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Quantum-classical transition of the escape rate of a biaxial ferromagnetic spin with an external magnetic field

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ABSTRACT

We study the model of a biaxial single ferromagnetic spin Hamiltonian with an external magnetic field applied along the medium axis. The phase transition of the escape rate is investigated. Two different but equivalent methods are implemented. Firstly, we derive the semi-classical description of the model which yields a potential and a coordinate dependent mass. Secondly, we employ the method of spin-particle mapping which yields a similar potential to that of semi-classical description but with a constant mass. The exact instanton trajectory and its corresponding action, which have not been reported in any literature is being derived. Also, the analytical expressions for the first- and second-order crossover temperatures at the phase boundary are derived. We show that the boundary between the first-and the second-order phase transitions is greatly influenced by the magnetic field.

1. Introduction

In recent years, the study of single ferromagnetic spin systems has been of considerable interest to condensed matter physicists. These systems have been pointed out [1,3] to be a good candidate for investigating first- and second-order phase transitions of the quantum-classical escape rate. The quantum-classical escape rate transition takes place in the presence of a potential barrier. At very low temperature (close to zero), transitions occur by quantum tunnelling through the barrier and the rate is governed by $\Gamma \sim e^{-B}$, where B is the instanton (imaginary time solution of the classical equation of motion) action. At high temperatures, the particle has the possibility of hopping over the barrier (classical thermal activation), in this case transition is governed by $\Gamma \sim e^{-\Delta V/T}$, where ΔV is the energy barrier. At the critical point when these two transition rates are equal, there exists a crossover temperature (first-order transition) $T_0^{(1)}$ from a quantum to a thermal regime, it is estimated as $T_0^{(1)} = \Delta V/B$. In principle these transitions are greatly influenced by the anisotropy constants and the external magnetic fields. The second-order phase transition occurs for particles in a cubic or quartic parabolic potential, it takes place at the temperature $T_0^{(2)}$, below $T_0^{(2)}$ one has the phenomenon of thermally assisted tunnelling and above $T_0^{(2)}$ transition occurs due to thermal activation to the top of the potential barrier [1,3]. The order of these transitions can also be determined from the period of oscillation $\tau(E)$ near the bottom of the inverted potential. Monotonically increasing $\tau(E)$ with the amplitude of oscillation gives a second-order transition while nonmonotonic behaviour of $\tau(E)$ (that is a minimum in the $\tau(E)$ vs E curve, with E being the energy of the particle) gives a first-order transition [1].

The model of a uniaxial single ferromagnetic spin with a transverse magnetic field, which is believed to describe the molecular magnet $MnAc_{12}$ was considered by Garanin and Chudnovsky [1], the Hamiltonian is of the form $\hat{H} = -D\hat{S}_z^2 - h_x\hat{S}_x$, using the spin-particle mapping version of this Hamiltonian [5–7], they showed that the transition from the thermal to the quantum regime is of the first-order in the regime $h_x < sD/2$ and of the second-order in the regime $sD/2 < h_x < 2sD$. For other single-molecule magnets such as Fe_8, a biaxial ferromagnetic spin model is a good approximation. In this case, Lee et al. [13] considered the model $\hat{H} = K(\hat{S}_z^2 + \lambda\hat{S}_y^2) - 2\mu_B h_y \hat{S}_y$, using spin coherent state path integral, they obtained a potential and a coordinate dependent mass from which they showed that the boundary between the first and the second-order transition set in at $\lambda = 0.5$ for $h_y = 0$ while the order of the transitions is greatly influenced by the magnetic field and the anisotropy constants for $h_y \neq 0$. Zhang et al. [14] studied the model $\hat{H} = K_1\hat{S}_z^2 + K_2\hat{S}_y^2$ using spin-particle mapping and periodic instanton method. The phase boundary between the first- and the second-order transitions was shown to occur at $K_2 = 0.5K_1$. The model with z-easy axis in an applied field has also been studied by numerical and perturbative methods [2]. In this paper, we study a biaxial spin system with an external magnetic field applied along the medium axis using spin-coherent state path integral and the

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formalism of spin-particle mapping. Unlike other models with an external magnetic field [4,12,14], the spin-particle mapping yields a simplified potential and a constant mass which allows us to solve for the exact instanton trajectory and its corresponding action in the presence of a magnetic field. We also present the analytical results of the crossover temperatures for the first- and the second-order transitions at the phase boundary.

2. Spin model and spin coherent state path integral

Consider the Hamiltonian of a biaxial ferromagnetic spin (single-molecule magnet) in an external magnetic field

$$\hat{H} = \mathcal{D}\hat{\mathcal{S}}_z^2 + \mathcal{E}\hat{\mathcal{S}}_x^2 - h_x\hat{\mathcal{S}}_x \tag{1}$$

where $\mathcal{D} \gg \mathcal{E} > 0$, and $\mathcal{S}_i,\ i = x, y, z$, is the components of the spin. This model possesses an easy XOY plane with an easy-axis along the y-direction and an external magnetic field along the x-axis. At zero magnetic field, there are two classical degenerate ground states corresponding to the minima of the energy located at $\pm y$, these ground states remain degenerate for $h_x \neq 0$ in the easy XY plane. The semi-classical form of the quantum Hamiltonian can be derived using spin coherent state path integral. In the coordinate dependent form, spin-coherent-state is defined by [15,16]

$$|\hat{\mathbf{n}}\rangle = \left(\cos\frac{1}{2}\theta\right)^{2s} \exp\left\{\tan\left(\frac{1}{2}\theta\right)e^{i\phi}\hat{\mathcal{S}}^-\right\}\| s, s\rangle \tag{2}$$

where $\hat{\mathbf{n}} = s(\sin\,\theta\,\cos\,\phi,\ \sin\,\theta\,\sin\phi,\ \cos\,\theta)$ is the unit vector parametrizing the spin on a two-sphere S^2. The overlap between two coherent states is found to be

$$\langle\hat{\mathbf{n}}'|\hat{\mathbf{n}}\rangle = \left[\cos\,\frac{1}{2}\theta\,\cos\,\frac{1}{2}\theta' + \sin\,\frac{1}{2}\theta\,\sin\frac{1}{2}\theta' e^{-i\Delta\phi}\right]^{2s} \tag{3}$$

where $\Delta\phi = \phi' - \phi$. The expectation value of the spin operator in the large s limit is approximated as $\langle\hat{\mathbf{n}}'|\hat{\mathcal{S}}|\hat{\mathbf{n}}\rangle \approx s[\hat{\mathbf{n}} + O(\sqrt{s})]\langle\hat{\mathbf{n}}'|\hat{\mathbf{n}}\rangle$. For an infinitesimal separated angle, $\Delta\theta = \theta' - \theta$, Eq. (3) reduces to

$$\langle\hat{\mathbf{n}}'|\hat{\mathbf{n}}\rangle \approx 1 - is\Delta\phi(1 - \cos\,\theta). \tag{4}$$

These states satisfy the overcompleteness relation (resolution of identity)

$$\mathcal{N}\int d\phi\, d(\cos\,\theta)|\hat{\mathbf{n}}\rangle\langle\hat{\mathbf{n}}| = \hat{I}. \tag{5}$$

Using these equations, the transition amplitude is easily obtained as

$$\langle\hat{\mathbf{n}}_f|e^{-\beta\hat{H}}|\hat{\mathbf{n}}_i\rangle = \int \mathcal{D}\phi\mathcal{D}(\cos\,\theta)e^{-S} \tag{6}$$

The Euclidean action ($t \to -i\tau$) is given by $S = \int_{-\beta/2}^{\beta/2} d\tau\,\mathcal{L}$, with

$$\mathcal{L} = is\dot{\phi}(1 - \cos\,\theta) + V(\theta, \phi) \tag{7}$$

$$V(\theta, \phi) = \mathcal{D}s^2\,\cos^2\theta + \mathcal{E}s^2\,\sin^2\theta\,\cos^2\phi - sh_x\,\sin\,\theta\,\cos\,\phi \tag{8}$$

These two equations (7) and (8) describe the semi-classical dynamics of the spin on S^2. Two degenerate minima exist for $h_x < h_c = 2\mathcal{E}s$, which are located at $\theta = \pi/2 : \phi = 2\pi n \pm \arccos\,\alpha_x$, where $\alpha_x = h_x/h_c$, $n \in \mathbb{Z}$, and the maximum is at $\theta = \pi/2$: $\phi = n\pi$ with the height of the barrier ($n = 0$) given by

$$\Delta V = \mathcal{E}s^2(1 - \alpha_x)^2 \tag{9}$$

Taking into consideration the fact that $\mathcal{D} \gg \mathcal{E}$, the deviation away from the easy plane is very small, thus one can expand $\theta = \pi/2 - \eta$, where $\eta \ll 1$. Integration over the fluctuation η in Eq. (6) yields an effective theory described by

$$\mathcal{L}_{\text{eff}} = is\dot{\phi} + \tfrac{1}{2}m(\phi)\dot{\phi}^2 + V(\phi) \tag{10}$$

where

$$V(\phi) = \mathcal{E}s^2(\cos\,\phi - \alpha_x)^2 \tag{11}$$

and

$$m(\phi) = \frac{1}{2\mathcal{D}(1 - \kappa\,\cos^2\phi + 2\alpha_x\kappa\,\cos\,\phi)} \tag{12}$$

with $\kappa = \mathcal{E}/\mathcal{D}$. An additional constant of the form $\mathcal{E}s^2\alpha_x^2$ has been added to the potential for convenience. The first term in the effective Lagrangian is a total derivative which does not contribute to the classical equation of motion, however, it has a significant effect in the quantum transition amplitude, producing a quantum phase interference in spin systems [10,11]. The two classical degenerate minima which correspond to $\phi = 2\pi n \pm \arccos\,\alpha_x$ are separated by a small barrier at $\phi = 0$ and a large barrier at $\phi = \pi$. The phase transition of the escape rate of this model can be investigated using the potential Eq. (11) and the mass Eq. (12) [13], in this paper, however, we will study this transition via the method of mapping a spin system onto a quantum mechanical particle in a potential field. A classical trajectory (instanton) exists for zero magnetic field, in this case the classical equation of motion

$$m(\overline{\phi})\ddot{\overline{\phi}} + \frac{1}{2}m(\overline{\phi})'\dot{\overline{\phi}} = \frac{dV}{d\overline{\phi}} \tag{13}$$

integrates to

$$\sin\,\overline{\phi} = \pm\frac{\sqrt{(1-\kappa)}\,\tanh(\omega\tau)}{\sqrt{1 - \kappa\,\tanh^2(\omega\tau)}} \tag{14}$$

where $\omega = 2s\sqrt{\mathcal{E}\mathcal{D}}$ and the upper and lower signs are for instanton and anti-instanton respectively. The corresponding action for this trajectory yields [10,17] $S_0 = B \pm is\pi$

$$B = s\,\ln\left(\frac{1 + \sqrt{\kappa}}{1 - \sqrt{\kappa}}\right) \tag{15}$$

For small anisotropy parameters, $\kappa \ll 1$, the coordinate dependent mass can be approximated as $m \approx 1/2\mathcal{D}$, the approximate instanton trajectory in this limit yields

$$\sin\,\overline{\phi} = \pm\frac{2\sqrt{\dfrac{1-\alpha_x}{1+\alpha_x}}\,\tanh(\omega\tau)}{\left[1 + \dfrac{1-\alpha_x}{1+\alpha_x}\,\tanh^2(\omega\tau)\right]} \tag{16}$$

where $\omega = s\sqrt{\mathcal{E}\mathcal{D}(1 - \alpha_x^2)}$ and the corresponding action is

$$B = 2s\sqrt{\kappa}[\sqrt{1 - \alpha_x^2} \pm \alpha_x\,\arcsin(\sqrt{1 - \alpha_x^2})] \tag{17}$$

The upper and the lower sign in the action correspond to the large and small barriers respectively while that in the trajectory is for instanton and anti-instanton. At zero magnetic field, the instanton interpolates between the classical degenerate minima $\overline{\phi} = \pm\pi/2$ at $\tau = \pm\infty$. For coordinate dependent mass the classical trajectory can be integrated in terms of the Jacobi elliptic functions. This solution will be presented in the next section using a simpler method.

3. Particle mapping

In this section, we will consider the formalism of mapping a spin system to a quantum-mechanical particle in a potential field [5]. In this formalism one introduces a non-normalized spin coherent state, the action of the spin operators on this state yields