Quantum-classical phase transition of the escape rate in a biaxial spin system with an arbitrarily directed magnetic field

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(August 10, 2018)

We investigate the escape rate of a biaxial spin particle with an arbitrarily directed magnetic field in the easy plane, described by Hamiltonian $H = -AS_z^2 - BS_x^2 - H_xS_x - H_zS_z$, ($A > B > 0$). We derive an effective particle potential by using the method of particle mapping. With the help of the criterion for the presence of a first-order quantum-classical transition of the escape rate we obtained various phase boundary curves depending on the anisotropy parameter $b \equiv B/A$ and the field parameters $\alpha_{x,z} \equiv H_{x,z}/AS$: $\alpha_{zc}(b_c)'s$, $\alpha_{xc}(b_c)'s$, and $\alpha_{zc} = \alpha_{zc}(\alpha_{xc})$. It is found from $\alpha_{zc}(b_c)'s$ and $\alpha_{xc}(b_c)'s$ that the first-order region decreases as $b$ and $\alpha_x$ (or $\alpha_z$) increase. The phase boundary line $\alpha_{zc} = \alpha_{zc}(\alpha_{xc})$ shows that compared with the uniaxial system, both the first- and second-order regions are diminished due to the transverse anisotropy. Moreover, it is observed that, in the limit $\alpha_{xc} \to 0$, $\alpha_{zc}$ does not coincides with the coercive field line, which yields more reduction in the first-order region. We have also computed the crossover temperatures at the phase boundary : $T_c(b_c), T_c(\alpha_{xc}, \alpha_{zc})$.

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

Recently there have been intensive studies on the quantum-classical phase transition of the escape rate in a single domain magnetic particle with many spins. In such a particle the magnetization direction of a collection of spins is oriented such that the magnetocrystalline anisotropy energy is at a stable or metastable minimum depending on the existence of an external magnetic field. The escape from a stable or metastable state is governed by classical thermal activation rate, which is proportional to \( \exp(-E_0/k_B T) \), at high temperature, and by quantum tunneling at temperatures below the energy barrier \( E_0 \). When these two escape rates are same there exists a crossover temperature \( T_0 \) at which the transition between classical and quantum regimes occurs. The transition can be either first-order or second-order. In the first-order transition the escape rate abruptly changes from the temperature-dependent thermal activation process to a practically temperature-independent quantum tunneling, so that the first derivative of the escape rate at the crossover temperature changes discontinuously. In the second-order transition, however, the escape rate changes smoothly from classical regime to temperature-dependent quantum tunneling (thermally assisted tunneling), and has a discontinuity of the second derivative. The determination of the transition order is closely related to the shape of the potential barrier which is controlled by the anisotropy constant and the external magnetic field.

The quantum-classical transition of the escape rate was investigated by Affleck \[2\] and Larkin and Ovchinnikov. \[3\] By using instanton technique they demonstrated that under certain assumption on the shape of potential barrier a smooth interpolation between the periods of oscillations at the bottom and the top of Euclidean potential well can be made, which leads to a second-order phase transition between classical and quantum regimes. Later, Chudnovsky \[4\] observed that the order of the phase transition in the crossover from classical activation to thermally assisted tunneling completely depends on the shape of the potential barrier. He has shown that the behavior of the energy-dependent period of oscillations \( \tau(E) \) in Euclidean potential determines the order of the quantum-classical transition; if the Euclidean period increases monotonically with the increasing energy \( E \) from the bottom
of the Euclidean potential the transition is second-order, but if \( \tau(E) \) is nonmonotonic such that it has a minimum at \( E = E_1 \) which is smaller than the potential barrier \( E_0 \) the first-order transition occurs. More recently, Gorokhov and Blatter \cite{5} have obtained a sufficient condition for the first-order quantum-classical phase transition by looking at the behavior of the oscillation period in Euclidean time as a function of oscillation amplitude near the barrier top in the two-dimensional string model. In this case the first-order transition appears when the amplitude-dependent period \( \tau(a) \), where \( a \) is the amplitude, is smaller than the zero amplitude period \( \tau(0) \) near the barrier top. This method has been subsequently extended to a quantum-mechanical model where mass has coordinate-dependence. \cite{6}

The above approaches have been applied to the quantum-classical phase transition of the escape rate in a single domain spin system. Up to now two types of spin systems have been studied intensively: uniaxial and biaxial systems. For the uniaxial system, such as high-spin molecular magnet Mn\(_{12}\) Ac, \cite{7} two models have been considered: one with a transverse field and the other with an arbitrarily directed field, described by Hamiltonians \( \mathcal{H} = -DS_z^2 - H_xS_x \) \cite{8} and \( \mathcal{H} = -DS_z^2 - H_xS_x - H_zS_z \), \cite{9} respectively. The biaxial spin system, such as iron cluster Fe\(_8\), \cite{10} has attracted more attention, and several models have been taken into account. Liang \textit{et al.} \cite{11} considered a model without an applied field, \( \mathcal{H} = K(S_z^2 + \lambda S_y^2), (0 < \lambda < 1) \) by using the periodic instanton approach and demonstrated that the coordinate-dependent effective mass plays an important role for the presence of the first-order transition. Based on the same approach Lee \textit{et al.} \cite{12} investigated the biaxial spin model with a transverse field, \( \mathcal{H} = K(S_z^2 + \lambda S_y^2) - H_yS_y \), and showed that the nonconstant mass which depends on both coordinate and field is important for the occurrence of the first-order transition. However, this approach involves a restricted range of applicability of \( \lambda << 1 \) for the biaxial spin system with a field. Such a restriction can be avoided by introducing the method of particle mapping. \cite{13} The same model has been considered by Kim \cite{14} who used a quasiclassical method based on the particle mapping and found an analytical form of the phase boundary curve between first- and second-order transitions. The phase boundary of a biaxial system with longitudinal field described by \( \mathcal{H} = K(S_z^2 + \lambda S_y^2) - H_xS_x \) has also been obtained by Garanin and Chudnovsky \cite{15} employing a perturbation approach.
with respect to the transverse anisotropy, by Kim [14] using the quasiclassical method based on particle mapping, and by Park et al. [16] with the help of the Gorokhov and Blatter's criterion [5] and the particle mapping.

In this paper we study the phase transition of the escape rate of a biaxial spin system with an arbitrarily directed magnetic field in the easy plane. In this case the transverse and longitudinal fields coexist, and there are three parameters which can be controlled by experiment: the anisotropy constant and two field parameters. In order to find a phase diagram for the transition orders these should be considered simultaneously. We will use the method of particle mapping to derive an effective particle potential from the spin Hamiltonian. Then, by applying the criterion developed in Refs. [5, 6] to this potential we will find various phase diagrams depending on the three parameters. Especially, we will obtain a phase boundary curve between the first- and the second-order transitions for the iron cluster Fe$_8$ in which a relation between the transverse and longitudinal field parameters is given. We will also find the crossover temperature at the phase boundary.

In the following section, we present a derivation of the effective particle potential based on the method of particle mapping. Here, we will take a constant mass so that the effective potential includes all the coordinate dependence. We will also review the sufficient condition for the presence of the first-order transition. In Sec. III we show various phase boundary curves between the first- and second-order transitions. For completeness we will also briefly discuss two special cases: the biaxial systems with the transverse field only and with the longitudinal field only. These diagrams are compared with the previously obtained results in uniaxial system. The crossover temperature at the phase boundary is also computed here. Finally, there will be a summary and discussions in Section IV.

II. PARTICLE MAPPING AND THE CRITERION FOR THE FIRST-ORDER TRANSITION

Consider a biaxial single domain magnetic particle with $XOZ$ easy-plane and the easy $Z$-axis in the $XZ$ plane. When an external field is applied along an arbitrary direction in
the $XZ$ plane the Hamiltonian can be described by

$$\mathcal{H} = -AS_z^2 - BS_x^2 - H_xS_x - H_zS_z,$$

(2.1)

where $A$ and $B$ are the longitudinal and transverse anisotropy constants, respectively satisfying $A > B > 0$. Our model is equivalent to $\mathcal{H} = K(S_z^2 + \lambda S_y^2) - H_xS_x - H_yS_y$ if we set $A = K$, $B = (1 - \lambda)K$. For convenience, we introduce dimensionless transverse anisotropy parameter $b \equiv B/A(< 1)$ and field parameters $\alpha_x \equiv H_x/SA$, $\alpha_z \equiv H_z/SA$ where $S$ is the spin number. Following Ref. [13] this spin problem can be reduced to a particle moving in an effective potential. The equivalent particle Hamiltonian can be written as

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

(2.2)

where $m \equiv 1/2A$ is the particle mass, and $V(x) \equiv Av(x)$ is the effective particle potential given by

$$v(x) = \frac{1}{4dn^2 x}[S^2(\alpha_x snx - \alpha_z cnx)^2 - 4bS(S + 1)$$

$$- 2S(2S + 1)(b\alpha_z snx + \alpha_x cnx)]$$

(2.3)

in which $snx, cnx, dnx$ are the Jacobian elliptic functions with modulus $k^2 = 1 - b$. The Schrödinger-like equation corresponding to this Hamiltonian is $\mathcal{H}\Psi(x) = E\Psi(x)$ where $\Psi(x)$ is the particle wave function given by

$$\Psi(x) = \left(\frac{cnx}{dnx}\right)^S \exp \left[\frac{\alpha_x S}{2\sqrt{1 - b}} \tanh^{-1}(\sqrt{1 - bsnx})\right]$$

$$\times \exp \left[-\frac{\alpha_x S}{2\sqrt{b(1 - b)}} \tan^{-1}\left(\frac{b}{1 - b cnx}\right)\right]\Phi(x)$$

(2.4)

with

$$\Phi(x) = \sum_{\sigma = -S}^S \frac{C_\sigma}{\sqrt{(S + \sigma)!(S - \sigma)!}} \left(\frac{snx + 1}{cnx}\right)^\sigma.$$  

(2.5)

In Fig.1 the effective particle potential is drawn. It has a metastable minimum with asymmetric barriers: small and large ones. The heights and widths of these barriers are governed
by the anisotropy and field parameters $b$, $\alpha_x$, $\alpha_z$. For a given value of $b$ the height of the small barrier decreases as $\alpha_x$ or $\alpha_z$ increases and vanishes at a critical value, while that of the large barrier increases. The critical value is determined by the metastability condition which, for the present model, is derived as \cite{17}

\begin{equation}
\alpha_{xm}^{2/3} + \alpha_{zm}^{2/3} = [2(1 - b)]^{2/3}.
\end{equation}

For a given value of $b$ the metastable state exists inside the region closed by the curve $\alpha_{xm} = \alpha_{xm}(\alpha_{zm})$. We note here that because of the transverse anisotropy the metastability region of the biaxial system is smaller than the uniaxial system which corresponds to the case of $b = 0$. \cite{18}

The escape rate at temperatures below the barrier height $E_0$ can be obtained by taking thermal average over tunneling probabilities. In the semiclassical approximation this can be expressed as

\begin{equation}
\Gamma \propto \int_0^{E_0} dE P(E) e^{-E/T},
\end{equation}

where the metastable minimum is chosen to be zero energy. The tunneling probability $P(E)$ can be approximated by the WKB exponent, $P(E) \sim e^{-S(E)}$ in which $S(E)$ is the Euclidean action defined by

\begin{equation}
S(E) = 2\sqrt{2m} \int_{x_1(E)}^{x_2(E)} dx \sqrt{v(x) - E},
\end{equation}

where $x_1(E), x_2(E)$ are the turning points corresponding to energy $E$. For large spin system the semiclassical approximation is well applicable so that we can neglect the contributions from paths fluctuating around the semiclassical saddle point path which minimizes the Euclidean action $S(E)$. Within this approximation the escape rate becomes

\begin{equation}
\Gamma \sim \exp \left[ -\frac{S_{T\min}(E)}{T} \right],
\end{equation}

where $S_{T\min}(E)$ is the minimum of an energy function

\begin{equation}
S_T(E) = S(E) + E/T,
\end{equation}
which is called *thermon action* [4] or *periodic instanton action* in field theory. The condition for \( S_T(E) \) to have minimum requires \( dS_T(E)/dE = 0 \). Since the first derivative of an action with respect to energy in a potential well brings about the oscillation period we can write

\[
\tau(E) = -\frac{dS(E)}{dE} = \frac{1}{T},
\]

(2.11)

where negative sign attributes to the Euclidean potential in which the energy has negative values, and hence \( \tau(E) \) is an Euclidean time oscillation period. Since the Euclidean action \( S(E) \) is zero at \( E = E_0 \) (i.e., at the top of the barrier) the minimum of the energy function becomes

\[
S_{T\min}(E_0) \equiv S_0 = \frac{E_0}{T}
\]

(2.12)

which is just the exponent of the thermal activation rate, i.e., the *thermodynamic action*. [4]

The type of phase transition is determined by the behavior of \( \tau(E) \) with energy \( E \). As has been well analyzed by Chudnovsky [4] a first-order quantum-classical transition takes place when \( \tau(E) \) has a minimum at some energy \( E_1 (< E_0) \). In this case the crossover temperature \( T_0 = 1/\tau(E_0) \) is lower than \( T_1 = 1/\tau(E_1) \), and there exists a temperature \( T_c \) \((T_0 < T_c < T_1)\) corresponding to an Euclidean oscillation period \( \tau_c \) at which the classical escape rate with \( S_0 \) and the quantum escape rate with the minimum thermon action \( S_{T\min} \) are connected, but their first derivatives with respect to \( T \) are discontinuous. That is, the semiclassical saddle point path jumps from the quantum regime to the classical one at temperature \( T_c \). It can be noted from the behavior of \( \tau(E) \) that the first-order transition exist if the Euclidean oscillation period at an energy near the bottom of the Euclidean potential is smaller than \( \tau(E_0) \), the period corresponding to the crossover temperature.

The above argument can be generalized to the case in which the period is a function of the oscillation amplitude \( a \) near the bottom of the Euclidean potential. When \( E \) approaches to \( E_0 \) the dynamics in the Euclidean potential well becomes small oscillations. At \( E = E_0 \) the oscillation amplitude is zero, and the thermon action becomes the thermodynamic action. In this limit the period can be related to the crossover temperature \( T_0 \) as following:

\[
T_0 = 1/\tau(0) = \omega_0/2\pi,
\]

(2.13)
where \( \tau(0) \) is a period corresponding to zero amplitude, and \( \omega_0 \) is defined as

\[
\omega_0 = \sqrt{-\frac{v''(x_0)}{m}},
\]

where \( x_0 \) is the position of the top of potential barrier. The solution of small oscillations near this point can be obtained from the Euclidean Euler-Lagrange equation, and the oscillation period of the solution can be expressed by \( \tau(a) = 2\pi/\omega \). From the above discussion the condition for the presence of the first-order quantum-classical transition is then given by \( \tau(a) < \tau(0) \). The difference \( \tau(a) - \tau(0) \) satisfying this condition can be calculated by the perturbation method in which the oscillation amplitude \( a \) is used as a perturbation parameter. \[6\] For the case of constant mass the condition becomes

\[
-\frac{5}{24} \frac{v'''(x_0)}{v''(x_0)} + \frac{1}{8} v''''(x_0) < 0.
\]

(2.15)

Below we will use this criterion to compute the phase boundary curve between the first- and the second-order transitions. Once we obtain the boundary curve we can also calculate the crossover temperature at the phase boundary from Eq.(13).

III. PHASE BOUNDARY LINES BETWEEN THE FIRST- AND SECOND-ORDER TRANSITIONS

We start this section by considering two special cases where either the longitudinal field or the transverse field is applied, from which we deduce some important features that can be directly applied to the case with arbitrarily directed field. We first consider the model with transverse field only. When the longitudinal field is zero the model can be described by

\[
\mathcal{H} = -AS_z^2 - BS_x^2 - H_xS_z.
\]

(3.1)

This is equivalent to the previously studied models; if we set \( A = K \), \( B = (1 - \lambda)K \) it is same as the model considered in Ref. \[12\], and if \( A - B = K_\parallel \), \( B = K_\perp \) it becomes the model of Ref. \[14\]. The present approach based on the criterion described above, however,
is different from theirs. In this case since $\alpha_z = 0$ in Eq.(3) the effective particle potential is reduced as

$$v_t(x) = \frac{S^2\alpha_z^2 sn^2 x - 2S(2S + 1)\alpha_x cn x - 4bS(S + 1)}{4dn^2 x},$$

(3.2)

and the corresponding wave function is given by

$$\Psi_t(x) = \left(\frac{cn x}{dn x}\right)^S \times \exp \left[ -\frac{\alpha_x S}{2\sqrt{b}(1-b)} \tan^{-1}\left(\sqrt{\frac{b}{1-b} cn x}\right) \right] \Phi(x).$$

(3.3)

We note that the effective particle mass has no coordinate dependence, which is different from previous models where the coordinate dependent mass played a crucial role to present the first-order transition. [12,13] In converting the spin problem to a particle one, whether the mass depends on the coordinate is a matter of how to set up the Schrödinger-like equation in the process of particle mapping (see Appendix). In our case all the coordinate dependence is included in the effective particle potential. As we can see below our approach gives the same results as Ref. [14].

The potential $v_t(x)$ has now small and large barriers with same minima. Since the mass is independent of the coordinate it is obvious that the escape over the small barrier dominates. [19] For large spin system, such as $S \sim S + 1 \sim \tilde{S} \equiv S + 1/2$, the top of the small barrier is located at $x_0 = sn^{-1}0 = 0$. By equating both sides of the criterion Eq.(15) and evaluating the the derivatives of the potential $v_t(x)$ at $x = x_0$ we obtain an equation of the phase boundary line between first- and second-order transitions:

$$\alpha_{tc}(b_c) = \frac{1 - 16b_c + 16b_c^2 + \sqrt{1 + 32b_c - 32b_c^2}}{4(1 - 2b_c)}$$

(3.4)

where the subscript $c$ represents that the anisotropy and field parameters are the values taken at the phase boundary. From Eq.(13), the transition temperature at the phase boundary $T_c$ is then obtained to be

$$\frac{T_{tc}}{SA} = \frac{1}{2\pi} \sqrt{\frac{3\alpha_{tc}(b_c)}{2(1 - 2b_c)}},$$

(3.5)
where $\alpha_{tc}(b_c)$ is given in Eq.(19). These results are consistent with those in Ref. [14] if we realize that $\alpha_x = 2(1 - b)x, b = k_t/(1 + k_t)$.

The phase diagram for the case with longitudinal field only based on the present approach has already been obtained by us, [16] so we just quote the results here:

$$\alpha_{tc}(b_c) = 2(1 - b_c)\sqrt{\frac{1 - 2b_c}{1 + b_c}},$$

$$\frac{T_{lc}}{SA} = \frac{\sqrt{3b_c}}{\pi} \sqrt{\frac{1 - b_c}{1 + b_c}}.$$  (3.6)

In Fig.2 we have plotted the phase boundary curves $\alpha_{tc}(b_c), \alpha_{tc}(b_c)$ and the coercive field line $\alpha_{tm} = \alpha_{tm} \equiv 2(1 - b)$. The first-order transition exist below the curves $\alpha_{tc}(b_c)$ and $\alpha_{tc}(b_c)$. In the region between these lines and the coercive field line the second-order transition lies. From these results we observe some important features. First, as the transverse anisotropy $b$ increases both $\alpha_{tc}$ and $\alpha_{lc}$ decrease, and become zero at $b = 1/2$, i.e., the first-order region decreases in both cases. Thus, the biaxial system has smaller first-order region than the uniaxial system. Second, for a given value of $b(< 1/2)$ the longitudinal field case has larger first-order region than the transverse field case. As we will see below these are common in the case with an arbitrarily directed field.

The above features can be understood as follows. In the uniaxial system without transverse field the spin operator commutes with the Hamiltonian, and thus the spin becomes a constant of the motion (i.e., no dynamics). Moreover, in this case the barrier of the effective particle potential becomes infinitely thick so that there is no tunneling, and hence no quantum-classical phase transition. In the presence of the transverse field, however, the uniaxial spin system becomes a dynamical one, and so the tunneling occurs. In the limit of very small transverse field the top of the effective potential barrier becomes flat, which is favorable to the first-order transition. When the transverse field increases, however, the situation becomes unfavorable to the presence of the first-order. Now, in the biaxial system the transverse anisotropy also gives dynamical origin to the spin system, and thus enforces the escape process from the metastable state, which leads to the suppression of the first-order transition. Therefore, the decrease of $\alpha_{tc}(b_c)$ is caused by the transverse anisotropy only, whereas the boundary line $\alpha_{tc}(b_c)$ is affected by both the transverse field and transverse...
anisotropy.

We now consider the case with arbitrarily directed field. In this case the three parameters $b$, $\alpha_x$, $\alpha_z$ should be treated simultaneously, which is not a simple problem. In the present work we will fix one parameter and then compute the phase boundary with the other two parameters. We first calculate the phase boundary lines $\alpha_{zc}(b_c)$’s for several values of $\alpha_x$, and $\alpha_{xc}(b_c)$’s for different values of $\alpha_z$, which are shown in Fig.3. An immediate observation is that the first-order region for a given $\alpha_x$ (or $\alpha_z$) diminishes as $b$ increases, which shows the same trend as the $\alpha_x = 0$ (or $\alpha_z = 0$) case. We also note that the first-order region becomes smaller for increasing values of $\alpha_x$’s (or $\alpha_z$’s). This can be readily explained by the metastability condition in Eq.(6). When $\alpha_x$ and $b$ are given this condition yields the coercive field line for $\alpha_z$ such as $\alpha_{zm}(\alpha_x, b) = 2[(1 - b)^{2/3} - (\alpha_x/2)^{2/3}]^{3/2}$. From this it can be seen that the metastability region, where the escape process can be considered, is shrunk on the whole, which in turn suppresses the first-order transition region.

Next, we compute the phase boundary for a fixed value of $b$. This is the case when a specific sample is prepared in experiment. Here, we take the molecular iron cluster Fe$_8$ which has been studied in the previous experiment. In this case the transverse anisotropy parameter is given by $b = 0.29$. In Fig.4 we draw the phase boundary $\alpha_{zc}(\alpha_{xc})$ and the metastability condition line $\alpha_{zm}(\alpha_{xm})$ for $b = 0.29$. Since the metastability region of the biaxial system is smaller than the uniaxial system we can see both the first- and second-order regions become smaller than the uniaxial system. The point of intersection with $\alpha_x$-axis is larger than that of $\alpha_{zc}(0)$, which reveals that the case with longitudinal field only has larger first-order region than the transverse field only case. An interesting feature in this diagram is that unlike the uniaxial system $\alpha_{zc}$ does not coincides with the coercive field $\alpha_{zm}$ at zero transverse field. It can thus be realized that the first-order region is reduced more than the second-order region. In fact this is anticipated if we look into the Fig.2 which displays that the longitudinal phase boundary line $\alpha_{lc}$ concurs with the coercive field line only at the point $b = 0$, i.e., the uniaxial system without transverse field. As is mentioned above this is the case of no tunneling, and thus we cannot think of any phase transition. Once $b \neq 0$, i.e., in the biaxial system, the two lines are separated such that the second-order transition
region becomes larger.

The crossover temperature at the phase boundary is calculated numerically by using the values on the phase boundary lines in Fig.3 and the formula \( T_c/S_\alpha = (1/2\pi)\sqrt{-u''(x_0)/m} \).

Fig.5 shows \( T_{zc}/S_\alpha \) and \( T_{xc}/S_\alpha \) as a function of \( b_c \) for several values of \( \alpha_x \)’s and \( \alpha_z \)’s, respectively. Since the \( b_c \) at \( \alpha_{zc} = 0 \) decreases as \( \alpha_x \) increases (see Fig.3) the corresponding \( T_{zc}(b_c)/S_\alpha \) ends at smaller value of \( b_c \) for increasing \( \alpha_x \). The same trend can be found in \( T_{xc}(b_c) \). It is noted that for a given \( b_c \) the crossover temperature at the phase boundary, \( T_{zc}(b_c)/S_\alpha \), rises as the transverse field grows, while \( T_{xc}(b_c)/S_\alpha \) is lowered as the longitudinal field increases. By fixing the value of \( b_c \) and using Fig.4 we can investigate how the crossover temperature at the phase boundary varies with the field parameters. For \( b_c = 0.29 \) we have shown a 3d plot of \( T_c(\alpha_{xc}, \alpha_{zc}) \) in Fig.6. From this picture it can be easily seen that \( T_c \) is lowered with increasing \( \alpha_{zc} \), but rises as \( \alpha_{xc} \) increases.

IV. SUMMARY

We have studied the phase transition of the escape rate of a biaxial spin particle with an arbitrarily directed magnetic field. By using the method of particle mapping we have obtained an effective particle potential to which the criterion for the presence of the first-order quantum-classical phase transition developed in Refs. [3,4] has been applied. From this approach we have computed several phase boundary lines depending on the transverse anisotropy and field parameters. In the field vs transverse anisotropy plot, \( \alpha_{xc}(b_c) \) or \( \alpha_{zc}(b_c) \), it is found that the first-order region decreases as \( b_c \) and \( \alpha_{x,z} \) increase. In the case of longitudinal field vs transverse field plot, \( \alpha_{zc}(\alpha_{xc}) \), we have observed that compared with the uniaxial system, both the first- and second-order transition regions are reduced (with more decrease of the first-order region) due to the transverse anisotropy. The crossover temperatures at the phase boundary corresponding to these diagrams have also been obtained. In the 3d plot of \( T_c(\alpha_{xc}, \alpha_{zc}) \) we have observed that \( T_c \) increases with \( \alpha_{zc} \), but decreases with \( \alpha_{zc} \).

Experimentally, the phase diagram of \( \alpha_{zc}(\alpha_{xc}) \) can be found in the octanuclear iron
cluster Fe$_8$ in which $S = 10$ and $b_c = 0.29$. In this case the intersection with the $\alpha_x$-axis in the phase diagram (see Fig.4) is given by $\alpha_{xc} = 0.274$ for which the magnetic field is estimated to be $H_{xc} = 0.64$T, and for the longitudinal field, $\alpha_{zc}(0) = 0.81$ in the diagram for which $H_{zc} = 1.9$T, about three times of $H_{xc}$. The corresponding temperatures are also estimated to be $T_{zc} = 0.39$K which is the upper limit of the $T_{zc}(b_c = 0.29)$’s for different values of $\alpha_x$’s, and $T_{xc} = 0.5$K which determines the lower limit of $T_{xc}(b_c = 0.29)$’s for different values of $\alpha_z$’s.

**APPENDIX A: COORDINATE DEPENDENT MASS**

In converting the spin Hamiltonian of Eq.(1) to an effective particle Hamiltonian, if we set the Schrödinger-like equation to be

$$\left[ -\frac{1}{2m(x)} \frac{d^2}{dx^2} + V(x) \right] \Psi(x) = E \Psi(x) \quad (A1)$$

the coordinate dependent mass $m(x)$, effective particle potential $V(x)$, and the wave function $\Psi(x)$ are derived as following:

$$m(x) = \frac{1}{2A(1 + b \sinh^2 x)} \quad (A2)$$

$$V(x) = \frac{A}{4(1 + b \sinh^2 x)} \times \left[ \alpha_z^2 S^2 - 4b(S^2 + S - 1/2) - 4\alpha_x S(S + 1/2) \cosh x \right. \left. + b^2 \sinh^4 x - \{b^2 + 4b(S^2 + S - 1) - \alpha_x^2 S^2\} \sinh^2 x \right. \left. - \{4b\alpha_z S(S + 1/2) \cosh x + 2\alpha_x \alpha_z S^2\} \sinh x \right], \quad (A3)$$

$$\Psi(x) = \left(1 + b \sinh^2 x\right)^{\frac{S-1}{2}} \times \exp \left[ -\frac{\alpha_z S}{2\sqrt{1-b}} \tanh^{-1} \left( \sqrt{1-b} \tanh x \right) \right] \times \exp \left[ -\frac{\alpha_x S}{2\sqrt{b(1-b)}} \tan^{-1} \left( \sqrt{\frac{b}{1-b}} \cosh x \right) \right] \Phi(x) \quad (A4)$$
with
\[ \Phi(x) = \sum_{\sigma=-S}^{S} \frac{C_{\sigma}}{(S + \sigma)!(S - \sigma)!} (\sinh x + \cosh x)^{\sigma}. \]  

(A5)

In this case, since the mass depends on coordinate the criterion for the first-order transition becomes more complicated. Following Ref. [6] the general first-order transition condition which includes the coordinate-dependent mass is given by

\[ [V''''(x_0)(g_1 + g_2/2) + \frac{1}{8}V''''(x_0) + m'(x_0)\omega_0^2 g_2 \\
+ m'(x_0)\omega_0^2(g_1 + g_2/2) + \frac{1}{4}m''(x_0)\omega_0^2] < 0, \]

(A6)

where

\[ g_1(\omega_0) = -\frac{\omega_0^2 m'(x_0) + V'''(x_0)}{4V''(x_0)}, \]
\[ g_2(\omega_0) = -\frac{2m'(x_0) + V'''(x_0)}{4[m(x_0)\omega_0^2 + V''(x_0)]}, \]

(A7)

and \( \omega_0^2 \) is the sphaleron oscillation defined as \( \omega_0^2 = -V''(x_0)/m(x_0) \) from Eq.(15). Calculating the derivatives of \( m(x) \) and \( V(x) \) and substituting these into above condition we have same results as the constant mass case.
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[18] Comparing with the model in Ref. [9] we see $\alpha_{x,z} = 2\tilde{h}_{x,z}$ in the large spin limit.

[19] It has also been proved in Ref. [12] that the small barrier escaping process always dominates even when the mass depends on the coordinate.
FIGURES

FIG. 1. The effective particle potential with \( b = 0.3, \alpha_x = 0.2, \alpha_z = 0.3 \). The local minimum at \( x = -x_m \) corresponds to a metastable state of the spin system described in Eq. (1). The inversion process of a spin magnetization vector takes place by the escape from the local minimum to global minimum at \( x = x_m \) along the path which passes through the small barrier.

FIG. 2. The phase boundary lines \( \alpha_{tc}(b_c) \) and \( \alpha_{lc}(b_c) \), and the coercive field line \( \alpha_{tm} = \alpha_{lm} \equiv 2(1 - b) \).

FIG. 3. The phase boundary lines (a) \( \alpha_{zc}(b_c)'s \) and (b) \( \alpha_{xc}(b_c)'s \) for several values of \( \alpha_x 's \) and \( \alpha_z 's \), respectively.

FIG. 4. Phase diagram for \( b = 0.29 \) which is the case of the iron cluster Fe_8. The solid line is the phase boundary \( \alpha_{zc}(\alpha_{xc}) \), and the dashed line corresponds to the line of metastability condition for \( b = 0.29 \).

FIG. 5. Crossover temperatures at the phase boundary as a function of \( b_c \): (a) \( T_{lc}/\tilde{S}A \)'s for \( \alpha_x = 0, 0.1, 0.3 \); (b) \( T_{tc}/\tilde{S}A \)'s for \( \alpha_z = 0, 0.3, 0.7 \).

FIG. 6. 3d plot of the crossover temperature \( T_c/\tilde{S}A \) at the phase boundary as a function of field parameters \( \alpha_{xc} \) and \( \alpha_{zc} \).
Fig. 1

\[ V(x) \]

\[ b = 0.3 \]
\[ \alpha_x = 0.2 \]
\[ \alpha_z = 0.3 \]
Fig. 2
Fig. 3a
Fig. 3b
Fig. 4
Fig. 5a
Fig. 6