Phase transition between quantum and classical regimes for the escape rate of a biaxial spin system

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Employing the method of mapping the spin problem onto a particle one, we have derived the particle Hamiltonian for a biaxial spin system with a transverse or longitudinal magnetic field. Using the Hamiltonian and introducing the parameter $p[\equiv(U_{\max}-E)/(U_{\max}-U_{\min})]$ where $U_{\max}(U_{\min})$ corresponds to the top (bottom) of the potential and E is the energy of the particle, we have studied the first- or second-order transition around the crossover temperature between thermal and quantum regimes for the escape rate, depending on the anisotropy constant and the external magnetic field. It is shown that the phase boundary separating the first- and second-order transition and its crossover temperature are greatly influenced by the transverse anisotropy constant as well as the transverse or longitudinal magnetic field. [S0163-1829(99)04117-X]

In recent years, quantum classical escape rate transition in the spin system has emerged as good candidates to display first- or second-order transition (FST).[1–4] Such a system is a single domain ferromagnetic particle with the magnetization **M** whose direction is subject to the magnetocrystalline anisotropy. In this situation the direction $\hat{M}$ has at least two equivalent stable orientations separated by an energy barrier U. Even though **M** is initially directed along one of these equivalent orientations, $\hat{M}$ can be changed by thermal activation whose rate is proportional to $\exp(-U/k_BT)$ at high temperature, and by quantum tunneling at a temperature low enough to neglect the thermal activation. In general, since the tunneling rate is dominated by $\exp(-U/\hbar\omega)$ where ω characterizes the width of the parabolic top of the barrier hindering the tunneling process, there is a crossover temperature T_0 from the thermally activated to the quantum tunneling process. Whether the transition about the crossover temperature is a first- or second-order one is determined by the external magnetic field and the magnetic anisotropy constant. By controlling the field and choosing the anisotropy constant in an appropriate way, we expect that there exists the crossover temperature $T_0^{(c)}$ at the phase boundary between first- and second-order transition.

Theoretical studies of the transition have been around for some time. Affleck[5] and Larkin and Ovchinnikov[6] demonstrated that a second-order transition from thermal to quantum regimes can occur at the crossover temperature by using the standard instanton technique. Later, Chudnovsky[7] discussed the criterion to determine FST based on the behavior of the period of oscillations in the inverted potential. Since then, based upon the mapping of a spin problem onto a particle one,[8] Chudnovsky, Garanin, and Martínes[1–3] suggested the spin system with the uniaxial crystal symmetry which shows FST in the presence of a transverse and longitudinal field. A biaxial spin model without an applied field has been considered by J.-Q. Liang *et al.*[4] who demonstrated that FST is determined from the ratio of the transverse to the longitudinal anisotropy constant by using the periodic instanton method. Even though they presented the analytical results without field, their approach could not be simply extended to the situation in the presence of field.[9] Very recently, in an effort to treat FST of the biaxial spin system with a longitudinal field, Garanin and Chudnovsky[12] used a perturbation approach and obtained the phase boundary between the first- and second-order transition which is numerically corrected by the Liang *et al.*'s exact value in the absence of the field. Thus a relevant approach to treat a biaxial spin system with a transverse or longitudinal field has been highly required for FST. In fact, complete analytical solution of the problem seems to be considerably important for the possibility of FST in molecular magnetic systems as well as a single domain ferromagnetic particle with many spins. In this paper, employing the method of mapping a spin problem onto a particle one, we obtain the particle Hamiltonian of the biaxial spin problem with a transverse or longitudinal field.[4,7] Actually, such a mapping is not a regular procedure and its form strongly depends on the form of the spin Hamiltonian. Using the Hamiltonian and introducing the dimensionless energy variable p, we will study the first- or second-order transition around the crossover temperature between thermal and quantum regimes for the escape rate, depending on the anisotropy constant and the external magnetic field, and present the analytic form of the phase boundary separating first- from second-order transition and the crossover temperature at the phase boundary.

Consider the biaxial spin model in a transverse field H_x described by the Hamiltonian

$$\mathcal{H}=-K_{||}S_z^2+K_{\perp}S_y^2-H_xS_x, \tag{1}$$

where $K_{||}$ and $K_{\perp}$ are the longitudinal and transverse anisotropy constants, respectively. This Hamiltonian can be mapped onto a particle problem[8] which describes the exact correspondence between the spin-wave function

$$\psi=\sum_{M=-S}^{S} a_M\left[\frac{(S+M)!(S-M)!}{(2S)!}\right]^{1/2}|SM\rangle, \tag{2}$$

where $|SM\rangle$ are the eigenstates of S_z, and the particle wave function $\Psi(x)=\exp[-\gamma(x)]\Sigma_{M=-S}^{S}a_M\exp(Mx)$ where

$$\frac{d\gamma(x)}{dx}=\frac{\tilde{S}h_x(1-k)\sinh(x)+(\tilde{S}-1)k\sinh(2x)}{1+k\cosh(2x)}, \quad (3)$$

$\tilde{S}=S+1/2$, $k=k_t/(2+k_t)$, $k_t=K_\perp/K_{||}$, and $h_x=H_x/(2\tilde{S}K_{||})$. The particle Hamiltonian is

$$\mathcal{H}=-\frac{1}{2m(x)}\frac{d^2}{dx^2}+U(x), \quad (4)$$

where $1/m(x)=K_{||}(2+k_t)[1+k\cosh(2x)]$, $U(x)=\tilde{S}^2K_{||}u(x)$, and

$$\begin{aligned}u(x)=\{&1/(1-k)[1+k\cosh(2x)]\}(h_x^2\sinh^2(x)(1-k)^2\\&-2h_x(1-k)\cosh(x)-k\{2h_x(1-k)\cosh(x)-1\\&+\cosh(2x)-[5\cosh(2x)-1]/(4\tilde{S}^2)\}\\&+k^2\{\cosh(2x)-1+[\cosh^2(2x)-\cosh(2x)\\&+4]/(4\tilde{S}^2)\}).\end{aligned} \quad (5)$$

Here we notice that the potential and the mass without transverse anisotropy are reduced to the results in Ref. 8. Even though $\tilde{S}$ appears in the potential, the terms associated with $\tilde{S}$ can be neglected in the large S limit including $S=10$. So, we will not include the terms with $1/(4\tilde{S}^2)$ in the subsequent consideration.[13]

In the quasiclassical approximation the transition rate becomes

$$\Gamma\sim\int dE W(E)\exp[-(E-U_{\min})/T], \quad (6)$$

where $W(E)$ is the probability of tunneling at an energy E. Since this is defined via the imaginary-time action $W(E)\sim e^{-S(E)}$, the transition rate is approximately given by

$$\Gamma\sim\exp(-F_{\min}/T), \quad (7)$$

where $F_{\min}$ is the minimum of the effective ''free energy'' $F\equiv E+TS(E)-U_{\min}$ with respect to E. Then, writing $F(E)/T=\int_{-1/2T}^{1/2T}d\tau[(m(x)/2)(dx/d\tau)^2+U(x)-E_{\min}]$, the imaginary time action is given by[1,2]

$$S(E)=2\int_{-x_1(E)}^{x_1(E)}dx\sqrt{2m(x)}\sqrt{U(x)-E}, \quad (8)$$

where $\pm x_1(E)$ are the turning points for the particle with energy $-E$ in the inverted potential $-U(x)$. We note that the mass is coordinate dependent, which is crucial in changing the boundary between first- and second-order transition, as will be seen. In order to determine FST in the crossover regime, we need to consider the behavior of $S(E)$ near the top of the barrier. Since the potential in Eq. (5) is even, we expand the integrand in Eq. (8) near $x=0$ which corresponds to the top of the barrier. Introducing dimensionless energy variable[1,3] $p[\equiv(U_{\max}-E)/(U_{\max}-U_{\min})]$ where $U_{\max}(U_{\min})$ corresponds to the top (bottom) of the potential, the action becomes near the top of the barrier:

$$S(E)=\frac{\pi\Delta U}{\sqrt{U_2(K_{||}+K_\perp)}}[p+\beta p^2+O(p^3)], \quad (9)$$

where $U_2=-\tilde{S}^2K_{||}u^{(2)}(0)/2$, $U_4=\tilde{S}^2K_{||}u^{(4)}(0)/4!$, and

$$\beta=\frac{3}{8}\left(\frac{U_4\Delta U}{U_2^2}\right)\left[1-\frac{2}{3}\left(\frac{k}{1+k}\right)\left(\frac{U_2}{U_4}\right)\right]. \quad (10)$$

Here $\Delta U=U_{\max}-U_{\min}$, and the second term in the bracket comes from the coordinate dependence of the mass. Using the analogy with the Landau theory of phase transitions and the general conditions for first- and second-order quantum classical transition of the escape rate discussed in Refs. 6 and 7, the factor in front of p^2 in Eq. (9) determines the boundary between the first- and second-order transition.

For the model without transverse field we have $U_2=2\tilde{S}^2K_{||}k/(1+k)$ and $U_4=-2\tilde{S}^2K_{||}k(1-5k)/[3(1+k)^2]$ from Eq. (5). If the mass does not depend on the coordinate, the sign of the factor of U_4 determines whether the system becomes the first- or the second-order transition due to the anharmonicity of the potential. However, as has been already noticed from Eq. (4), the mass is a function of x. So, we cannot simply obtain the phase boundary from the anharmonicity of the potential near the top of the barrier. Now, using $\Delta U=\tilde{S}^2K_{||}$, the action (9) is given by

$$S(E)=(\pi\tilde{S}/\sqrt{k_t})[p+\beta p^2+O(p^3)], \quad (11)$$

where $\beta=(1-1/k_t)/8$. Thus the critical value of k_t is 1 implying that smaller values of k_t lead to the first-order transition which is consistent with the result in Ref. 4.[14]

In case of the second-order transition the crossover occurs at temperature $T_0^{(2)}=\omega_0/(2\pi)$ where ω_0 is oscillation frequency near the bottom of the inverted potential. To estimate the frequency, we set up the Euclidean Euler-Lagrange equation $m(x)\ddot{x}+m'(x)\dot{x}^2/2-U'(x)=0$, and insert $x=x_b+\delta x$ into the equation, where x_b is x coordinate of the barrier. Expanding to second order in δx, we have $\delta\ddot{x}+\omega_0^2\delta x=0$, where $\omega_0=\sqrt{|U''(0)|/m(0)}$. This yields the crossover temperature given by $T_0^{(2)}=\tilde{S}\sqrt{K_{||}K_\perp}/\pi$, which leads to the crossover temperature at the phase boundary, $T_0^{(c)}=\tilde{S}K_{||}(2+k_t)/(3\pi)$.

For the first-order transition the approximate form of the crossover temperature for small k can be analytically calculated from the relation $T_0^{(1)}\simeq\Delta U/S(E_{\min})$. At the bottom of the barrier, $S(E_{\min})$ can be calculated directly from the integral expression (8) or following from[4]

$$S(E)=4\tilde{S}\sqrt{\frac{2k-\mathcal{E}}{1-k}}[\mathcal{K}(q)-(1-\alpha^2)\Pi(\alpha^2,q)], \quad (12)$$

which is derived from Eqs. (5) and (8). Here $\mathcal{E}=E/[\tilde{S}^2(K_{||}+K_\perp/2)]$, and $\mathcal{K}$ and Π are complete elliptic integrals of the first and third kind with $q^2=(z_1-1)/(z_1+1)$, $\alpha^2=q^2(1-k)/(1+k)$, and $z_1=[k(k-1)+\mathcal{E}]/[k(k-1)-k\mathcal{E}]$. This yields $T_0^{(1)}\simeq(\tilde{S}K_{||}/2)/\ln[(1+\sqrt{1-k^2})/k]$, which is approximate form for the first-order transition in the region of small k.

We now consider the model with a transverse magnetic field. Expanding the potential (5) in powers of x, we obtain

$$U_2=[\tilde{S}^2K_{||}/(1+k_t)](h_x+k_t)(1-h_x), \quad (13)$$

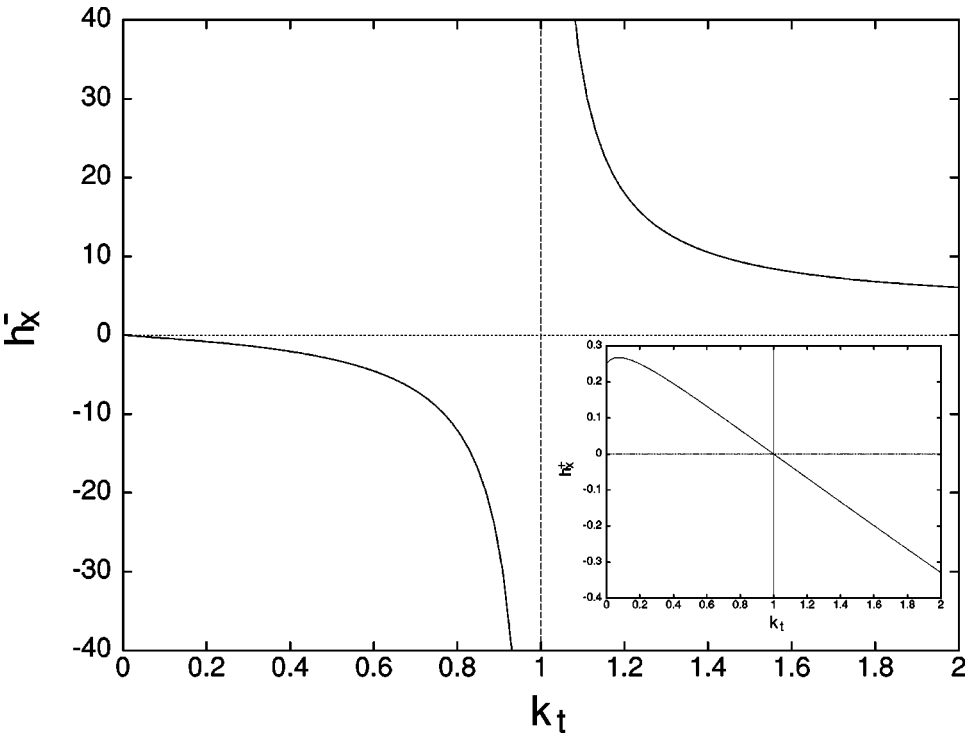

FIG. 1. h_x^- as a function of k_t. Inset: h_x^+ as a function of k_t.

$$U_4=\{\tilde{S}^2K_{||}/[12(1+k)^2]\}[4(1-5k)(1-k)h_x^2 + (-1+38k-57k^2)h_x-8k(1-5k)], \quad (14)$$

and $\Delta U=\tilde{S}^2K_{||}(1-h_x)^2$. It is noted that the condition in which the barrier does not vanish is $U_2>0$, i.e., $h_x<1$. Then, the action near the top of the barrier becomes, from Eq. (9),

$$S(E)=\pi\tilde{S}\frac{(1-h_x)^{3/2}}{(k_t+h_x)^{1/2}}[p+\beta p^2+O(p^3)], \quad (15)$$

where

$$\beta=\frac{(1-k_t)}{8(h_x+k_t)^2}(h_x-h_x^+)(h_x-h_x^-), \quad (16)$$

$$h_x^\pm(k_t)=\frac{1-14k_t+k_t^2\pm(1+k_t)\sqrt{1+34k_t+k_t^2}}{8(1-k_t)}. \quad (17)$$

As is shown in Fig. 1, the behavior of $h_x^\pm$ shows that β is negative for $k_t<1$ and $h_x<h_x^+$ which corresponds to the first-order transition. As is shown in Fig. 2, in the absence of the transverse anisotropy, $h_x=1/4$ is the critical value for FST,[1] and in the presence of very small transverse anisotropy we have $h_x^+\simeq(1+7k_t/2)/4$ in which the boundary becomes wider for $0\leqslant k_t<0.2$. As k_t continues to increase, the region where a first-order transition occurs, is smaller for the biaxial model than for the uniaxial model. This is intuitively understood that, since the transverse anisotropy drives the decay of the metastable state, it plays the role of the transverse field in the uniaxial case and so, for a given small transverse field the region for the first-order transition decreases as the transverse anisotropy increases. It is also noted that h_x^+ decreases linearly for $k_t\lesssim 1$, i.e., just as $h_x^+\simeq(1-k_t)/3$.

Continuing in the present case as without transverse case, we have the crossover temperature for the second-order transition

$$T_0^{(2)}(k_t,h_x)=(\tilde{S}K_{||}/\pi)\sqrt{(k_t+h_x)(1-h_x)}. \quad (18)$$

Using Eq. (17) for h_x^+, the crossover temperature at the phase boundary between the first- and second-order transition is written as

$$T_0^{(c)}=[\sqrt{3}/(2\pi)]\tilde{S}K_{||}(1+k_t)\sqrt{h_x^+(k_t)/(1-k_t)}, \quad (19)$$

which is illustrated in Fig. 3. We note that $T_0^{(c)}\simeq\tilde{S}K_{||}[\sqrt{3}/(4\pi)](1+3k_t)$ for small k_t and $T_0^{(c)}\simeq[\tilde{S}K_{||}/(2\pi)](1+k_t)$ for $k\lesssim 1$.

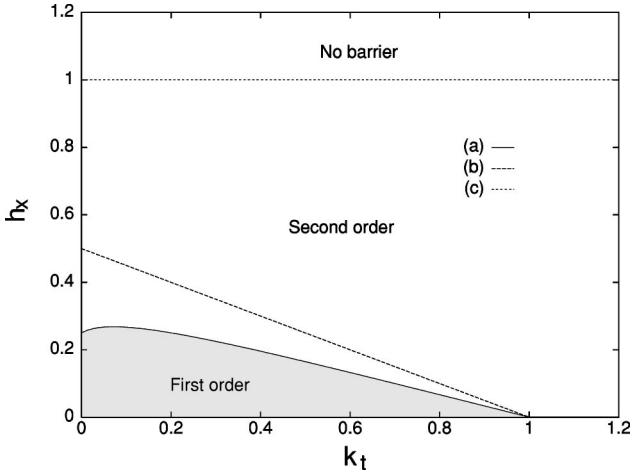

FIG. 2. Boundary between the first- and the second-order transitions, where (a) $h_x^+(k_t)$, (b) $h_x^{\max}(k_t)$ which gives the maximal crossover temperature in the quantum classical transition, and (c) the boundary whether the barrier vanishes or not.

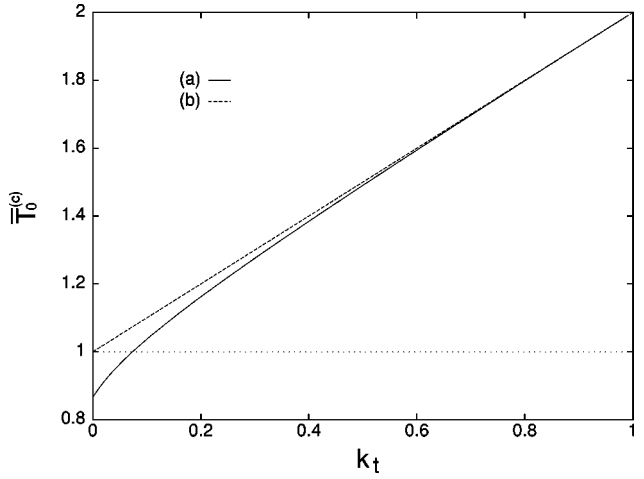

FIG. 3. Dependence of (a) the crossover temperature $T_0^{(c)}$ at the phase boundary and (b) the maximum crossover temperature $T_0^{\max}$ on the scaled anisotropy constant k_t where $\overline{T}_0^{(c)}=T_0^{(c)}/(\tilde{S}K_{||}/2\pi)$.

In the case of the first-order transition the crossover temperature as a function of k_t for small h_x is approximately estimated from the ground-state tunneling exponent given by Eq. (8) or the direct integral expression[15] and ΔU considered above, which gives

$$T_0^{(1)}(k_t,h_x)\simeq\tilde{S}K_{||}(1-h_x)^2/[2g(k_t,h_x)], \tag{20}$$

where

$$g(k_t,h_x)=\ln\left(\frac{\sqrt{1+k_t}+\sqrt{1-h_x^2}}{\sqrt{1+k_t}-\sqrt{1-h_x^2}}\right) - \frac{2h_x}{\sqrt{k_t}}\arctan\left(\sqrt{k_t/(1+k_t)}\sqrt{1/h_x^2-1}\right). \tag{21}$$

Simple analysis for the crossover temperature shows that there exists a maximum of the crossover temperature $T_0^{\max}$ in the regime of the second-order transition, which from Eq. (18) gives $T_0^{\max}=[\tilde{S}K_{||}/(2\pi)](1+k_t)$ at $h_x=(1-k_t)/2$. It is noted that this is the asymptotic form of $T_0^{(c)}$ for $k_t\lesssim 1$, as discussed previously. As the transverse anisotropy increases, $h_x^{\max}$ for the maximal crossover temperature decreases, (Fig. 2) while $T_0^{\max}$ increases linearly (Fig. 3). As is summarized in Table I and shown in Fig. 3, as k_t increases, the difference between $T_0^{\max}$ and T_0^c decreases and becomes zero at $k_t=1$ which is the critical value in the fieldless case.

In the presence of a longitudinal field of the spin model $\mathcal{H}=-K_{||}S_z^2+K_\perp S_y^2-H_zS_z$, we can proceed similarly, and we will briefly discuss the essential points. In order to obtain the relation between $h_z[=H_z/(2\tilde{S}K_{||})]$ and k_t at the phase boundary, we need to have the coefficient of p^2 in the action, i.e., β including U_3 and $m'(x_b)$ as well as U_2 and U_4 where x_b is the position of the barrier, because the potential given by

TABLE I. Summary of (a) the maximum crossover temperature and (b) the crossover temperature at the phase boundary at two end points in Fig. 3.

k_t	h_x	$T_0/(\tilde{S}K_{||}/2\pi)$
0	1/2	1 (a)
1	0	2 (a)
0	$h_x^+=1/4$	$\sqrt{3}/2$ (b)
1	$h_x^+=0$	2 (b)

$$u(x)=\frac{1}{(1-k)[1+k\cosh(2x)]}(h_z^2(1-k)^2 - k\{2h_z(1-k)\sinh(2x)-1+\cosh(2x) - [5\cosh(2x)-1]/(4\tilde{S}^2)\}+k^2[\cosh(2x)-1 + [\cosh^2(2x)-\cosh(2x)+4]/(4\tilde{S}^2)\}), \tag{22}$$

is not an even function. Following the procedure discussed previously, we have the boundary between first- and second-order transition

$$k_t=(1-h_z^2)/(1+2h_z^2). \tag{23}$$

The ratio of two anisotropy constants, k_t decreases parabolically for $h_z\ll 1$, as $k_t\simeq 1-3h_z^2$, and linearly for $h_z\lesssim 1$, as $k_t\simeq(2/3)(1-h_z)$. This can be understood from the fact that, since the height of barrier decreases as h_z increases, the first-order transition is expected for the larger width of the barrier which comes from the smaller value of the trasverse anisotropy. The crossover temperature $T_0^{(c)}$ at the phase boundary can be obtained by using Eq. (23) and $T_0^{(2)}(k_t,h_z)=(1/2\pi)\sqrt{|U''(x_b)|/m(x_b)}$ in the second-order transition, which leads to

$$T_0^{(c)}=(\tilde{S}K_{||}/\pi)(1-h_z^2)/\sqrt{1+2h_z^2}. \tag{24}$$

Simple analysis shows that our results are consistent with the ones deduced from the correction of the perturbative calculation performed in Ref. 12 up to the numerical factors. Even though the perturbative approach is less justified at large value of the transverse anisotropy constant, its boundary and crossover temperature are strikingly the same trend as our analytical results obtained from the quasiclassical method.

In this paper we have considered the quantum classical escape rate transition of a biaxial spin system in the presence of a transverse or longitudinal field by using the particle Hamiltonian mapped from the spin system. The coordinate dependence of the particle mass was crucial in changing the boundary between the first- and second-order transition and its boundary was greatly influenced by the transverse anisotropy constant and external field, whose effect is expected to be observed in future experiments including Fe_8 molecular magnet.[16]

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[9] Actually, based on the periodic instanton method, Lee *et al.* (Ref. 10) studied the phase transition in the biaxial spin system with a transverse magnetic-field. In order to obtain the effective Lagrangian, they expanded the quantity $s \sin\theta (=\sqrt{s^2-p^2})$ of the magnetic-field term of the Hamiltonian in p up to quadratic terms [the x-y approximation (Ref. 11)]. However, this approximation, often used for planar magnet, breaks down in the presence of a field perpendicular to the z axis in their Hamiltonian because such a field forces the spins to move out of the x-y plane. Correspondingly, their starting effective Lagrangian is valid for $\lambda \ll 1$. Thus, in our understanding, it is not appropriate to discuss the quantum classical transition depending on the whole magnitude of the parameter $\lambda = K_2/K_1$ because λ was initially very small in the Lagrangian.

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[13] We check this point numerically and confirm that, up to the small numerical values which can be neglected, there is no difference of $u(x)$ between with and without $1/(4\tilde{S}^2)$ terms in the integration range from $-x_1$ to x_1 in Eq. (8).

[14] Since the Hamiltonian $\mathcal{H} = K_1 S_z^2 + K_2 S_y^2$ is used in Ref. 4, our anisotropy constants are related by $K_{||} = K_2$ and $K_\perp = K_1 - K_2$. This yields $k_t = -1 + 1/\lambda$, in which $k_t = 1$ corresponds to $\lambda = 1/2$.

[15] We have also performed the calculation for the action in the whole range of E. In this case the formula is very complicated and we do not present it here. We checked that the formula in the bottom of the potential agrees with the result (20) obtained from the action at the ground-state tunneling.

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