Tunnel splitting in biaxial spin models investigated with spin-coherent-state path integrals

Zhi-De Chen,[1] J.-Q. Liang,[2] and F.-C. Pu[1,3]

[1]*Department of Physics and Institute of Modern Condensed Matter Physics, Guangzhou University, Guangzhou 510405, China*

[2]*Institute of Theoretical Physics and Department of Physics, Shanxi University, Taiyuan, Shanxi 030006, China*

[3]*Institute of Physics and Center for Condensed Matter Physics, Chinese Academy of Sciences, Beijing 100080, China*

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Tunnel splitting in biaxial spin models is investigated with a full evaluation of the fluctuation functional integrals of the Euclidean kernel in the framework of spin-coherent-state path integrals which leads to a magnitude of tunnel splitting quantitatively comparable with the numerical results in terms of diagonalization of the Hamilton operator. An additional factor resulted from a global time transformation converting the position-dependent mass to a constant one seems to be equivalent to the semiclassical correction of the Lagrangian proposed by Enz and Schilling. A long standing question whether the spin-coherent-state representation of path integrals can result in an accurate tunnel splitting is therefore resolved.

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I. INTRODUCTION

Quantum tunneling in mesoscopic systems called the macroscopic quantum tunneling (MQT) has attracted considerable attentions in recent years[1–10] since the MQT may help to establish a bridge between the quantum and classical realms in physics and might also be of significance for information storage in the future.[2,9] Experimental realization of the macroscopic quantum phenomena is extremely difficult and up to now, magnetic molecular clusters have been the most promising candidates to observe the phenomenon, for instance, the predicted macroscopic quantum coherence (MQC) i.e., the coherent tunneling between degenerate ground states in an octanuclear iron oxohydroxo cluster Fe_8 with a biaxial anisotropy[10] which has subsequently triggered active theoretical researches. However, most of the investigations[7,11–13] are concentrated on the quantum phase interference and little work has been done toward the evaluation of the pre-exponential factor of the tunnel splitting which gives rise to an accurate magnitude of the splitting. The tunnel splitting can be obtained from the transition amplitude between degenerate vacua which has a Euclidean path-integral representation called the instanton method. In the present paper we provide a systematic study of the functional fluctuation of the Euclidean kernel for the quantum tunneling in spin systems in the framework of spin-coherent-state path integral which to our knowledge has not been given in literature. Theoretical results of the pre-exponential factor of tunnel splitting are justified by comparing with the numerical diagonalization of Hamilton operator of the spin system considered.

Theoretically, tunnel splitting of the ground states is calculated by the instanton method developed originally for a particle moving in a double-well potential.[14,15] Tunnel splitting in a spin system has been investigated in terms of the instanton method by either a straightforward application of spin-coherent-state path integrals[7,8] to map the spin system on to a classical Lagrangian of angle variables of spin vector or other approaches[3,11,13,17] to map the spin system on to a particle (actually a rigid rotor) in a potential field. In the present paper, we show that, for tunnel splitting in the biaxial model, the spin-coherent-state path integral is equivalent to the traditional mapping technique[3,11,13] up to the one loop approximation except for a factor $(2s+1)/(2s)$ which can be omitted in the large spin approximation.

The tunnel splitting in biaxial spin model was first studied by Enz and Schilling[17,18] in the large spin approximation for which a standard way in coherent-state representation of path integral is that the expectation value of the Hamilton operator is replaced by a classical Hamiltonian (or Lagrangian) regarding the spin as a classical vector. They, however, "demonstrated the failure of the coherent-state representation of path integral for spin systems with a large spin s" and used a semiclassical Hamiltonian (or semiclassical Lagrangian) containing the first two leading terms of order-$1/s$ to obtain a better level splitting.[17] A significant modification from the semiclassical Hamiltonian is that a factor s in the classical action is replaced by the factor $\sqrt{s(s+1)}\simeq s+1/2$. Although the application of the instanton method to spin-coherent-state path integral was reconsidered by Garg and Kim[8] with much details, it was believed that spin-coherent-state path integral can only provide a qualitative description for the tunneling process, but cannot "yield answers that are correct beyond the leading exponential order."[19] The bad reputation of spin-coherent-state path integral in the application to calculate the magnitude of quantum tunneling in spin systems is obviously because of the observation that the tunnel splitting resulted from the spin-coherent-state representation of path integral by means of the classical Hamiltonian "strongly deviates from the exact one following from numerical diagonalization."[17] We argue that the deviation is actually due to the method used for evaluation of the pre-exponential factor of tunnel splitting which is suitable only for a Lagrangian of particle with a constant-mass,[17,18,20] however, is not adequate to the mapped particle in terms of the spin-coherent-state path integral which has a position-dependent mass.[11] An accurate magnitude of tunnel splitting can be obtained if fluctuation functional integrals are carefully manipulated taking the position-dependent mass into account while using the classical Hamiltonian as in the standard way. We demonstrate our formalism, which results in a satisfactory magnitude of level splitting beyond the leading exponential approximation as well as the level space in comparison with the numerical result by diagonalization of the Hamilton operator, for the case of large spin approximation in which the classical Hamiltonian works well. The organization of the paper is as follows. In Sec. II, we present a brief review on the instanton method for tunnel splitting in biaxial spin model in the framework of spin-coherent-state path integrals. Calculations of the fluctuation functional integral of the tunneling kernel with a great detail are presented in Sec. III. A conclusion is given in Sec. IV.

II. GENERAL FORMULATION OF INSTANTON METHOD FOR TUNNEL SPLITTING

For evaluation of the transition amplitude induced by quantum tunneling in spin systems, a customary procedure is to evaluate the imaginary time Feynman kernel from an initial state $|\mathbf{n}_1\rangle$ to a final state $|\mathbf{n}_2\rangle$ with the help of spin-coherent-state path integrals[5–7]

$$\mathcal{K}=\langle \mathbf{n}_2|e^{-\hat{H}t/\hbar}|\mathbf{n}_1\rangle=\int [d\mathbf{n}]e^{-(1/\hbar)S_e[\mathbf{n}]}, \tag{1}$$

where $S_e[\mathbf{n}]=\int_{-T/2}^{T/2}L_e[\mathbf{n}]d\tau$, $|\mathbf{n}\rangle$ denotes the spin-coherent state which is the eigenstate of spin operator $\hat{S}\cdot\mathbf{n}$ such that $\hat{S}\cdot\mathbf{n}|\mathbf{n}\rangle=s|\mathbf{n}\rangle$, $\mathbf{n}=(\sin\theta\cos\phi,\sin\theta\sin\phi,\cos\theta)$ is an arbitrary unit vector with a polar angle θ and an azimuthal angle ϕ and s is the total spin of the system. L_e is the Euclidean Lagrangian given by

$$L_e[\mathbf{n}]=i\hbar(1-\cos\theta)\dot{\phi}(\tau)+H(\theta,\phi), \tag{2}$$

where $H(\theta,\phi)=\langle\mathbf{n}|\hat{H}|\mathbf{n}\rangle$, and $\hat{H}$ is the Hamilton operator of the spin system. In the semiclassical approximation,[3,8] the spin is treated as a classical vector, thus the $H(\theta,\phi)$ is replaced with the classical anisotropy energy $E(\theta,\phi)$. In the following, we shall evaluate the Feynman kernel Eq. (1) in the large spin approximation.[11] For the sake of simplicity, we set $\hbar=1$ from now on.

The dominant contribution to the Feynman kernel Eq. (1) is from the classical path which is governed by the dynamic equation[3,7,8]

$$-is\sin\theta\dot{\theta}=\frac{\partial E(\theta,\phi)}{\partial\phi}\equiv E_\phi(\theta,\phi), \tag{3a}$$

$$is\sin\theta\dot{\phi}=\frac{\partial E(\theta,\phi)}{\partial\theta}\equiv E_\theta(\theta,\phi). \tag{3b}$$

The solutions of the above equation $[\theta_c(\tau),\phi_c(\tau)]$ could be instanton,[7] bounce,[16] or periodic instanton[21] depending on the boundary condition of the problem. Substituting $[\theta_c(\tau),\phi_c(\tau)]$ back into the Lagrangian (2), one can find the classical action

$$S_{\rm cl}=\int_{-T/2}^{T/2}[is(\cos\theta_c-1)\dot{\phi}_c-E(\theta_c,\phi_c)]d\tau. \tag{4}$$

As a matter of fact, the dynamic equation (3) implies the energy conservation along the classical path

$$\frac{d}{d\tau}E(\theta,\phi)=E_\phi(\theta,\phi)\dot{\phi}+E_\theta(\theta,\phi)\dot{\theta}=0, \tag{5}$$

namely, $E(\theta_c,\phi_c)=E_0$ with E_0 denoting the energy constant. Hence the classical action can be calculated from the energy conservation Eq. (5) with $\cos\theta$ expressed as a function of the angle ϕ. To explain the procedure explicitly we consider the well studied biaxial spin model with a field applied along the hard axis which results in the effect of quantum phase interference of tunneling paths.

The Hamilton operator of the spin system is given by[7,13]

$$\hat{H}=K_1\hat{S}_z^2+K_2\hat{S}_y^2-\alpha\hat{S}_z, \tag{6}$$

where $K_1>K_2>0$, $\alpha=g\mu_B h$, h is the magnitude of the applied field along the hard axis, and K_1, K_2 are the anisotropy constants. μ_B is the Bohr magneton and g is the spin g factor which is taken to be 2 here. Up to a constant, we obtain the classical energy seen to be

$$E(\theta,\phi)=K_1s^2(\cos\theta-u)^2+K_2s^2\sin^2\phi\sin^2\theta, \tag{7}$$

where $u=\alpha/(2K_1s)$. This implies two degenerate ground states located at $(\theta=\theta_0,\phi=0)$ and $(\theta=\theta_0,\phi=\pi)$ with $\theta_0=\cos^{-1}u$. The dynamic equation which gives rise to the classical path is now given by

$$i\dot{\phi}=-2K_1s(1-\lambda\sin^2\phi)\cos\theta+2K_1su, \tag{8a}$$

$$-i\dot{\theta}=2K_2s\sin\phi\cos\phi\sin\theta, \tag{8b}$$

where $\lambda=K_2/K_1$. For instanton solution dominating the tunneling at the ground state we can set $E_0=0$ and obtain the following relation from Eq. (7):

$$\cos\theta_c=\frac{u\pm i\lambda^{1/2}\sin\phi_c(1-u^2-\lambda\sin^2\phi_c)^{1/2}}{1-\lambda\sin^2\phi_c}, \tag{9}$$

where the subscript "c" denotes the classical solution, i.e., the instanton configuration. Using the above relation (9) the classical action is found to be

$$S_{cl}^{\pm}=is\int_0^{\pm\pi}(\cos\theta_c-1)d\phi_c=S_c\mp i\theta_s, \tag{10}$$

where

$$S_c=2s\,\text{arctanh}\sqrt{\frac{\lambda}{1-u^2}}-2s\sqrt{\frac{u^2}{1-\lambda}}\,\text{arctanh}\sqrt{\frac{u^2\lambda}{(1-u^2)(1-\lambda)}}, \tag{11}$$

and

$$\theta_s=s\pi-\frac{\alpha\pi}{2K_1\sqrt{1-\lambda}}. \tag{12}$$

Substituting Eq. (9) back to Eq. (8a), the explicit instanton trajectory is obtained as

$$\phi_c = \pm \cos^{-1} \frac{\sqrt{1-\lambda_h}\tanh(\omega_h \tau)}{\sqrt{1-\lambda_h \tanh^2(\omega_h \tau)}}, \tag{13}$$

where

$$\omega_h = 2s\sqrt{K_1 K_2 (1-u^2)}, \quad \lambda_h = \frac{\lambda}{1-u^2}. \tag{14}$$

The sign $\pm$ represents two symmetric tunneling paths of opposite windings (i.e., instanton and anti-instanton).[7] The Feynman kernel then can be found by summing up the multi-instanton contributions according to Refs. 5,14

$$\mathcal{K} \simeq e^{-\varepsilon_0 T} \sum_{m,l>0}^{m+l \mathrm{odd}} \frac{(BT)^{l+m}}{m!\,l!} e^{i\theta_s(m-l)} e^{-S_c(m+l)}$$
$$= e^{-\varepsilon_0 T} \sinh(2Be^{-S_c}\cos\theta_s T), \tag{15}$$

where B comes from fluctuation around the classical path and $\epsilon_0 = \omega/2$ is the zero-point energy. From Eq. (15), one can read off the tunnel splitting[5,14]

$$\Delta E = 4Be^{-S_c}|\cos\theta_s|. \tag{16}$$

The factor $|\cos\theta_s|$ indicates the well known effect of the quantum phase interference induced by the magnetic field along the hard axis of the biaxial model.[7,10,11,13] The destructive interference (whenever $\theta_s = 2n\pi + \pi/2$) between two symmetric tunneling paths of opposite windings leads to the quenching of tunnel splitting which is not related to Kramers' degeneracy[7] since the external magnetic field breaks the time reversal symmetry of the spin system. The effect of quantum phase interference has been well studied. However, little attention has been paid to the prefactor B which gives rise to a quantitative magnitude of level splitting. The main goal of the present paper is to study the prefactor B for which a systematic formulation is still lacking.

III. EVALUATION OF THE PREFACTOR B

The prefactor B is resulted from the fluctuation integrals around the classical path characterized by θ_c and ϕ_c. We begin with evaluation of the fluctuation functional integral.

A. Fluctuation integral

We consider small fluctuations (η_1, η_2) around the classical path

$$\theta = \theta_c + \eta_1, \quad \phi = \phi_c + \eta_2, \tag{17}$$

with fixed end points such that

$$\eta_1(\pm T/2) = \eta_2(\pm T/2) = 0. \tag{18}$$

Up to one loop approximation, the one-instanton contribution to imaginary time transition-amplitude is given by

$$\mathcal{K}_1 = Ie^{-S_{\mathrm{cl}}}, \tag{19}$$

where I denotes the fluctuation functional integral

$$I = \mathcal{N}\int d\eta_1 d\eta_2 e^{-(1/2)\int_{-T/2}^{T/2}\eta_i M_{ij}\eta_j d\tau}, \tag{20}$$

$\mathcal{N}$ is a normalization factor to match the integral measure $d[\eta]$,[8]

$$\mathcal{N} = \frac{2s+1}{4\pi}\lim_{n\to\infty}\prod_{k=1}^{n} \sin\theta(\tau_k) \simeq \frac{2s+1}{4\pi}\lim_{n\to\infty}\prod_{k=1}^{n} \sin\theta_c(\tau_k) \tag{21}$$

and

$$\eta_i M_{ij}\eta_j = [\eta_1 \quad \eta_2]\begin{bmatrix} M_{11} & M_{12} \\ M_{21} & M_{22}\end{bmatrix}\begin{bmatrix}\eta_1 \\ \eta_2\end{bmatrix}. \tag{22}$$

A general expression of the matrix element M_{ij} can be found by taking the second order functional derivatives of L_e with respect to (θ,ϕ),

$$M_{ij} = \left(\frac{\partial L_e}{\partial x_i} - \frac{d}{d\tau}\frac{\partial L_e}{\partial \dot{x}_i}\right)\left(\frac{\partial L_e}{\partial x_j} - \frac{\partial L_e}{\partial \dot{x}_j}\frac{d}{d\tau}\right), \tag{23}$$

where $x_{1,2} = (\theta,\phi)$. With the help of Eq. (3), we can write it out explicitly

$$M_{11} = E_{\theta\theta}(\theta_c,\phi_c) - \cot\theta_c E_\theta(\theta_c,\phi_c), \quad M_{12} = m_{12} + l_{12}\frac{d}{d\tau},$$

$$M_{21} = m_{21} + l_{21}\frac{d}{d\tau}, \quad M_{22} = E_{\phi\phi}(\theta_c,\phi_c), \tag{24}$$

where

$$m_{12} = E_{\theta\phi}(\theta_c,\phi_c), \quad l_{12} = -l_{21} = is\sin\theta_c,$$
$$m_{21} = E_{\phi\theta}(\theta_c,\phi_c) - \cot\theta_c E_\phi(\theta_c,\phi_c). \tag{25}$$

Noting the equality $E_{\theta\phi}(\theta_c,\phi_c) = E_{\phi\theta}(\theta_c,\phi_c)$ and employing the condition (18) of the fixed end point, one can easily verify the following relation:

$$\int_{-T/2}^{T/2}\eta_1 M_{12}\eta_2 d\tau = \int_{-T/2}^{T/2}\eta_2 M_{21}\eta_1 d\tau, \tag{26}$$

which implies that matrix M is Hermitian for a given energy function $E(\theta,\phi)$.

On the other hand, taking time derivative of Eq. (3), one can find

$$\begin{bmatrix} M_{11} & M_{12} \\ M_{21} & M_{22}\end{bmatrix}\begin{bmatrix}\dot{\theta}_c \\ \dot{\phi}_c\end{bmatrix} = 0, \tag{27}$$

which shows that the matrix M has a zero mode $(\dot{\theta}_c, \dot{\phi}_c)$ due to the time translation invariance since the Lagrangian does not depend on time τ explicitly.[14–16] To obtain the fluctuation integral I in Eq. (20) we, first of all, have to carry out the integration of one variable which is seen to be Gaussian.[14–16]

Using the hermiticity of M we can reexpress the fluctuation integration I as

$$I=\mathcal{N}\int d\eta_1 d\eta_2 e^{-(1/2)\int_{-T/2}^{T/2}[M_{11}\eta_1^2+2\eta_1(M_{12}\eta_2)+M_{22}\eta_2^2]d\tau}$$
$$=\mathcal{N}\int d\eta_1 d\eta_2 e^{-(1/2)\int_{-T/2}^{T/2}[M_{11}\eta_1^2+2\eta_2(M_{21}\eta_1)+M_{22}\eta_2^2]d\tau}. \tag{28}$$

Since the classical path (θ_c,ϕ_c) is a saddle point,[14–16] we have the following condition:

$$M_{11}>0 \quad \text{or} \quad M_{22}>0, \tag{29}$$

which implies that either η_1 (if $M_{11}>0$) or η_2 (if $M_{22}>0$) would be a Gaussian integration and thus can be carried out.

For the biaxial spin model of large spin s with the classical energy $E(\theta,\phi)$ given by Eq. (7), we have

$$M_{11}=2K_1 s^2(1-\lambda\sin^2\phi_c)\sin^2\theta_c>0. \tag{30}$$

We could perform the Gaussian integration over η_1 as shown in Ref. 8. Here we shall do the integration in an alternative way. The fluctuation integral I can also be expressed as (i.e., the Van Vleck determinant)[14–16]

$$I=\mathcal{N}_1\frac{1}{\sqrt{\det M}}, \tag{31}$$

where $\mathcal{N}_1=(2\pi)^{1/2}\mathcal{N}$. On the other hand, since

$$\begin{bmatrix} M_{11} & M_{12}\\ M_{21} & M_{22}\end{bmatrix}=\begin{bmatrix} M_{11} & 0\\ M_{21} & 1\end{bmatrix}\begin{bmatrix} 1 & M_{11}^{-1}M_{12}\\ 0 & M_{22}-M_{21}M_{11}^{-1}M_{12}\end{bmatrix}, \tag{32}$$

and M_{11} is a positive c number, we have

$$I=\mathcal{N}_1\frac{1}{\sqrt{\det M_{11}\det(M_{22}-M_{21}M_{11}^{-1}M_{12})}}$$
$$=\mathcal{N}_2\frac{1}{\sqrt{\det(M_{22}-M_{21}M_{11}^{-1}M_{12})}}. \tag{33}$$

From Eqs. (24),(25), we obtain

$$M_{22}-M_{21}M_{11}^{-1}M_{12}=-\frac{d}{d\tau}A[\theta_c(\tau),\phi_c(\tau)]\frac{d}{d\tau}+C[\theta_c(\tau),\phi_c(\tau)], \tag{34}$$

where

$$A[\theta_c(\tau),\phi_c(\tau)]=-\frac{l_{12}^2}{M_{11}}=\frac{s^2\sin^2\theta_c}{M_{11}}>0 \tag{35}$$

since $M_{11}>0$,

$$C[\theta_c(\tau),\phi_c(\tau)]=\left(\frac{d}{d\tau}\frac{m_{12}l_{12}}{M_{11}}\right)+M_{22}-\frac{m_{12}^2}{M_{11}}$$
$$=\frac{E_{\theta\phi}^2}{E_{\theta\theta}-\cot\theta_c E_\theta}+is\frac{d}{d\tau}\frac{\sin\theta_c E_{\phi\theta}}{E_{\theta\theta}-\cot\theta_c E_\theta}, \tag{36}$$

and

$$\mathcal{N}_2\simeq\frac{2s+1}{2s}\lim_{n\to\infty}\prod_{k=1}^{n}\left(\frac{A[\theta_c(\tau_k),\phi_c(\tau_k)]}{\tau_k-\tau_{k-1}}\right)^{1/2}. \tag{37}$$

It is interesting to see the difference between our fluctuation integration I and that of traditional approach in terms of the first order Euclidean Lagrangian[11]

$$L_e^{\text{eff}}(\phi,\dot\phi)=\frac{1}{2}m(\phi)\dot\phi^2+V(\phi)-i\Theta(\phi)\dot\phi, \tag{38}$$

which is obtained from the spin-coherent-state path integral Eq. (1) by regarding ϕ and $p=s\cos\theta$ as a pair of conjugate canonical variables and working out the momentum integration,[11] where $\Theta=s-\alpha/m(\phi)$ and

$$m(\phi)=1/[2K_1(1-\lambda\sin^2\phi)],$$
$$V(\phi)=K_2 s^2\sin^2\phi-\frac{\alpha^2\lambda\sin^2\phi}{4K_1(1-\lambda\sin^2\phi)} \tag{39}$$

denote position-dependent mass of the mapped particle and the potential, respectively. The fluctuation functional integral directly obtained from the effective Lagrangian (38) is seen to be

$$I_{\text{eff}}=N\frac{1}{\sqrt{\det\left[-\frac{d}{d\tau}m(\phi_c)\frac{d}{d\tau}+V''(\phi_c)\right]}}, \tag{40}$$

where

$$N=\lim_{n\to\infty}\prod_{k=1}^{n}\left[\frac{m[\phi_c(\tau_k)]}{\tau_k-\tau_{k-1}}\right]^{1/2},$$

and it can also be verified that

$$A[\theta_c(\tau),\phi_c(\tau)]=m[\phi_c(\tau_k)]. \tag{41}$$

We now show the relation between I_{eff} and I. From Eq. (27), one can find $(M_{22}-M_{21}M_{11}^{-1}M_{12})\dot\phi_c=0$, namely,

$$\left(-\frac{d}{d\tau}A[\theta_c(\tau),\phi_c(\tau)]\frac{d}{d\tau}+C[\theta_c(\tau),\phi_c(\tau)]\right)\dot\phi_c=0. \tag{42}$$

Since the zero-mode wave function $\dot\phi_c$ also satisfies[13,14]

$$\left(-\frac{d}{d\tau}m[\phi_c(\tau)]\frac{d}{d\tau}+V''[\phi_c(\tau)]\right)\dot\phi_c=0, \tag{43}$$

seen from the expression of I_{eff}, we have

$$\{C[\theta_c(\tau),\phi_c(\tau)]-V''[\phi_c(\tau)]\}\dot\phi_c=0. \tag{44}$$

From Eq. (13), it is seen that

$$\dot\phi_c(\tau)\neq 0, \quad \text{unless} \quad \tau\to\pm\infty, \tag{45}$$

and

$$\lim_{\tau\to\pm\infty} V''[\phi_c(\tau)] = \lim_{\tau\to\pm\infty} C[\theta_c(\tau),\phi_c(\tau)] = m^*\omega_h^2, \tag{46}$$

where $m^* = m[\phi_c(\tau=\pm\infty)] = 1/(2K_1)$ and ω_h is given in Eq. (14). We therefore conclude

$$C[\theta_c(\tau),\phi_c(\tau)] = V''[\phi_c(\tau)] \tag{47}$$

and

$$I = \frac{2s+1}{2s} I_{\rm eff} \simeq I_{\rm eff}, \tag{48}$$

namely, I and $I_{\rm eff}$ are essentially the same in the large spin approximation. The above derivation shows that tunnel splitting by the spin-coherent-state path integral is equivalent to that by starting from traditional mapping technique[3,11,17,18] in the large spin approximation up to the second order fluctuation.

For other spin systems which may have the condition $M_{22} > 0$, Eq. (33) then can be rewritten as

$$I = \mathcal{N}_1 \frac{1}{\sqrt{\det M_{22} \det(M_{11} - M_{12}M_{22}^{-1}M_{21})}} = \mathcal{N}_2' \frac{1}{\sqrt{\det(M_{11} - M_{12}M_{22}^{-1}M_{21})}}. \tag{49}$$

From Eqs. (24), (25), we obtain

$$M_{11} - M_{12}M_{22}^{-1}M_{21} = -\frac{d}{d\tau} A'[\theta_c(\tau),\phi_c(\tau)] \frac{d}{d\tau} + C'[\theta_c(\tau),\phi_c(\tau)], \tag{50}$$

where

$$A'[\theta_c(\tau),\phi_c(\tau)] = -\frac{l_{21}^2}{M_{22}} = \frac{s^2 \sin^2\theta_c}{M_{22}} > 0, \tag{51}$$

$$C'[\theta_c(\tau),\phi_c(\tau)] = \left(\frac{d}{d\tau}\frac{m_{21}l_{21}}{M_{22}}\right) + M_{11} - \frac{m_{21}^2}{M_{22}} = E_{\theta\theta} - \cot\theta_c E_\theta - \frac{(E_{\theta\phi} - \cot\theta_c E_\phi)^2}{E_{\phi\phi}} - is\frac{d}{d\tau}\frac{\sin\theta_c E_{\phi\theta} - \cos\theta_c E_\phi}{E_{\phi\phi}}. \tag{52}$$

This case is irrelevant to our tunneling model Eq. (6) and we will not give further discussion.

B. Position-dependent mass

To obtain the explicit tunneling kernel $\mathcal{K}$, a convenient way is that the position-dependent-mass may be converted to a constant one so that we can adapt the standard procedure given in literature to evaluate the fluctuation determinant. To this end, we make a global time transformation in Eqs. (31)–(34) by defining a "new" time ξ:

$$\frac{d\xi}{d\tau} = \frac{m^*}{A[\theta_c(\tau),\phi_c(\tau)]}, \tag{53}$$

with which the fluctuation functional integral becomes

$$I = \mathcal{N}_2 \frac{1}{\sqrt{\det\left[\frac{m^*}{A}\left(-m^*\frac{d^2}{d\xi^2} + \frac{AC}{m^*}\right)\right]}} = \mathcal{N}_3 \frac{1}{\sqrt{\det\left(-m^*\frac{d^2}{d\xi^2} + \frac{AC}{m^*}\right)}}, \tag{54}$$

where

$$\mathcal{N}_3 = \frac{2s+1}{2s} \lim_{n\to\infty} \prod_{k=1}^{n} \left(\frac{m^*}{\tau_k - \tau_{k-1}}\right)^{1/2}$$

and we omit the arguments of $A[\theta_c(\tau),\phi_c(\tau)]$ and $C[\theta_c(\tau),\phi_c(\tau)]$ for clarity. From Eqs. (34), (35), and (42), one can verify that $\dot{\phi}_c$ is the zero mode wave function, namely,

$$\left(-m^*\frac{d^2}{d\xi^2} + \frac{AC}{m^*}\right)\dot{\phi}_c(\xi) = 0, \tag{55}$$

where $\dot{\phi}_c = (d/d\tau)\phi_c$.

Now the fluctuation integral I can be evaluated from the fluctuation determinant for a particle with constant mass m^*. It is worthwhile to remark that the time transformation to remove the position dependence of the mass is the key point of our formulation which, We will see, results in a better tunnel splitting equivalent to that obtained from semiclassical correction of the Lagrangian.[17,18,22–24] The fluctuation integral Eq. (33) is ill defined because of the zero mode $\dot{\phi}_c$ seen from Eq. (43) which leads to divergence of the integral. However the divergence problem can be cured by means of the Faddeev-Popov procedure[14–16] from which the fluctuation integral is found to be $I = \sqrt{S_c/m^*}T\tilde{I}$, where

$$\frac{S_c}{m^*} = \int_{-T/2}^{T/2} \dot{\phi}_c^2(\tau)\,d\tau \tag{56}$$

and

$$\tilde{I} = D\left[\frac{\det'\left(-m^*\frac{d^2}{d\xi^2} + \frac{AC}{m^*}\right)}{\det\left(-m^*\frac{d^2}{d\xi^2} + m^*\omega_h^2\right)}\right]^{-1/2}. \tag{57}$$

Here D is the fluctuation determinant of the harmonic oscillator with mass m^*,

$$D=\mathcal{N}_3\left[\det\left(-m^*\frac{d^2}{d\xi^2}+m^*\omega_h^2\right)\right]^{-1/2}=\left(\frac{m^*\omega_h}{\pi}\right)^{1/2}e^{-\omega_h\Pi/2}, \tag{58}$$

and Π denotes the time period in new time scale ξ. By a standard procedure,[14–16] we can find

$$I=D\left(\frac{m^*\omega_h}{\pi}\right)^{1/2}LT, \quad L=\lim_{\xi\to\infty}\dot{\phi}_c(\xi)e^{\omega_h\xi}. \tag{59}$$

Then the prefactor is seen to be

$$B=\frac{2s+1}{2s}\left(\frac{m^*\omega_h}{\pi}\right)^{1/2}L. \tag{60}$$

Using the instanton solution given in Eq. (13), Eqs. (41) and (53) lead to the time period

$$\int d\xi=\int d\tau-\lambda\int \sin^2\phi_c d\tau$$
$$=\int d\tau-\frac{m^*}{2s}\ln\frac{\sqrt{1-u^2}+\sqrt{\lambda}\tanh(\omega_h\tau)}{\sqrt{1-u^2}-\sqrt{\lambda}\tanh(\omega_h\tau)}. \tag{61}$$

If we ignore the second term on the right hand side of the above equation which is seen to be of $1/s$ order, namely if we take the approximation such that

$$\lim_{\xi\to\infty}\dot{\phi}_c(\xi)e^{\omega_h\xi}\simeq\lim_{\tau\to\infty}\dot{\phi}_c(\tau)e^{\omega_h\tau}=\tilde{L}, \tag{62}$$

we then find the following prefactor:

$$\tilde{B}=\frac{2s+1}{2s}\left(\frac{m^*\omega_h}{\pi}\right)^{1/2}\tilde{L}\simeq\left(\frac{m^*\omega_h}{\pi}\right)^{1/2}\tilde{L}, \tag{63}$$

where

$$\tilde{L}=2\omega_h(1-u^2)^{1/2}(1-u^2-\lambda)^{-1/2}. \tag{64}$$

$\tilde{B}$ is exactly the prefactor used by Enz and Schilling [see Eqs. (8b) and (8d) in Ref. 17 and Eqs. (13b) and (13d) in Ref. 18] and also Kou *et al.*[13] This result shows explicitly that, in the previous calculations,[13,17,18] a term [i.e., the second term on the right hand side of Eq. (61)] of $(1/s)$ order is omitted in the global time transformation in the evaluation of the prefactor. We show that it is the omitted term which leads to an extra factor contributing to the magnitude of tunnel splitting significantly.

We now find this extra factor. Setting an arbitrary constant of integration to zero, Eq. (61) leads to

$$e^{\omega_h\xi}=e^{\omega_h\tau}\left(\frac{\sqrt{1-u^2}-\sqrt{\lambda}\tanh(\omega_h\tau)}{\sqrt{1-u^2}+\sqrt{\lambda}\tanh(\omega_h\tau)}\right)^c, \tag{65}$$

where $c=\frac{1}{2}\sqrt{\lambda(1-u^2)}$. It follows that

$$L=\lim_{\xi\to\infty}\dot{\phi}_c(\xi)e^{\omega_h\xi}=\lim_{\tau\to\infty}\dot{\phi}_c e^{\omega_h\tau}\left(\frac{\sqrt{1-u^2}-\sqrt{\lambda}\tanh(\omega_h\tau)}{\sqrt{1-u^2}+\sqrt{\lambda}\tanh(\omega_h\tau)}\right)^c$$
$$=Q\tilde{L}, \tag{66}$$

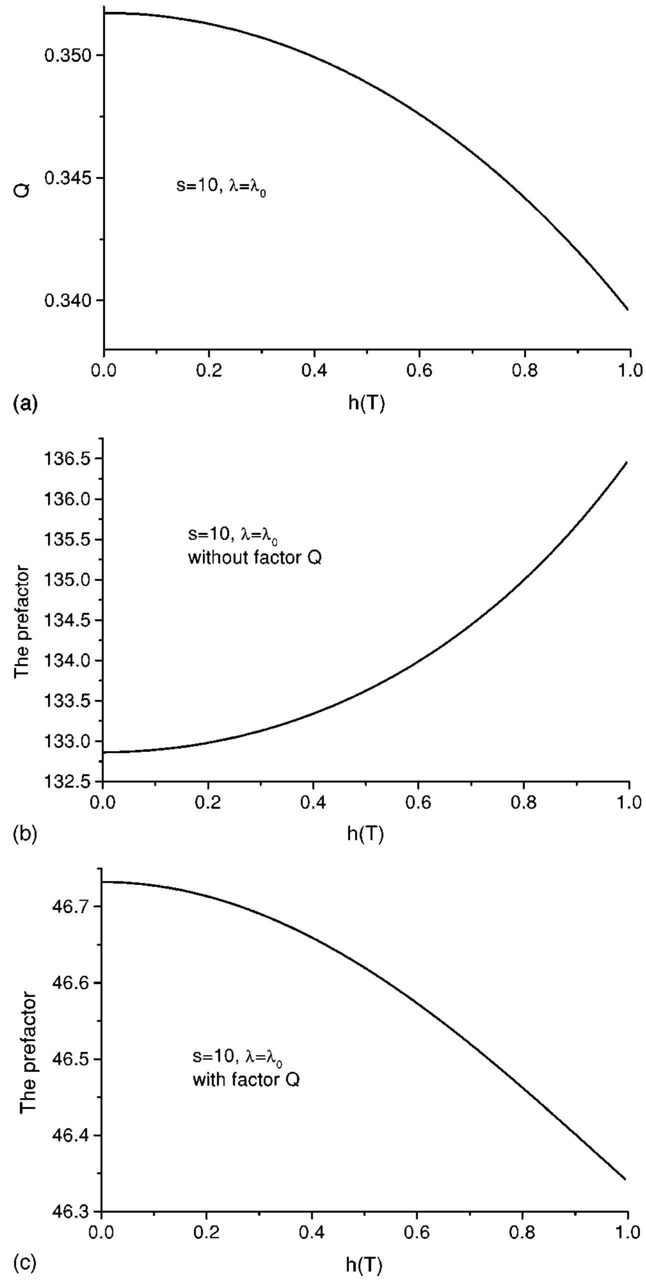

FIG. 1. Field dependence of Q (a) and prefactors $4\tilde{B}$ (b), $4Q\tilde{B}$ (c) for $s=10$ and $K_1=0.321$ K; $K_2=0.229$ K.

where Q is the extra factor

$$Q=\left(\frac{\sqrt{1-u^2}-\sqrt{\lambda}}{\sqrt{1-u^2}+\sqrt{\lambda}}\right)^c. \tag{67}$$

Substituting the result back to Eq. (60), we can find

$$B=Q\tilde{B}. \tag{68}$$

Therefore, the ground state tunnel splitting in biaxial spin model is expressed as

$$\Delta E=4Q\tilde{B}e^{-S_c}|\cos\theta_s|. \tag{69}$$

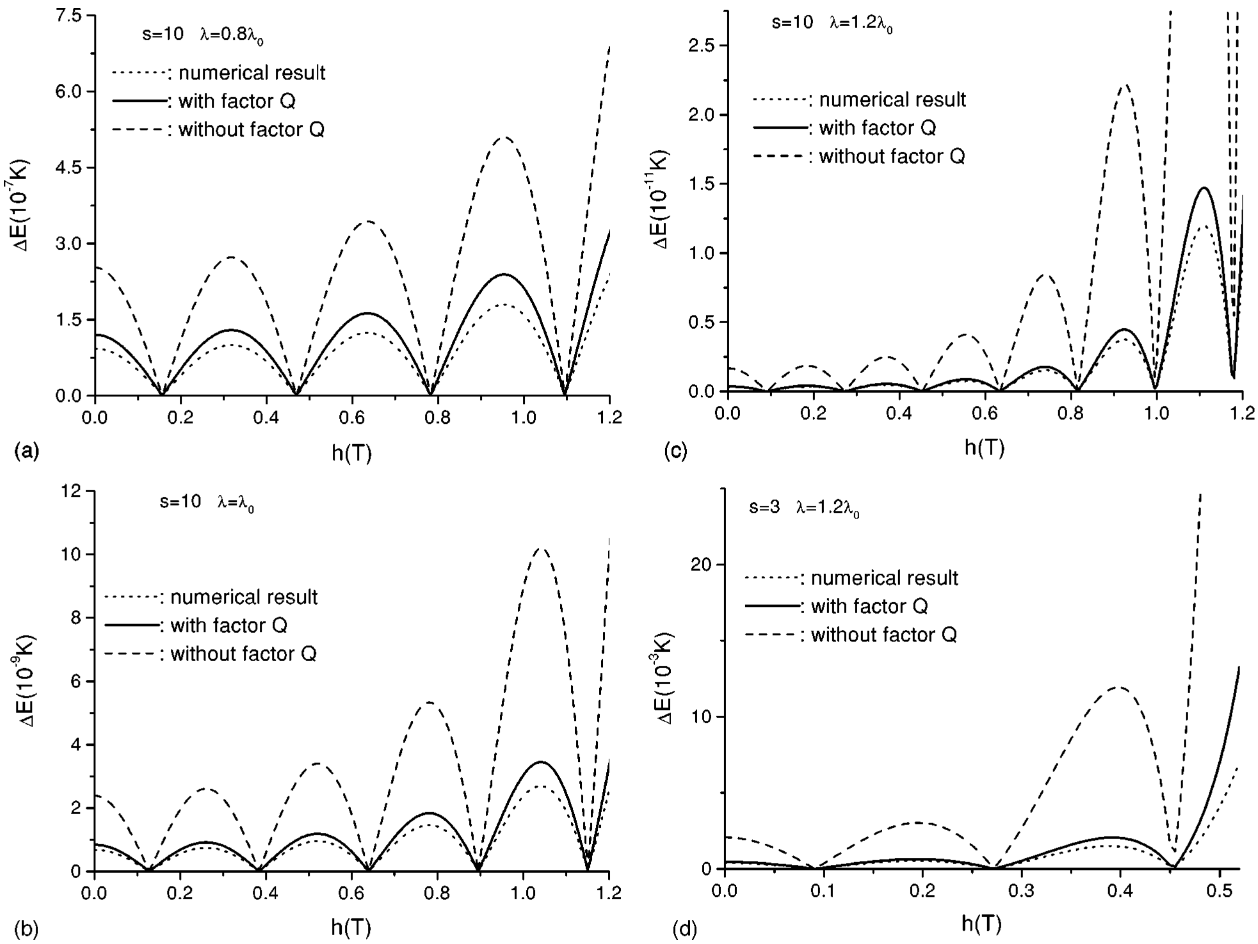

FIG. 2. Field dependence of tunnel splitting at ground state for $s=10$, 3 and various $\lambda=K_2/K_1$, $\Delta E'=4\tilde{B}e^{-Sc}\cos\theta_s$ (dash line), $\Delta E=4Q\tilde{B}e^{-Sc}\cos\theta_s$ (solid line), and numerical result of diagonalization(dotted line). (a) $s=10$, $K_1=0.321$ K; $K_2=0.8\times 0.229$ K, (b) $s=10$, $K_1=0.321$ K; $K_2=0.229$ K, (c) $s=10$, $K_1=0.321$ K; $K_2=1.2\times 0.229$ K, (d) $s=3$, $K_1=0.321$ K; $K_2=1.2\times 0.229$ K.

The pre-exponential factor of the level splitting Eq. (69) is different from that in literature[13,17] by a factor Q which is missing in the standard procedure for evaluation of the fluctuation integral suitable only to the model with a constant mass.[19] To see the effect of the factor Q, we present here a numerical estimation of external field dependence of Q, $4\tilde{B}$ and $4Q\tilde{B}$. Making the use of the anisotropy constants in Ref. 10 such that $K_1=0.321$ K, $K_2=0.229$ K, field dependence of Q, $4\tilde{B}$ and the prefactor $4Q\tilde{B}$ are shown in Fig. 1 for $s=10$. One can see that the factor Q lowers the energy splitting by about 3 times, and a more important effect is that Q modifies the behavior of field dependence of the prefactor, namely, $4\tilde{B}$ increases with the applied field, while $4Q\tilde{B}$ decreases with the field. Analytical value of energy splitting with and without the correction factor Q is shown in Fig. 2, together with numerical result by performing a diagonalization of the Hamilton operator given in Eq. (6). As we can see from Fig. 2, if the factor Q is missing in the prefactor, the analytical splitting strongly deviates from numerical result especially in high field regions (see Fig. 2 dot-and-dash line), confirming the observation of Enz and Schilling,[17] and this is the reason why the so-called semiclassical Lagrangian for the spin system was used in Refs. 17,24 to obtain a better magnitude of the tunnel splitting. However, when the factor Q is included, the analytical splitting (69) agrees quantitatively with numerical result for spin number as low as $s=3$ with $\lambda\simeq 0.856$. Our result shows that spin-coherent-state path integral representation which gives rise to the classical energy Eq. (7) results in an accurate energy splitting of the ground state, provided that the fluctuation determinant for the spin model with a position-dependent mass is evaluated more exactly. It is worthwhile to remark that the instanton method is, strictly speaking, valid in the weak coupling region where the barrier should be high enough. The applied magnetic field lowers the barrier height seen from Eq. (39), therefore, increases the coupling. The theoretical tunnel splitting evaluated by instanton method becomes worse with increasing magnetic field. Our method is valid until a critical value of magnetic field

$$h_c=\frac{2s(1-\lambda)}{g\mu_B}\sqrt{K_1K_2/\lambda}$$

obtained from the condition that $V''(\pi/2)=0$. Beyond the critical magnetic field the top of potential barrier becomes concave.

TABLE I. Energy levels $(E_n - E_0)$ by quantization rule in Eq. (70) and by numerical diagonalization of the Hamilton operator. The unit of energy is K and $E_0 = -29.7995$.

n	0	1	2	3	4	5
Numerical result	0.0	5.14164	9.72755	13.7499	17.1886	19.9547
Quantization rule	0.0	5.14708	9.74084	13.77567	17.2403	20.10912

To show the validity of our method, we present two more numerical results in the absence of external magnetic field to show that the classical energy Eq. (7) is good for the semiclassical description of the biaxial spin model in the path integral formulation. The quantized low-lying levels for a double-well potential can be found by Bohr-Sommerfeld rule,[25–27] i.e., $\oint p dx = n 2\pi$ numerically. With the help of classical Hamiltonian Eq. (7) for zero-field case regarding ϕ and $p = s\cos\theta$ as a pair of conjugate canonical variables which becomes

$$H = \frac{p^2}{2m(\phi)} + K_2 s^2 \sin^2\phi,$$

the quantized levels are determined by

$$2\int_{-\phi_n}^{\phi_n} \sqrt{2m(\phi)(E_n - K_2 s^2 \sin^2\phi)}\, d\phi = n2\pi, \tag{70}$$

where ϕ_n is the turning point given by $\phi_n = \sin^{-1}(E_n/K_2 s^2)^{1/2}$, and $n = 0,1,2,\ldots$. Using the parameters in Ref. 10 for Fe_8 system, the first five levels obtained from Eq. (70) are shown in Table I, together with the exact energy levels E_n determined by numerical diagonalization of the Hamilton operator. We can see from the table that the energy levels obtained with the classical energy are in good agreement with the exact ones. This implies that the representation of the spin-coherent-state path integral provides a reasonable description for determining the energy levels as well as the level splitting in semiclassical approximation. On the other hand, the s-dependence of zero-field action given by Eq. (11), namely,

$$S_c(h=0) = s \ln\frac{1+\sqrt{\lambda}}{1-\sqrt{\lambda}}, \tag{71}$$

which is seen to be the same as that given in Ref. 3, can be easily verified by numerical calculation. To this end, we present a semi-log-plot of the zero-field tunnel splitting ΔE in the ground state for integer s in Fig. 3 obtained in terms of the numerical diagonalization. It is seen that, for $\lambda = 0.7134$ in Fe_8 molecular cluster, the curve of $\ln\Delta E$ as a function of spin number s (integer) is fitted best by $F(s) = -2.3s + 1.18133$ (dotted line in Fig. 3). Neglecting the s dependence of the prefactor (which is of order $\ln s$),[8] numerical result agrees with the above expression Eq. (71) since $\ln(1+\sqrt{\lambda})/(1-\sqrt{\lambda}) \simeq 2.474$.

In this paper we show that the failure of coherent state representation of path integral in computation of tunnel splitting in spin systems can be cured by more exactly dealing with the position-dependent mass in terms of a global time transformation which is seen to be equivalent to the correction of $1/s$ order in Lagrangian first proposed by Enz and Schilling[17] and subsequently used by Liang *et al.*[24] in the case of absence of magnetic field. It is interesting to see how the apparently very different viewpoints lead to the same correction of level splitting. To this end we rewrite the tunnel splitting of Enz and Schilling [see Eqs. (8b)–(9b) in Ref. 17] for the case of vanishing external field ($h=0$) with the approximation $\sqrt{s(s+1)} \simeq s + 1/2$ by factorizing out the classical action S_c such that[28]

$$\Delta E_{ES} = 4 Q_{ES} \tilde{B} e^{-S_c}, \tag{72}$$

where the factor

$$Q_{ES} = \left(\frac{1-\sqrt{\lambda}}{1+\sqrt{\lambda}}\right)^{1/2} \tag{73}$$

with subscript "ES" denoting the factor of Enz and Schilling resulted from the correction of $1/s$ order in the Lagrangian is similar to our factor Q in Eq. (69) which in the case of vanishing field reduces to

$$Q(h=0) = \left(\frac{1-\sqrt{\lambda}}{1+\sqrt{\lambda}}\right)^{\sqrt{\lambda}/2}. \tag{74}$$

The level splitting Eq. (72) is given exactly in Ref. 24 for the case of absence of magnetic field. The factor Q_{ES} has been obtained from a completely different approach by Liang, Müller-Kirsten and Rana[29] and our findings, therefore, receive a verifying support from the results in Ref. 29. The numerical magnitudes of tunnel splitting evaluated from formulas (72) and (69) are compared in Table II for $s=2$ to 7

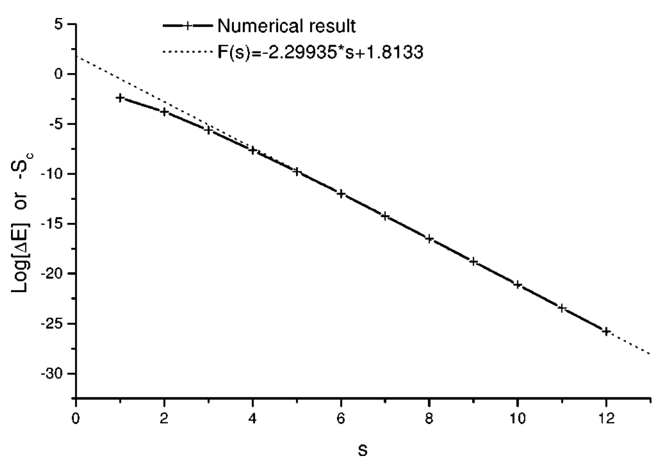

FIG. 3. Semi-log plot of $\ln\Delta E$ vs s.

TABLE II. Ground state tunnel splitting of Eq. (69) and ΔE_{ES} in comparison with the numerical result ΔE_0 by diagonalization of the Hamilton operator for some typical values of λ (The unit of the energy is K).

(a) $\lambda=\lambda_0=0.7134$						
s	2	3	4	5	6	7
ΔE_0	2.261×10^{-2}	3.604×10^{-3}	4.737×10^{-4}	5.618×10^{-5}	6.251×10^{-6}	6.658×10^{-7}
ΔE	2.865×10^{-2}	4.589×10^{-3}	5.951×10^{-4}	7.005×10^{-5}	7.756×10^{-6}	8.232×10^{-7}
ΔE_{ES}	3.758×10^{-2}	5.134×10^{-3}	6.219×10^{-4}	7.011×10^{-5}	7.535×10^{-6}	7.825×10^{-7}
(b) $\lambda=1.15\lambda_0$						
s	2	3	4	5	6	7
ΔE_0	8.47×10^{-3}	7.88×10^{-4}	6.067×10^{-5}	4.221×10^{-6}	2.756×10^{-7}	1.723×10^{-8}
ΔE	1.044×10^{-2}	9.488×10^{-4}	7.223×10^{-5}	4.992×10^{-6}	3.245×10^{-7}	2.022×10^{-8}
ΔE_{ES}	1.43×10^{-2}	1.138×10^{-3}	8.059×10^{-5}	5.319×10^{-6}	3.35×10^{-7}	2.04×10^{-8}
(c) $\lambda=0.85\lambda_0$						
s	2	3	4	5	6	7
ΔE_0	4.452×10^{-2}	1.056×10^{-2}	2.058×10^{-3}	3.611×10^{-4}	5.939×10^{-5}	9.349×10^{-6}
ΔE	6.172×10^{-2}	1.411×10^{-2}	2.702×10^{-3}	4.698×10^{-4}	7.683×10^{-5}	1.205×10^{-5}
ΔE_{ES}	7.3809×10^{-2}	1.498×10^{-2}	2.688×10^{-3}	4.485×10^{-4}	7.129×10^{-5}	1.205×10^{-5}

and various K_1, K_2 with the result of numerical diagonalization of the Hamilton operator $\hat{H}=K_1\hat{S}_z^2+K_2\hat{S}_y^2$ in the matrix representation in terms of eigenstates of operator $\hat{S}_z$ with the usual computational program MATHEMATICA. It is obviously that our formula is accurate same as Eq. (72) and better for small spin s.

IV. CONCLUSION

The existing instanton method in literature for evaluation of fluctuation determinant of tunneling kernel is valid only for a particle of constant mass, and is therefore not suitable to the spin model at hand which possesses a position dependent mass. We use a time transformation to remove the position dependence of the mass and obtain a additional factor Q in the prefactor of tunnel splitting which leads to the accurate magnitude of tunnel splitting as well as the level space in comparison with the numerical result of diagonalization of the Hamilton operator. We conclude that the representation of spin-coherent-state path integrals is sufficiently good for evaluation of tunneling amplitude.

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