

Periodic Instanton and Phase Transition in Quantum Tunneling of Spin Systems

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Abstract The quantum-classical transitions of the escape rates in a uniaxial spin model relevant to the molecular magnet $Mn_{12}Ac$ and a biaxial anisotropic ferromagnetic particle are investigated by applying the periodic instanton method. The effective free energies are expanded around the top of the potential barrier in analogy to Landau theory of phase transitions. We show that the first-order transitions occur below the critical external magnetic field $h_x = \frac{1}{4}$ for the uniaxial spin model and beyond the critical anisotropy constant ratio $\lambda = \frac{1}{2}$ for the biaxial ferromagnetic grains, which are in good agreement with earlier works.

PACS number(s): 75.45.+j, 75.50.Tt

Keywords: Periodic Instanton; Spin Tunneling; Phase Transition

Macroscopic quantum tunneling(MQT) of magnetization was intensively studied both theoretically and experimentally in the last decade [1]. In recent years much attention has been attracted to the phase transition problem in quantum tunneling of magnetization

[2-4]. The interest to this problem arises mainly from the successful experiments on the molecular magnet Mn_{12}Ac [5-7]. It has been shown that [2] the quantum-classical transition of the escape rate is analogous to the phase transition and the general conditions for the first- and second-order transitions are also analyzed. Quite recently an effective free energy $F = a\phi^2 + b\phi^4 + c\phi^6$ for the transitions of a spin system was introduced [3] as in Landau theory of phase transition. Here $a = 0$ corresponds to the quantum-classical transition and $b = 0$ to the boundary between first- and second-order transitions. In the common sense the first-order transitions are difficult to find in real systems and a uniaxial spin model is one of the very few examples which would exhibit the first-order transition. Various theoretical methods were used to deal with the spin tunneling problem [8-13]. We present in this letter the periodic instanton calculations for the quantum-classical transitions of the escape rates for the uniaxial spin model and a biaxial anisotropy ferromagnetic particle. The latter provides another candidate in which we may observe the first-order transition at the proper anisotropy constant.

A rather simple and experimentally important system which may exhibit the first-order transition is the uniaxial spin model in a magnetic field parallel to x -axis H_x described by the Hamiltonian

$$\mathcal{H} = -DS_z^2 - H_x S_x \quad (1)$$

which is generic for problems of spin tunneling. This model is believed to have relevance to the molecular magnet Mn_{12}Ac with D the anisotropy constant. It was found recently that there exists a critical value of the external field below which the first-order escape rate transition occurs [3]. Here the periodic instanton method is used to deal with this problem which should be more accurate as we need not resort to the double well potential approximation. Using the method of mapping the spin model onto a particle problem [3,9], the equivalent particle Hamiltonian is $\mathcal{H} = \frac{p^2}{2m} + U(x)$, where

$$U(x) = S(S+1)D(h_x \cosh x - 1)^2 \quad (2)$$

and $m = 1/2D$, $h_x = H_x/2\tilde{S}D$, $\tilde{S} = S + 1/2$. The minimum of the effective potential, $x_0 = \cosh^{-1}(1/h_x)$, has been moved to zero and $S \gg 1$ is used throughout. Recently the single kink and kink lattice solutions for a class of quasi-exactly solvable potential model in field theory, the Double sinh-Gordon(DSHG) potential [14], have been found and the statistical mechanics of DSHG kinks are studied. Our motivation is based on that the potential (2) is a special case of DSHG model and corresponding instanton and periodic instanton configurations may be found following the similar procedure.

The vacuum instanton solution is nothing but the zero-energy solution of the equation of motion in imaginary time τ

$$\frac{1}{2}m \left(\frac{dx}{d\tau} \right)^2 - U(x) = 0. \quad (3)$$

The instanton/anti-instanton located at τ_0 is given by

$$x_c(\tau) = \pm 2 \tanh^{-1} \left[\tanh \frac{x_0}{2} \tanh \left(\frac{\tau - \tau_0}{\xi} \right) \right], \quad \xi = \frac{1}{\sqrt{1 - h_x^2 \tilde{S}D}} \quad (4)$$

where $\tanh \frac{x_0}{2} = \sqrt{\frac{1-h_x}{1+h_x}}$ and the corresponding Euclidean action is

$$S_c = \int_{-\infty}^{+\infty} m \dot{x}_c^2 d\tau = 2\tilde{S} \left(\ln \frac{1 + \sqrt{1 - h_x^2}}{h_x} - \sqrt{1 - h_x^2} \right). \quad (5)$$

To understand the finite temperature tunneling behavior we construct the periodic instanton configuration satisfying the periodic boundary condition similar to the kink lattice method in double sinh-Gordon(DSHG) theory. This solution is obtained by integrating the equation of motion

$$\frac{1}{2}m \left(\frac{dx}{d\tau} \right)^2 - U(x) = -E \quad (6)$$

and is given by

$$x_p(\tau) = \pm 2 \tanh^{-1} \left[\tanh x_1 \operatorname{sn} \left(\frac{\tau - \tau_0}{\xi_P}, k \right) \right] \quad (7)$$

with

$$k = \sqrt{\frac{1 - (\sqrt{E'} + h_x)^2}{1 - (\sqrt{E'} - h_x)^2}} = \frac{\tanh x_1}{\tanh x_2}, \quad (8)$$

$$\xi_P = \frac{1}{\tilde{S}D\sqrt{1 - (h_x - \sqrt{E'})^2}} = \frac{k}{2h_x\tilde{S}D \sinh x_1 \cosh x_2}.$$

where $\text{sn}(\tau, k)$ is the Jacobi elliptic function with modulus k and the complementary modulus $k' = \sqrt{1 - k^2}$. Equivalently,

$$\tanh^2 x_{1,2} = \frac{1 - h_x \mp \sqrt{E'}}{1 + h_x \mp \sqrt{E'}} = \frac{1 - h_x (1 + k^2)h_x \mp k'^2(1 + h_x) + \sqrt{4h_x^2k^2 + k'^4}}{1 + h_x (1 + k^2)h_x \mp k'^2(1 - h_x) + \sqrt{4h_x^2k^2 + k'^4}} \quad (9)$$

and the characteristic length of the periodic instanton

$$\xi_P^2 = \frac{1}{\tilde{S}^2 D^2} \left(1 - h_x^2 \left(\frac{(1 + k^2)h_x^2 - k'^2(1 - h_x^2) + h_x\sqrt{4h_x^2k^2 + k'^4}}{(1 + k^2)h_x^2 + h_x\sqrt{4h_x^2k^2 + k'^4}} \right)^2 \right)^{-1}. \quad (10)$$

This description corresponds to the movement of a pseudo-particle in the inverted potential $-U(x)$ with energy $-E$ and $E' = E/\tilde{S}^2 D$. The periodicity of the solution (7) is $\tau_p(E) = 4\beta$, $\beta = K(k)\xi_P$, where $K(k)$ is the complete elliptic integral of the first kind. The topological charge of periodic instanton

$$Q_p = 2x_p(K(k)) = 2x_1 \quad (11)$$

is smaller than the vacuum instanton case

$$Q = \int_{-\infty}^{+\infty} \frac{\partial x_c(\tau)}{\partial \tau} dx = 2x_0. \quad (12)$$

Similarly, the periodic instanton size ξ_P is also smaller than the zero-energy one ξ . The potential and the instanton configurations are depicted in Fig. 1. The particle starts from the turning point $-x_1$ at imaginary time $-\beta$ and reaches the other turning points x_1 at β . After the same interval, at time 2β , the particle returns to its original position, i.e., it tunnels through the barrier twice in the whole period.

The Euclidean action of the periodic instanton configuration is

$$S_p = \int_{-\beta}^{\beta} [m \dot{x}_p^2 + E] d\tau = W + 2E\beta \quad (13)$$

where

$$W = \frac{2}{D\xi_P\alpha^2} [(\alpha^4 - k^2) \Pi(\alpha^2, k) + k^2 K(k) + \alpha^2 (K(k) - E(k))] \quad (14)$$

Here $\alpha^2 = \tanh^2 x_1 < k^2$ and $E(k)$, $\Pi(\alpha^2, k)$ are the complete elliptic integral of second and third kind, respectively.

The temperature dependent tunneling rate can be estimated by a Boltzmann average over the tunneling probabilities from excited states with energy E which is approximated by the semiclassical WKB exponents, $\Gamma_n \sim e^{-2W(E)}$. In the quasi-classical approximation the transition rate is given by $\Gamma \sim e^{-F_{\min}/T}$, where F_{\min} is the minimum of the effective "free energy" [3]

$$F = E + TW'(E), \quad W'(E) = 2W(E) \quad (15)$$

with respect to E . In the case of second-order transition the crossover temperature is given by

$$T_0^{(2)} = \frac{\tilde{\omega}_0}{2\pi}, \quad \tilde{\omega}_0 = \frac{\tilde{S}D}{\pi} \sqrt{h_x(1-h_x)} \quad (16)$$

where $\tilde{\omega}_0 = \sqrt{|U''(0)|/m}$ is the instanton frequency. It is convenient to introduce dimensionless temperature and energy variables

$$\theta = \frac{T}{T_0^{(2)}}, \quad P = \frac{\Delta U - E}{\Delta U}, \quad (17)$$

where $\Delta U = U_{\max} - U_{\min}$ is the barrier height. To investigate the phase transition behavior, we need to expand the free energy around the top of the potential barrier. Near the potential maximum ($k \sim 0$) the expansion of elliptic integrals up to order of k^6 is seen to be [15]

$$\begin{aligned} K(k) &= \frac{\pi}{2} \left[1 + \frac{k^2}{4} + \frac{9}{64}k^4 + \frac{25}{256}k^6 + \dots \right] \\ E(k) &= \frac{\pi}{2} \left[1 - \frac{k^2}{4} - \frac{3}{64}k^4 - \frac{5}{256}k^6 - \dots \right] \\ \Pi(\tanh^2 x_1, k) &= \frac{\pi}{2} + \frac{\pi}{8}(3 - 2h_x)k^2 + \frac{\pi}{128} (32h_x^3 - 8h_x^2 - 60h_x + 45) k^4 \\ &\quad + \frac{\pi}{512} (-256h_x^5 + 64h_x^4 + 480h_x^3 - 88h_x^2 - 350h_x + 175) k^6. \end{aligned} \quad (18)$$

The other parameters in the free energy, such as α^2 and ξ_p , are also calculated in the same way and we obtain the result

$$F/\Delta U = 1 + 4h_x(\theta - 1)k^2 + 4h_x \left(h_x^2 + 2h_x - 1 - \theta \left(h_x^2 + \frac{3}{2}h_x - \frac{7}{8} \right) \right) k^4 + 4h_x \left(\theta \left(2h_x^4 + 3h_x^3 - h_x^2 - \frac{11}{4}h_x + \frac{51}{64} \right) - \left(2h_x^4 + 4h_x^3 - 4h_x + 1 \right) \right) k^6. \quad (19)$$

There exists an exact relation between k^2 and P

$$k^2 = \frac{-2h_x - P + h_x P + 2h_x \sqrt{1 - P}}{-2h_x - P + h_x P - 2h_x \sqrt{1 - P}}. \quad (20)$$

Expressing k^2 in power series of P , we have

$$F/\Delta U = 1 + (\theta - 1)P + \frac{\theta}{8} \left(1 - \frac{1}{4h_x} \right) P^2 + \frac{3\theta}{64} \left(1 - \frac{1}{3h_x} + \frac{1}{16h_x^2} \right) P^3 + O(P^4) \quad (21)$$

which coincides with Ref. [3] exactly. There indeed exists a phase boundary between the first- and second-order transitions, i.e., $h_x = \frac{1}{4}$, at which the factor in front of P^2 changes the sign. We conclude that this critical boundary is inherent in the DSHG model and plays dominant role in the tunneling process of the uniaxial spin system with an external magnetic field, eq. (1).

Turning now to the computation of level splittings of excited states. We have a more general formula for the double- well-like potentials in WKB approximation [16,17,3]

$$\Delta E = \frac{\omega(E)}{\pi} \exp[-W] \quad (22)$$

where $\omega(E) = \frac{2\pi}{t(E)}$ is the energy-dependent frequency and $t(E)$ is the period of the real-time oscillation in the potential well. This level splitting formula is valid for entire energy region $0 < E < \Delta U$. The calculation of the period $t(E)$, equivalently the normalization constant of WKB wave functions [18], results in

$$t(E) = \sqrt{2m} \int_{x_1}^{x_2} \frac{dx}{\sqrt{E - U(x)}} = \frac{2K(k')}{\tilde{S}D\sqrt{1 - (\sqrt{E'} - h_x)^2}}. \quad (23)$$

For energies near the bottom of the potential well the energy-dependent frequency tends to the classical frequency of small oscillations at the bottom of the well $\omega_0 = 2\tilde{S}D\sqrt{1 - h_x^2}$

while near the barrier top this frequency reduces to the instanton frequency $\tilde{\omega}_0$. Here we discuss the low energy limit of the level splitting. Under the condition $E \ll \Delta U$, the action W may be expanded in power series of k' according to another group of formulae

$$\begin{aligned} K(k) &= \ln \frac{4}{k'} + \frac{1}{4} \left(\ln \frac{4}{k'} - 1 \right) k'^2 + \frac{9}{64} \left(\ln \frac{4}{k'} - \frac{7}{6} \right) k'^4 \quad (24) \\ E(k) &= 1 + \frac{1}{2} \left(\ln \frac{4}{k'} - \frac{1}{2} \right) k'^2 + \frac{3}{16} \left(\ln \frac{4}{k'} - \frac{13}{12} \right) k'^4 \\ \Pi(\tanh^2 x_1, k) &= -\frac{\sqrt{1-h_x^2}}{2h_x} x_0 + \frac{1+h_x}{2h_x} \ln \frac{4}{k'} - \frac{1+h_x+\sqrt{1-h_x^2}x_0}{8h_x} k'^2 + \frac{h_x+1}{8h_x} k'^2 \ln \frac{4}{k'} \\ &\quad - \left(\frac{4h_x^2-1}{64h_x^3} \sqrt{1-h_x^2}x_0 + \frac{3}{256} \frac{7h_x^3+8h_x^2-1}{h_x^3} \right) k'^4 + \frac{9h_x^3+11h_x^2-2}{128h_x^3} k'^4 \ln \frac{4}{k'} \end{aligned}$$

and $k'^4 \approx 16E'h_x^2/(1-h_x^2)^2$. Inserting them back into (14) we obtain

$$W = 2\tilde{S}\sqrt{1-h_x^2} \left(\frac{2x_0}{\sqrt{1-h_x^2}} - 1 - \frac{1-h_x^2}{64h_x^2} k'^4 - \frac{1-h_x^2}{16h_x^2} k'^4 \ln \frac{4}{k'} \right) \quad (25)$$

which reduces to the ground state action (5) at $k' = 0$. This expression may be rewritten in a more compact form

$$W = S_c(0) - \frac{E}{\omega_0} \ln \frac{eq\omega_0}{E} \quad (26)$$

with

$$q = \frac{8\tilde{S}(1-h_x^2)^{3/2}}{h_x^2}. \quad (27)$$

Approximating the energy levels in the well by a harmonic oscillator, i.e., $E = (n + \frac{1}{2})\omega_0$, and taking into account corrections from the functional-integral technique [16,17,3], simplify (26) into

$$W = S_c(0) - \ln \left(\frac{8\tilde{S}e(1-h_x^2)^{\frac{3}{2}}}{(n + \frac{1}{2})h_x^2} \right)^{(n+\frac{1}{2})}, \quad (28)$$

so the low-lying energy level splitting takes the form [16,10]

$$\Delta E_n = \frac{\Delta E_0}{n!} q^n \quad (29)$$

where the ground state splitting

$$\Delta E_0 = \frac{8\tilde{S}^{\frac{3}{2}}D}{\pi^{\frac{1}{2}}} \left[\frac{\exp \sqrt{1-h_x^2}}{1+\sqrt{1-h_x^2}} \right]^{2\tilde{S}} (1-h_x^2)^{5/4} h_x^{2\tilde{S}-1} \sim h_x^{2S} \quad (30)$$

is proportional to power $2S$ of the perturbative transverse field h_x [10].

Now consider the phase transition in a biaxial anisotropic ferromagnetic grain without applied magnetic field

$$\mathcal{H} = K_1 S_z^2 + K_2 S_y^2 \quad (31)$$

which has been investigated intensively [8,13,19]. This model possesses a XOY easy plane and an easy axis along the x direction with the anisotropy constants $K_1 > K_2 > 0$. We show here that it provides another example which exists first-order transition from classical to quantum regimes. With the help of the coherent-state path integral the effective Hamiltonian in terms of continuum field variable can be written in the form $\mathcal{H} = \frac{p^2}{2m(\phi)} + U(\phi)$ and

$$m(\phi) = \frac{1}{2K_1(1-\lambda \sin^2 \phi)}, \quad U(\phi) = K_2 S(S+1) \sin^2 \phi \quad (32)$$

where $\lambda = \frac{K_2}{K_1} < 1$ and mass $m(\phi)$ is field dependent. The periodic instanton solution is given by [19]

$$\phi_p = \arcsin \sqrt{\frac{1-k^2 \operatorname{sn}^2(\omega\tau, k)}{1-\lambda k^2 \operatorname{sn}^2(\omega\tau, k)}} \quad (33)$$

with

$$k = \sqrt{\frac{n_1^2 - 1}{n_1^2 - \lambda}}, \quad n_1^2 = \frac{K_2 S(S+1)}{E_{cl}} \quad (34)$$

where $\omega = \omega_0 \sqrt{1-\lambda/n_1^2}$ denotes the small oscillation frequency at the position of periodic instanton and $\omega_0^2 = 4S(S+1)K_1K_2$. The non-monotonically decreasing behavior of the periodicity of the solution, $\tau(E) = 4K(k)/\omega$, has been pointed in Ref. [4] where the authors proved that beyond a critical value of coupling the spin system acquired a first order transition as a result of the field dependence of its effective mass. We now turn to evaluate

the effective free energy dependence on dimensionless energy scale P for various anisotropy constants.

The Euclidean action evaluated for the periodic instanton trajectory is given by eq. (13) with

$$W = \frac{\omega}{\lambda K_1} \left[K(k) - (1 - \lambda k^2) \Pi(\lambda k^2, k) \right]. \quad (35)$$

The second order transition temperature for this model is defined by eq.(16) with the instanton frequency $\tilde{\omega}_0 = \sqrt{|U''(\pi/2)|/m(\pi/2)} = \sqrt{1 - \lambda} \omega_0$. Near the top of the barrier the third kind of elliptic integral should be expanded as

$$\Pi(\lambda k^2, k) = \frac{\pi}{2} + (2\lambda + 1) \frac{\pi}{8} k^2 + (8\lambda^2 + 4\lambda + 3) \frac{3\pi}{128} k^4 + (16\lambda^3 + 8\lambda^2 + 6\lambda + 5) \frac{5}{512} \pi k^6. \quad (36)$$

After a straightforward calculation we obtain the expansion result for the free energy (15) in terms of k up to the sixth order

$$F/\Delta U = 1 - P + \theta(1 - \lambda) \left(k^2 + (6\lambda + 1) \frac{k^4}{8} + (40\lambda^2 + 8\lambda + 3) \frac{k^6}{64} \right). \quad (37)$$

The effective free energy analogous to Landau theory of phase transitions near the top of the barrier ($P \ll 1$) reads

$$F/\Delta U = 1 + (\theta - 1) P + \frac{\theta}{8(1 - \lambda)} (1 - 2\lambda) P^2 + \frac{\theta}{64(1 - \lambda)^2} (8\lambda^2 - 8\lambda + 3) P^3 + O(P^4) \quad (38)$$

with the exact relation between k^2 and P

$$k^2 = \frac{P}{1 - \lambda(1 - P)}. \quad (39)$$

The factor before P changes sign at the phase transition temperature $T = T_0^{(2)}$. The boundary between the first- and the second-order transition is obviously seen to be $\lambda_c = \frac{1}{2}$. The computed dependence of F on P for the entire range of energy is plotted in Fig. 2 for $S^2 = 1000$, $K_1 = 1$. At $\lambda = 0.3$, there is only one minimum of F at the top of the barrier

for all $T > T_0^{(2)}$. Below $T_0^{(2)}$ it continuously shifts to the bottom as the temperature is lowered. This corresponds to the second-order transition from thermal activation to thermally assisted tunneling. At $\lambda = 0.8$, however, there can be one or two minima of F , depending on the temperature. The actual phase transition (in this case the first-order) occurs at the temperature when the two minima have the same free energy, which for $\lambda = 0.8$ takes place at $T_0^{(1)} = 1.122T_0^{(2)}$.

The relation between tunneling at excited states and finite temperature can be understood as follows: Below the crossover temperature T_0 the particle tunnels through the barrier at the most *favorable* energy level $E(T)$ which goes down from the top of the barrier to the bottom of the potential with lowering temperature. Such a regime is called thermally assisted tunneling (TAT). The second order transition from the classical thermal activation to TAT is smooth, with no discontinuity of $d\Gamma/dt$ at T_0 , and the transition temperature is given by $T_0^{(2)}$. In the situation under which the first order transition may occur tunneling just below the top of the barrier is *unfavorable*, the TAT is suppressed, and the thermal activation competes with the ground state tunneling directly, leading to the first order transition.

The flatness of the barrier top is not the necessary condition under which the first order transition may occur. For the constant mass model the more favorable potential for the first order transition is that the top of the barrier should be wider so that the particle doesn't have more advantage to tunnel through the barrier from the excited states than from the ground state. In our second model the pseudo-particles near the top of the barrier are "heavier" than those in the bottom of the well, i.e. $m(\pi/2) = m(0)/(1 - \lambda) > m(0)$. The advantage for the particles to tunnel through the barrier at higher excited states is again not very obvious, which leads to the first order transition from the thermal activation directly to ground state tunneling.

A simple estimation for the crossover temperature $T_0^{(0)}$ is given by

$$T_0^{(0)} = \frac{\Delta U}{2S_c(E=0)} = \frac{K_2(S + \frac{1}{2})}{2 \ln \frac{1+\sqrt{\lambda}}{1-\sqrt{\lambda}}} \quad (40)$$

where the superscript of $T_0^{(0)}$ indicates that the ground state tunneling is considered. In Fig.

3 we plot the dependence of the actual crossover temperature on λ . For $\lambda < \frac{1}{2}$, the actual crossover temperature $T_0^{(1)}$ is read out from Fig. 2(b). The escape rate can be conveniently represented in terms of the effective temperature defined by

$$\Gamma \sim \exp\left[-\frac{\Delta U}{T_{eff}(T)}\right] = \exp\left[-\frac{F_{\min}}{T}\right] \quad (41)$$

The dependence $T_{eff}(T) = \Delta UT/F_{\min}$ is represented in Fig. 4 for different anisotropy constant ratio λ . The most significant difference between the crossover temperature $T_0^{(0)}$ and the actual crossover temperature T_0 arises in the limit $\lambda \rightarrow 0$, that is, $T_0^{(0)}/T_0^{(2)} = \pi/4 \approx 0.785$.

The first-order escape-rate transition considered above is the transition from thermal activation to thermally assisted tunneling near the bottom of the potential and not directly to the ground-state tunneling [3] due to the logarithmic divergence of the instanton period $\tau(E)$ for the energies near U_{\min} . In some field-theoretical models, as, e.g., the reduced nonlinear $O(3)$ - σ model, τ approaches 0 near the bottom of the potential. Accordingly, the second derivative of $W(E)$ and $F(E)$ is negative everywhere, as for the rectangular potential for particles. In such a situation, as it is clear from Fig. 2(b), the minimum of $F(E)$ can only be at the barrier top or potential bottom. That is, thermal activation competes directly with the ground-state tunneling. It was shown that [20] adding a small Skyrme term to the reduced nonlinear $O(3)$ - σ model causes τ to diverge near the bottom of the potential, with the accordingly small amplitude. This is, in a sense, similar to the situation realized in this spin model for $\lambda \rightarrow 1$.

In conclusion, we present in this letter the periodic instanton calculation for the first- and second-order transitions between quantum and classical regimes for two spin models. The level splitting formula for the excited states is derived and checked for the uniaxial spin system. Our results for uniaxial spin model confirm Ref. [3] and for the biaxial anisotropic spin system we find that the transition from the classical regimes with lowering temperature is of the first order for λ above the phase boundary line $\lambda_c = \frac{1}{2}$, and of the second-order below this critical value.

Helpful discussions with Dr. X. B. Wang, Prof. Lee Chang and Prof. B. Z. Li are appreciated. This Work is partly supported by the National Natural Science Foundation of China under Grant Nos. 19677101 and 19775033.

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Figure Captions:

Fig. 1. The DSHG potential and the periodic instanton configuration for $S = 10, D = 0.6K, h_x = 0.01$

Fig. 2. Effective free energy for the escape rate: (a) $\lambda = 0.3$, second-order transition; (b) $\lambda = 0.8$, first order-transition.

Fig. 3. Dependence of the actual phase transition temperature T_0 on the anisotropy constant ratio.

Fig. 4. Dependence of the effective temperature T_{eff} on T for different values of λ