Quantum-classical crossover for biaxial antiferromagnetic particles with a magnetic field along the hard axis

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Quantum-classical crossover of the escape rate is studied for biaxial antiferromagnetic particles with a magnetic field along the hard axis. The phase boundary line between first- and second-order transitions is calculated, and the phase diagrams are presented. Comparing with the results of different directed fields, the qualitative behavior of the phase diagram for the magnetic field along the hard axis is different from the case of the field along the medium axis. For the hard axis the phase boundary lines $k(y)$ shift downwards with increasing $h$, but upwards for the medium axis. It is shown that the magnetic field along the hard axis favors the occurrence of the first-order transition in the range of parameters under the certain constraint condition. The results can be tested experimentally for molecular magnets Fe$_8$ and Fe$_4$.

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Quantum spin tunneling at low temperatures has attracted considerable attention in view of a possible experimental test of the tunneling effect for mesoscopic single-domain particles. Up until now, molecular magnets are the most promising candidates to observe macroscopic quantum coherence.\textsuperscript{1} When the temperature increases the thermal hopping becomes dominant beyond some crossover temperatures. Since the first- and second-order transitions between the quantum and classical behaviors of the escape rates in spin systems were introduced by Chudnovsky and Garanin,\textsuperscript{2,3} a lot of work has been done theoretically and experimentally.\textsuperscript{4–14} Most theoretical studies focus on ferromagnetic particles. It is shown that quantum tunneling occurs at higher temperatures and higher frequencies in antiferromagnetic particles. It is shown that quantum tunneling occurs in the presence of an external magnetic field along the hard axis for molecular magnet Fe$_8$, and is actually ferrimagnetic, in the study of the quantum-classical crossover of biaxial antiferromagnetic particles, the case of a magnetic field along the hard axis deserves further investigation. In this paper we attempt to investigate the effect of a magnetic field along the hard axis on the quantum-classical crossover behavior of the escape rate of biaxial antiferromagnetic particles. Comparing with the results of different directed fields, the qualitative behavior of the phase diagram for the magnetic field along the hard axis is different from the case of the field along the medium axis. For the hard axis the phase boundary lines $k(y)$ shift downwards with increasing $h$, but upwards for the medium axis. It is shown that the magnetic field along the hard axis favors the occurrence of the first-order transition in the range of parameters under the certain constraint condition.

We consider a small biaxial antiferromagnetic particle with two magnetic sublattices whose magnetizations, $\mathbf{m}_1$ and $\mathbf{m}_2$, are coupled by the strong exchange interaction $\mathbf{m}_1 \cdot \mathbf{m}_2 / \chi_s$, where $\chi_s$ is the perpendicular susceptibility. The system has a noncompensation of sublattices with $m(=m_1 - m_2>0)$, with the easy axis $x$, the medium axis $y$, and the hard axis $z$. In the presence of a magnetic field along the hard axis, the Euclidean action is written as\textsuperscript{15}

$$S_E(\theta, \phi) = V \int d\tau \left( \frac{m_1 + m_2}{\gamma} \dot{\phi} - i \frac{m}{\gamma} \cos \theta ight. + \frac{\chi_s}{2\gamma^2} \left[ \dot{\theta}^2 + (\dot{\phi} - i \gamma H)^2 \sin^2 \theta \right] + K_1 \cos^2 \theta + K_2 \sin^2 \theta \sin^2 \phi - mH \cos \theta \right),$$

where $V$ is the volume of the particle, $\gamma$ the gyromagnetic ratio, and $\chi_s = \chi_s(m_2/m_1)$. $K_1$ and $K_2(K_1 > K_2)$ are the transverse and longitudinal anisotropic coefficients, respectively. The polar coordinate $\theta$ and the azimuthal coordinate $\phi$ for the angular components of $\mathbf{m}_1$ in the spherical coordinate system determine the direction of the Néel vector. A dot over a symbol denotes a derivative with respect to the Euclidean time $\tau$. The classical trajectory to the Euclidean action (1) is determined by
where \( n=m/(K_1 \gamma) \), \( x=x_1 / (K_1 \gamma^2) \), \( k=K_2 / K_1 \), \( b=\gamma H \), \( h=H/H_c \), and \( H_c=2K_2/m \).

In the high-temperature regime the sphaleron solution of Eqs. (2) and (3) is \((\theta_0, \phi_0, \eta_0(\tau/2))\), where \( h_0=\gamma h/(1-k+xb^2/2) \approx 1 \). The crossover behavior of the escape rate from the quantum tunneling to the thermal activation is obtained from the deviation of the period of the periodic instanton from that of the sphaleron.\(^{18,24}\) To this end we expand \((\theta, \phi)\) about the sphaleron configurations \( \theta_0 \) and \( \phi_0 \), i.e., \( \theta=\theta_0+\eta(\tau) \) and \( \phi=\phi_0+\xi(\tau) \), where \( \phi_0=\pi/2 \). Denote \( \delta\Omega(\tau+\beta h)=\delta\Omega(\tau) \). Thus it can be expanded in the Fourier series \( \delta\Omega(\tau)=\sum_{\omega_n} \delta\Omega_n \exp[i\omega_n \tau] \), where \( \omega_n=2\pi n/\beta h \). To the lowest order \( \eta=ia\theta_1 \sin(\omega \tau) \) and \( \xi=a\phi_1 \cos(\omega \tau) \). Here, \( a \) serves as a perturbation parameter. Substituting them into Eqs. (2) and (3), and neglecting the terms of order higher than \( a \), we obtain the relation

\[
\frac{\dot{\Theta}}{\Theta} = \frac{x \omega_2^2 + \sin^2\Theta(2-2k+x^2)}{\omega_n(n \sin \Theta - bx \sin 2\Theta)} = -\frac{\omega_n(2bx \cos \Theta)}{\sin \Theta(\omega_n^2 - 2k)},
\]

and the oscillation frequency

\[
\omega^2 = -\frac{1}{2x^2}[x \sin^2\Theta(2-2k+x^2) - 2k] + (n - 2bx \cos \Theta)^2
\]

\[
\pm \frac{1}{2x^2}[8kx^2(2-2k+x^2) \sin^2 \Theta_0
\]

\[
+ [x(\sin^2 \Theta_0(2-2k+x^2) - 2k) + (n - 2bx \cos \Theta_0)^2]^{1/2}.
\]

Next, let us write \( \eta=ia\theta_1 \sin(\omega \tau) \) and \( \xi=a\phi_1 \cos(\omega \tau) + i\phi_2 \), where \( \eta_2 \) and \( \xi_2 \) are of the order of \( a^2 \). Inserting them into Eqs. (2) and (3), we arrive at \( \omega = \omega_4 \) and

\[
\eta_2 = a^3 \phi_1 \sin(3\omega \tau), \quad \xi_3 = a^3 \phi_1 \cos(3\omega \tau),
\]

where the analytic forms of real coefficients \( \phi_1 \) and \( \phi_2 \) are cumbersome, and are not listed in the paper. We also have

\[
n^4y^2(\omega^2 - \omega_1^2)(\omega^2 - \omega_2^2) = a^2 \frac{\phi_1^2}{4} g(h,k,y),
\]

where

\[
g(h,k,y) = g_1(h,k,y) + g_2(h,k,y) \quad \text{and} \quad y=x/n(=x_1 / K_1/m^2).
\]

The forms of \( g_1(h,k,y) \) and \( g_2(h,k,y) \) are given by

\[
g_1(h,k,y) = (1 - 4hky \cos \Theta_0)w\left[ \frac{2}{\lambda^2} hkyw \cos \Theta_0
\right.
\]

\[
+ \frac{\sin \theta_0}{\lambda} \left[ k - \frac{5}{2}yw^2 \right]
\]

\[
- 4\tilde{q}_2 k \cos \Theta_0 - 8(\tilde{p}_0 - \tilde{p}_2)hkyw \sin \Theta_0
\]

\[
+ 4w \left[ (\tilde{p}_0 - \tilde{p}_2)k \cos \Theta_0 - k \sin \Theta_0
\right.
\]

\[
- \left( \tilde{p}_0 - \frac{3}{2}\tilde{p}_2 \right) y^2w \cos \Theta_0 \right],
\]

\[
g_2(h,k,y) = -\frac{n - 4hky \cos \Theta_0}{\sin \Theta_0}w\left[ \frac{1}{\lambda^2}[-hky \cos \Theta_0
\right.
\]

\[
+ 4(1-k-2h^2/2y^2)\cos \Theta_0)
\]

\[
+ \frac{12w}{\lambda^2} \left( \frac{1}{8} \sin \Theta_0 - hky \sin \Theta_0 \right)
\]

\[
+ \frac{4}{\lambda} \left[ \frac{1}{2} k + 4hky\tilde{q}_2 - \frac{3}{4} yw^2 \right] \cos \Theta_0
\]

\[
+ 2\tilde{p}_0 - \tilde{p}_2)hky \sin \Theta_0 - \tilde{q}_2 y^2w \cos \Theta_0
\]

\[
- 2(\tilde{p}_0 - \tilde{p}_2) \sin \Theta_0 \left[ 1 - k + 2h^2/2y^2 \right]
\]

\[
+ 4(k - y^2\tilde{q}_2) \sin \Theta_0 + 2(\tilde{p}_0 - \tilde{p}_2)w \cos \Theta_0
\]

\[
- 8(\tilde{p}_0 - \tilde{p}_2)hkyw \cos \Theta_0 \right],
\]

where \( w=n\omega_4 \) and \( \lambda=\phi_1/\Theta_0 \). The parameter \( y=x/n^2 \) (=\( x_1 K_1/m^2 \)) indicates the relative magnitude of the noncompensation. For a large noncompensation \((y \ll 1, \text{i.e., } m \gg \sqrt{\chi K_1})\) the system becomes ferromagnetic, while for a small noncompensation \((y \gg 1, \text{i.e., } m \ll \sqrt{\chi K_1})\) the system becomes nearly compensated antiferromagnetic.\(^{18}\) Also, \( \tilde{p}_0, \tilde{p}_2, \) and \( \tilde{q}_2 \) are obtained by replacing \( \Theta_1 \) by \( \phi_1/\lambda \) and dropping \( \phi_1 \) in \( p_0, p_2, \) and \( q_2 \), respectively. It is shown that for \( h=0 \), Eq. (10) is reduced to Eq. (17) in Ref. 18.

According to the theory by Chudnovsky,\(^{23}\) the order of quantum-classical crossover is determined by the behavior of the Euclidean time oscillation period \( \tau(E) \), where \( E \) is the energy near the bottom of the Euclidean potential. The existence of a minimum in the oscillation period with respect to
We discuss the molecular magnet Fe$_8$, which is actually ferromagnetic, and thereby $y$ should be taken into account in the biaxial symmetry. Take the measured value of the anisotropy parameter, e.g., $k=0.728$ for Fe$_8$. The phase boundary line $h(y)$ for $k=0.728$ is shown in Fig. 3 (dashed line). For Fe$_8$ the maximum of the critical value $y_c$, beyond which the first-order transition vanishes, is about 0.313. Next we consider molecular magnet Fe$_4$(OCH$_3$)$_4$(dpm)$_y$ Fe$_4$ for simplicity, as another example. The cluster Fe$_4$ is characterized by an $S=5$ ground state arising from antiferromagnetic interaction between central and peripheral iron spins.$^{25-28}$ According to the estimate of the transverse and longitudinal anisotropic coefficients for Fe$_4$ in Refs. 25–28, $k=0.822$. The phase boundary line $h(y)$ for $k=0.822$ is also shown in Fig. 3 (solid line). For Fe$_4$ the maximum of the critical value $y_c$ is about 0.378.

In conclusion, the quantum-classical crossover of the escape rate for biaxial antiferromagnetic particles is investigated in the presence of a magnetic field along the hard axis. The nonlinear perturbation method is applied to establish the phase diagrams for first- and second-order transitions. Comparing with the results of different directed fields, it shows that in the range of parameters under the constraint condition $h_0 \leq 1$, the magnetic field along the hard axis favors the occurrence of the first-order transition. For instance, in the case of $h=0.1$ and $k=0.85$ the first-order transition vanishes beyond $y=0.138$, 0.261, and 0.335 for the field along the medium axis, easy axis, and hard axis, respectively. To illustrate the above results with concrete examples, firstly we discuss the molecular magnet Fe$_8$, which is actually ferrimagnetic, and thereby $y$ should be taken into account in the biaxial symmetry. Take the measured value of the anisotropy parameter, e.g., $k=0.728$ for Fe$_8$. The phase boundary line $h(y)$ for $k=0.728$ is shown in Fig. 3 (dashed line). For Fe$_8$ the maximum of the critical value $y_c$, beyond which the first-order transition vanishes, is about 0.313. Next we consider molecular magnet Fe$_4$(OCH$_3$)$_4$(dpm)$_y$ Fe$_4$ for simplicity, as another example. The cluster Fe$_4$ is characterized by an $S=5$ ground state arising from antiferromagnetic interaction between central and peripheral iron spins.$^{25-28}$ According to the estimate of the transverse and longitudinal anisotropic coefficients for Fe$_4$ in Refs. 25–28, $k=0.822$. The phase boundary line $h(y)$ for $k=0.822$ is also shown in Fig. 3 (solid line). For Fe$_4$ the maximum of the critical value $y_c$ is about 0.378.

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\[ p_2 = -\left\{ \frac{h(k - 2x\omega^2)}{2} \sin \theta_0 + 2b x \omega (k \theta_1 \phi_1 + n \omega \theta_1^2 \sin \theta_0 - 2x \omega^2 \theta_1 \phi_1) + 2b x^2 \omega^2 (4\omega^2 - 3\omega_0^2) \theta_1 \phi_1 \cos^2 \theta_0 \\
+ k^2 (2\theta_1^2 + \phi_1^2) \sin 2\theta_0 - n k \omega \theta_1 \phi_1 \cos \theta_0 - k(2 + b^2 x + 4x^2) \phi_1^2 \sin 2\theta_0 - \frac{1}{2} k x \phi_1^2 (4\omega^2 - \omega_0^2) \sin 2\theta_0 \\
+ 4x \omega^2 \phi_1^2 \sin 2\theta_0 - n x \omega \theta_1 \phi_1 (2\omega^2 - 3\omega_0^2) \cos \theta_0 - x \omega^2 \omega_0^2 \phi_1^2 \sin 2\theta_0 \right\} \left\{ -4(h \cos \theta_0 + \cos 2\theta_0)k^2 + 2k[4hx \omega^2 \cos \theta_0 \\
+ 2 \cos^2 \theta_0(2 + b^2 x + 4x^2) - 2 - b^2 x - 8x \omega^2 + 4\omega^2 [n^2 - 4nbh \cos \theta_0 + 2x + b^2 x^2 + 4x^2 \omega^2 - 2 \cos^2 \theta_0(2 - b^2 x)] \right\}, \]

\[ q_2 = \{ 2 \theta_1 \omega [2nbx \omega \phi_1 + \theta_1(2 - 2k + b^2 x + 4x^2) \omega \phi_1 \sin \theta_0 + h k \omega \theta_1 \sin \theta_0] - 2 \theta_1 \phi_1 \cos^3 \theta_0 \omega^2 \omega_0^2 + 2k(2 + b^2 x + 3x^2 \omega^2) + 6x \omega_0^2 \\
+ 2b^2 x( -8\omega^2 + 3\omega_0^2) - 2 \cos^2 \theta_0 \omega [h k \theta_1 \phi_1(2k - 3x^2 \omega^2) + 2b x \omega (-2 + 2k - b^2 x) \phi_1] \sin \theta_0 + 3n \omega \theta_1 \phi_1 (2k + x \omega_0^2) \phi_1^2 \sin \theta_0 \right\} \\
+ \cos \theta_0 \omega [4k \theta_1 \phi_1 + 2n^2 \omega^2 \theta_1 \phi_1 - 2n \omega \sin \theta_0(2 + b^2 x) \phi_1^2 - x \omega^2 \omega_0^2 \phi_1^2 + x \theta_1 \phi_1 (6[1 + 2x \omega^2]) \omega^2 + b x \omega (-8\omega^2 + 3\omega_0^2) ] \\
+ 2k(4n \omega \theta_1^2 \sin \theta_0 + 2n \omega \phi_1^2 \sin \theta_0 - \theta_1 \phi_1 (2 + b^2 x + 4x^2 \omega^2 + 3x \omega_0^2)) \right\} \left\{ 4 \sin \theta_0[2 h \cos \theta_0 + \cos 2\theta_0]k^2 \\
+ k(8x \omega^2 \sin^2 \theta_0 - 4hx \omega^2 \cos \theta_0 - 2\theta_0(2 + b^2 x)) - 2\omega^2 [n^2 - 4nbh \cos \theta_0 + 2x + b^2 x^2 + 4x^2 \omega^2 - 2 \cos^2 \theta_0(2 - b^2 x)] \right\}. \]