to $\pi-\theta_0$ is found to be

$$\begin{aligned} \cos\bar{\theta} &= -\cos\theta_0\tanh(\omega_b\tau)\,, \\ \sin\bar{\phi} &= \frac{i}{2}\frac{\cot^2\theta_0\,\mathrm{sech}^2(\omega_b\tau)}{[1+\cot^2\theta_0\,\mathrm{sech}^2(\omega_b\tau)]^{1/2}}\,. \end{aligned} \tag{4.3}$$

Here, $\omega_b = (\gamma H/2)\cot\theta_0$, which is half the small oscillation frequency in either well. Note that we have the happy circumstance that the boundary conditions are satisfied by both $\bar{\theta}$ and $\bar{\phi}$, so no boundary layers are needed. The action associated with this instanton is found to be

$$S_{\mathrm{cl}} = 2J\left[-\cos\theta_0 + \frac{1}{2}\ln\left[\frac{1+\cos\theta_0}{1-\cos\theta_0}\right]\right]. \tag{4.4}$$

To find the prefactor, we note that

$$E_{\phi\phi} = 2K\sin\theta_0\sin\bar{\theta}\cos\bar{\phi} = K(\sin^2\bar{\theta}+\sin^2\theta_0)\,, \tag{4.5}$$

which is positive, so we can integrate out ϕ_1 directly. From Eq. (4.5), we get

$$A = \frac{\hbar J^2}{2Kv_0}\frac{\sin^2\bar{\theta}}{\sin^2\bar{\theta}+\sin^2\theta_0}\,. \tag{4.6}$$

Substituting the solution (4.3) in Eq. (4.6), we get

$$\frac{ds}{d\tau} = \frac{Kv_0}{\hbar J^2}\left[2-\frac{1}{2}\left[\frac{1}{1-i\tan\theta_0\cosh(\omega_b\tau)}+\mathrm{c.c.}\right]\right], \tag{4.7}$$

where c.c. stands for complex conjugate. It follows that as $\tau\to\infty$,

$$s \approx \frac{Kv_0}{\hbar J^2}\left[2\tau - \frac{\cos\theta_0}{2\omega_b}\ln\left[\frac{1+\cos\theta_0}{1-\cos\theta_0}\right]\right]. \tag{4.8}$$

We also have from Eq. (4.3),

$$\bar{\theta}(\tau) \approx \pi - \theta_0 - 2\cot\theta_0 e^{-2\omega_b\tau}\,, \tag{4.9}$$

which gives

$$d\bar{\theta}/d\tau \approx 4\cot\theta_0\,\omega_b e^{-2\omega_b\tau}\,. \tag{4.10}$$

Combining Eqs. (2.43), (2.44), (4.8), and (4.10), we finally get for the tunnel splitting Δ,

$$\Delta = 8\left[\frac{J}{\pi}\right]^{1/2}\frac{\gamma K}{M_0}\frac{\cos^{5/2}\theta_0}{\sin\theta_0}\left[\frac{1-\cos\theta_0}{1+\cos\theta_0}\right]^{(1/2)\cos\theta_0} e^{-S_{\mathrm{cl}}}\,. \tag{4.11}$$

In the limit $H\to H_c$, we get $\theta_0 = \pi/2-(2\epsilon)^{1/2}$, $\omega_b = (\epsilon/2)^{1/2}\gamma H$, $S_{\mathrm{cl}} = (2J/3)(2\epsilon)^{3/2}$, and

$$\Delta = 8\sqrt{3}\omega_b(S_{\mathrm{cl}}/2\pi)^{1/2}e^{-S_{\mathrm{cl}}}\,. \tag{4.12}$$

This is the answer for a particle in a one-dimensional quartic double-well potential, with $2\omega_b$ being the small oscillation frequency in either well. In fact, we also have $S_{\mathrm{cl}} = 8V_0/3\omega_b$, where $V_0 = v_0K\epsilon^2$ is the barrier height.

B. Hexagonal symmetry

Our second example of MQC is a system with hexagonal symmetry, with six easy axes in the basal plane. The anisotropy energy for a hexagonal system can be written as

$$\begin{aligned} E(\theta,\phi) &= K_1\sin^2\theta + K_2\sin^4\theta \\ &\quad + K_3\sin^6\theta - K_3'\sin^6\theta\cos(6\phi)\,. \end{aligned} \tag{4.13}$$

We would like $\theta=\pi/2$ to be the easy axis. We ensure that by having $K_1<0$ and $K_2,K_3,K_3' \ll |K_1|$. We will assume that $|K_1|$ is sufficiently larger than the other coefficients that the fluctuations of θ about $\pi/2$ are small. Writing $\theta=\pi/2+\alpha$, and expanding to second order in α, we get

$$E(\alpha,\phi) = -K(1-3\alpha^2)\cos(6\phi) + K'\alpha^2\,, \tag{4.14}$$

where $K=K_3'$, $K'=-K_1-2K_2-3K_3$; $K'\gg K>0$.

Energy conservation gives

$$\alpha^2 = -K[1-\cos(6\phi)]/[K'+3K\cos(6\phi)]\,. \tag{4.15}$$

Combining this with the equation of motion

$$i\cos\alpha\frac{d\phi}{d\tau} = -(2\gamma/M_0)\alpha[K'+3K\cos(6\phi)]\,, \tag{4.16}$$

and expanding to lowest order in α, we get

$$\frac{d\phi}{d\tau}=\pm(2\gamma/M_0)\{[1-\cos(6\phi)]\times[KK'+3K^2\cos(6\phi)]\}^{1/2}\ . \tag{4.17}$$

It is straightforward to integrate this equation, and the instanton that goes from $\phi=0$ to $\phi=2\pi/6$ is found to satisfy

$$\cos(3\bar{\phi})=-z_1\tanh(\omega_p\tau)/[z_2^2-\tanh^2(\omega_p\tau)]^{1/2}\ , \tag{4.18}$$

where

$$\omega_p=(6\gamma/M_0)(2KK'+6K^2)^{1/2} \tag{4.19}$$

is the small oscillation frequency, and

$$\begin{aligned} z_1&=[(K'-3K)/6K]^{1/2}\ ,\\ z_2&=[(K'+3K)/6K]^{1/2}\ . \end{aligned} \tag{4.20}$$

Note that since $z_1^2=z_2^2-1$, $|\cos(3\phi)|<1$, and $\bar{\phi}(\tau)$ is real. The solution for $\bar{\alpha}(\tau)$, on the other hand,

$$\bar{\alpha}(\tau)=-i\,\mathrm{sech}(\omega_p\tau)/\sqrt{3}z_1\ , \tag{4.21}$$

is purely imaginary. The action for the instanton (4.18) is given by

$$\begin{aligned} S_{\rm cl}&=-iJ\int_{-\infty}^{\infty}d\tau\sin[\bar{\alpha}(\tau)]\frac{d\bar{\phi}}{d\tau}\\ &=(2K)^{1/2}J\int_0^{\pi/3}\frac{\sin(3\phi)}{[K'+3K\cos(6\phi)]^{1/2}}d\phi\\ &=\frac{2}{3\sqrt{3}}J\ln[(1+z_2)/z_1]\approx\frac{2}{3}J\left[\frac{2K}{K'}\right]^{1/2}. \end{aligned} \tag{4.22}$$

We now turn to the prefactor. Now,

$$E_{\phi\phi}=36K(1-3\alpha^2)\cos(6\bar{\phi})\ , \tag{4.23}$$

which is not always positive, and so we cannot integrate out ϕ_1. Instead, we can integrate out θ_1, since

$$\begin{aligned} E_{\theta\theta}-\cot\bar{\theta}E_\theta&=2(1+\alpha^2)[K'+3K\cos(6\bar{\phi})]\\ &\approx 2K'+O(K)>0\ . \end{aligned} \tag{4.24}$$

The time variables s [see Eq. (2.48)] and τ are related by a multiplicative constant, so the determinant for the ϕ_1 integral is particularly easy to evaluate. We finally get the following one-instanton contribution to the matrix element (2.3):

$$\langle\hat{\mathbf{n}}_2|e^{-HT}|\hat{\mathbf{n}}_1\rangle_{\text{one-instanton}}=|\langle\hat{\mathbf{n}}_1|\psi_0\rangle|^2\Delta T e^{-E_0T}\ , \tag{4.25}$$

with

$$\Delta=2\omega_p(S_{\rm cl}/2\pi)^{1/2}e^{-S_{\rm cl}}\ . \tag{4.26}$$

Here, $\hat{\mathbf{n}}_1$ and $\hat{\mathbf{n}}_2$ are the directions with $\theta=\pi/2$, and $\phi=0,2\pi/6$, respectively. As before, $|\psi_0\rangle$ is the wave function for the particle to have $\hat{\mathbf{M}}\simeq M_0\hat{\mathbf{n}}_1$, and E_0 is the corresponding energy.

We can now sum over multi-instanton configurations to obtain the effective tunneling Hamiltonian $H_{\rm eff}$. Ignoring the constant E_0, and denoting the states with $\phi=2\pi j/6$ by $|j\rangle$ (and defining $|6\rangle\equiv|0\rangle$), we get the cyclic matrix

$$\langle j|H_{\rm eff}|i\rangle=-\hbar\Delta(\delta_{i,j-1}+\delta_{i,j+1})\ . \tag{4.27}$$

The energies are $\pm 2\hbar\Delta$ and $\pm\hbar\Delta$, the latter two being doubly degenerate.

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[14]If the particle is not spherical, but is still small enough to have a uniform magnetization, we must add the shape anisotropy energy to the magnetocryalline anisotropy energy E_a, and

minimize the sum to find the easy axes, etc.

[15]J. R. Klauder, in *Path Integrals,* Proceedings of the NATO Advanced Summer Institute, edited by G. J. Papadopoulos and J. T. Devreese (Plenum, New York, 1978).

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[17]In fact, except for a (erroneous) footnote in Ref. 9, these papers only deal with the WKB exponent, which is just $-S_{\rm cl}$, where $S_{\rm cl}$ is the least action. One does not explicitly need the time dependence of the paths to calculate $S_{\rm cl}$ since the second term in Eq. (2.7) is a constant of motion, and the first term can be integrated by using this constant to write $\bar{\phi}$ as a function of $\bar{\theta}$. The existence of the classical path is nevertheless assumed.

[18]In order to do this, we must have $E_{\phi\phi}>0$. We will discuss this point below. See the discussion surrounding Eqs. (2.45)–(2.48).

[19]In addition to the cases presented here, we have also done the calculation for hexagonal symmetry with an easy direction along the c axis. The formulas are very complicated, so we do not give them here. It is worth noting, however, that just as the driving term for tetragonal symmetry is K_2', it is now K_3', the term that breaks the rotational symmetry in the basal plane. The exponent $S_{\rm cl}$ is given in Ref. 9.

[20]The prefactor for this case can also be obtained by noting that $M_x \sim \epsilon^{1/2}$, $M_y \sim \epsilon$, and $M_z \approx M_0 - M_x^2/2M_0$, so that if only leading-order terms are kept, the equations of motion for M_x and M_y are identical to those to the position x and momentum p for a particle moving in a one-dimensional quartic potential well. The prefactor for the latter problem is known. In footnote 9 of Ref. 9, we cited the answer using this approach, but we forgot the all important factor of 2 in Eq. (2.5), and so our result printed there is half the correct one.

[21]We thank C. L. Henley for suggesting this example to us.