Volume 9, Number 11
1009-1963/2000/09(11)/0848-07
CHINESE PHYSICS
November, 2000
© 2000 Chin. Phys. Soc.

NUMERICAL STUDY ON TUNNELING SPLITTING IN BIAIXAL SPIN SYSTEMS*

CHEN ZHI-DE(陈芝得) and ZHANG SHU-QUN(张树群)
Department of Physics, Guangzhou Normal University, Guangzhou 510405, China

(Received 10 March 2000; revised manuscript received 6 July 2000)

Numerical study on tunneling splitting in biaxial spin systems is done by performing diagonalization of the Hamilton operator. It is found that the calculated energy splitting agrees quantitatively with theoretical prediction of instanton method. Our result shows that both the instanton method and the large spin limit work well for the total spin around 10. By including the fourth-order term in Hamiltonian, experimental observation can be re-covered quantitatively.

Keywords: tunneling splitting, biaxial spin systems, numerical study
PACC: 7500, 0365

I. INTRODUCTION

Tunnelling of magnetization in mescoscopic systems has become a very attractive field in recent years.[1−9] Biaxial spin model has been recently the object of most intensive researches[6,9−13] due to its unique macroscopic quantum coherence(MQC),[4−6] as compared with the uniaxial spin model. MQC in biaxial spin model with a field along the hard axis was first elucidated by Garg,[6] and lately by Kou *et al.* in more detail.[10] Encouragingly, such a MQC phenomenon is now seen in the biaxial magnetic cluster of formula $[Fe_8O_2(OH)_{12}(tacn)_6]^{8+}$ (shortly Fe_8).[9] Recently, quantum theory on tunneling splitting in this model was also developed.[13] Another convenient way to study tunneling splitting is to perform diagonalization of the Hamilton operator by numerical calculation, and some results can be found in both theoretical and experimental work mentioned above.[6,9,10,12] Numerical study is simple but by no means trivial, it can help to provide some guidance for theoretical study. For example, theoretical study on tunneling splitting in biaxial spin model in high field range predicted a new type of instanton with a new quantum interference phenomenon,[11] this prediction can be easily tested by numerical study, though experimental result is not yet available. As we know, however, a systematic numerical study on biaxial spin model with a detailed comparison with theoretical work is still lacking. In the present paper, we present a systematic numerical study on the biaxial spin model with and without the fourth order terms.[9] Numerical result is used to test theoretical prediction by instanton method, [14−17] as well as the S-dependent periodicity predicted in a recent theory.[1)] The organization of the paper is as follows: the model and explanation are given in section II. Oscillation of the tunneling splitting with the transverse field is demonstrated in section III for the model with and without fourth-order terms. Analysis of numerical result and the main conclusion will be provided in section IV.

II. THE MODEL AND THE EXPLANATION

The main interest will be concentrated on the biaxial spin model with Hamiltonian

$$H_0 = -DS_z^2 + E(S_x^2 - S_y^2) - g\mu_B h_z S_z - g\mu_B h_T(S_x\cos\varphi + S_y\sin\varphi), \quad (1)$$

where h_z is the field component along the easy axis, h_T is the transverse field applied in the XY plane at an azimuth angle φ. Anisotropic constants are chosen according to Ref.[9], such as

$$D = 0.275K, \quad E = 0.0462K, \quad g = 2, \quad (2)$$

h_z is always set to be zero except for the case of "resonant tunnelling" (see below), while the fourth-order term, namely[9]

$$H_1 = C(S_+^4 + S_-^4), \quad C = -2.95\times10^{-5}, \quad (3)$$

will be included in some cases for comparison.

Denoting the eigenstate along the easy axis as $|n\rangle, n = 0, \pm1, \pm2, \cdots, \pm S$, one can easily work out

*Project supported by Guangdong provincial Natural Science Foundation of China (Gront No. 994496).
1)Zhi-De Chen *et al.*, submitted to Europhys. Lett..

the matrix element

$$\langle i|H_0|j\rangle = \frac{E}{2}a_1\delta_{i,j+2} + \frac{E}{2}a_2\delta_{i,j-2} + \frac{D}{2}a_3\delta_{i,j} - a_4\delta_{i,j+1} - a_5\delta_{i,j-1} - g\mu_{\rm B}h_z j\delta_{i,j}, \quad (4)$$

where

$$a_1 = \sqrt{(S-j)(S-j+1)(S-j-1))(S+j+2)},$$
$$a_2 = \sqrt{(S+j)(S-j+1)(S+j-1))(S-j+2)}, \quad (5)$$
$$a_3 = [(S-j)(S+j+1) + (S+j)(S-j+1)], \quad (6)$$

and

$$a_4 = {\rm e}^{-i\varphi}\frac{1}{2}g\mu_{\rm B}h\sqrt{(S-j)(S+j+1)},$$
$$a_5 = {\rm e}^{i\varphi}\frac{1}{2}g\mu_{\rm B}h\sqrt{(S+j)(S-j+1)}, \quad (7)$$

The matrix element of H_1 is

$$\langle i|H_1|j\rangle = b_1\delta_{i,j+4} + b_2\delta_{i,j-4}, \quad (8)$$

where

$$b_1 = C\sqrt{(S-j)(S+j+1)(S-j-1)(S+j+2)(S-j-2))(S+j+3)(S-j-3)(S+j+4)}, \quad (9)$$

$$b_2 = C\sqrt{(S+j)(S-j+1)(S+j-1)(S-j+2)(S+j-2))(S-j+3)(S+j-3)(S-j+4)}. \quad (10)$$

The main work of numerical calculation is the diagonalization of the $(2S+1)\times(2S{+}1)$ matrix of the Hamilton operator, that is, to find out the $(2S{+}1)$ eigenvalues. If there is no tunneling, eigenstates $|n\rangle$ and $|-n\rangle$ should be exactly degenerate, while tunneling leads to energy splitting

$$\Delta E_n = [E(n) - E(-n)], \quad (11)$$

which is called tunneling splitting. $\Delta E_S = E(S) - E(-S)$ is called the tunneling splitting of the ground state, while $\Delta E_n (n < S)$ are called the tunneling splitting of the excited states. It should be noted that such an energy splitting is really small, as compared with the eigenvalues($\Delta E/E \sim 10^{-12}$ for ground state), therefore calculation accuracy should be controlled to ensure reliability of the output data. Before going to discuss the numerical result, we first review some main conclusion of theoretical analysis. The Hamiltonian of the biaxial model for theoretical study is expressed as[6,10]

$$H = K_1 S_x^2 + K_2 S_y^2 - g\mu_{\rm B}h(S_x\cos\varphi + S_y\sin\varphi),$$
$$K_1 > K_2, \quad (12)$$

comparing with Eq.(1), one can find

$$K_1 = E + D, \quad K_2 = D - E, \quad \lambda = \frac{K_2}{K_1} = \frac{D-E}{D+E}. \quad (13)$$

A notable subject in this system is MQC, which comes from the interference of spin paths with different windings.[4−6,9,10] When the field is applied along the hard axis (i.e., h_z=0 and $\varphi = 0$), it is predicted that the tunneling splitting of the ground state oscillates with the transverse field such as[6,10]

$$\Delta E \propto 2Q{\rm e}^{-S_c}\left|\cos\left(S\pi + \frac{h_T}{\Delta H}\pi\right)\right|,$$
$$S_c(h_T = 0) = S\log\frac{1+\sqrt{(D+E)/(D-E)}}{1-\sqrt{(D+E)/(D-E)}} \simeq 2.474S, \quad (14)$$

where S_c is the instanton action, Q is called the prefactor[17] and a detailed expression for S_c and Q can be found in Ref.[10]. ΔH is the oscillation periodicity given by[6,10]

$$\Delta H = \frac{2k_{\rm B}}{g\mu_{\rm B}}K_1(1-\lambda)^{1/2} = \frac{2k_{\rm B}}{g\mu_{\rm B}}\sqrt{2E(E+D)} \simeq 2.557T. \quad (15)$$

Since MQC effect is the unique characteristic of tunneling splitting, the re-production of the oscillation of energy splitting with the transverse field will be taken as a convincing guarantee for ensuring that the calculated energy splitting does represent the tunneling splitting. Equation(14) shows that zero-field splitting for S=10 is $\Delta E(h_T = 0) \sim 10^{-10}$, which is about 2 order lower than experimental result.[9] The predicted periodicity is substantially lower than the experimental(i.e., 0.41T).[9]

Ⅲ. ENERGY SPLITTING AS A FUNCTION OF TRANSVERSE FIELD

We first present the calculated energy splitting for ground state of S=10 and 10.5 in Fig.1, where theoretical prediction is also included for comparison. The result shows that the calculated energy splitting coincides with theoretical prediction quantitatively, and

thus guarantees that the calculated energy splitting indeed represents the tunneling splitting. In the case of $h_z = 0$, tunneling splitting depends on both the magnitude and the azimuth angle(i.e., φ) of the transverse field, numerical result is shown in Fig.2 for S=10 and S=10.5. This result is in quantitative agreement with theoretical prediction[18] and qualitatively agrees with experimental observation.[9] For S ranging from 1 to 13, the variation of the tunneling splitting with the transverse field is qualitatively the same(the same oscillation periodicity but the amount of splitting decreases exponentially with ground state energy, see below). It is interesting to note that, not just the ground state, the energy splitting of the excited states(say, $n = \pm(S-1), \pm(S-2), \cdots,$) shows the same oscillation behavior as the ground state. Some representative results for S=10 and 10.5 are shown in Figs.3 (a) and 3(b). This result is easy to understand since the energy splitting for all levels is controlled by the same off-diagonal elements. To illustrate the dependence of the energy splitting on the level-energy, we present a semi-log plot of zero-field-energy-splitting versus level energy in Fig.4. Our result shows that, for S ranging from 8 to 14,

$$\log\Delta E_n/\sqrt{E(n)} \simeq k\sqrt{E(n)} + \text{const},$$
$$k \in [-0.934, -1.34]. \qquad (16)$$

This implies that the energy splitting decreases exponentially with the level energy.

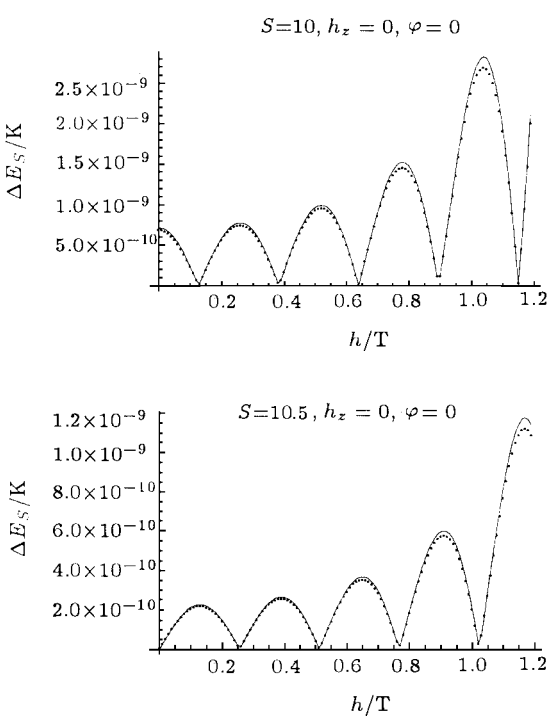


Fig.1. Oscillation of tunneling splitting with the transverse field in the case of h_z.=0 and φ=0. Theoretical prediction of Ref.[10] is shown in solid line.

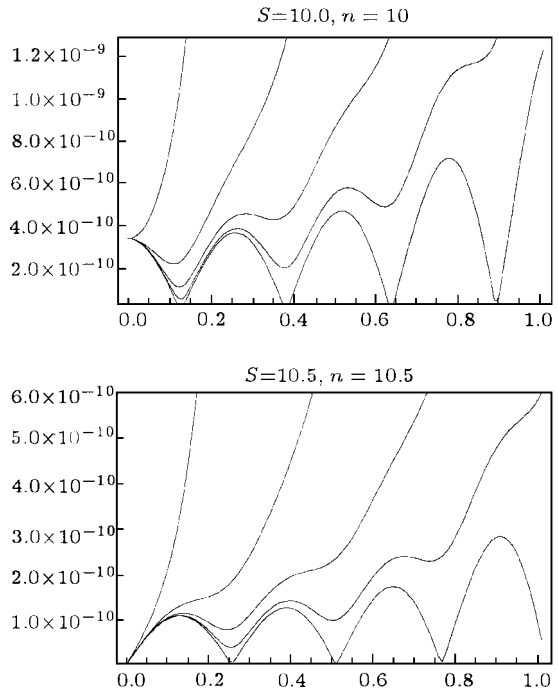


Fig.2. The calculated ground state splitting $\Delta E_n(n = \pm S)$ as a function of transverse field for S=10 and S=10.5 (from top to bottom: $\varphi = 90°, 20°, 10°, 0°$).

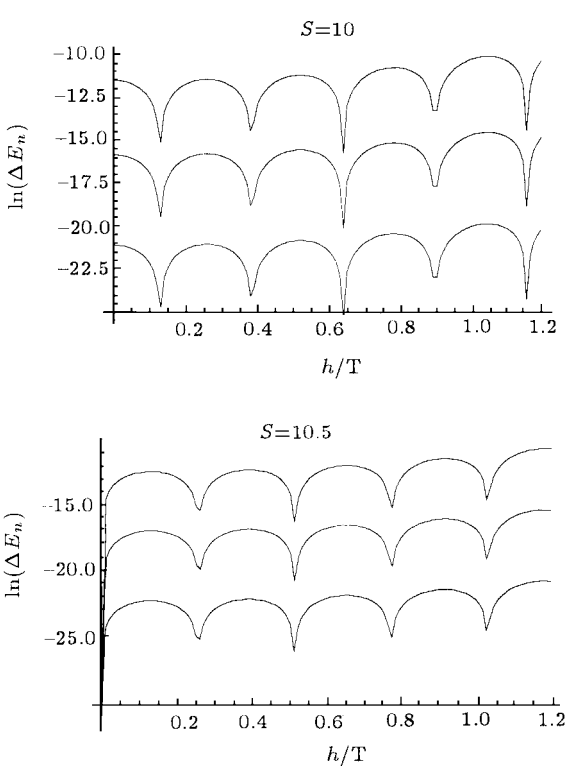


Fig.3.(a) Oscillation of tunneling splitting $\Delta E_n(n = S$(bottom), $S-1, S-2$(top)) with the field along the hard axis in semi-log plot (the result for S=10 can also be found in Fig.4 in Ref.[12]).

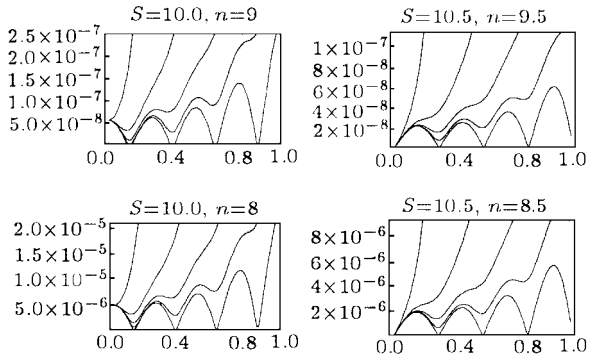


Fig.3.(b) Variation of the tunneling splitting of excited states $\Delta E_n(n = S-1, S-2$ with the transverse field for S=10 and 10.5 (from top to bottom: φ=90°, 20°, 10°, 0°).

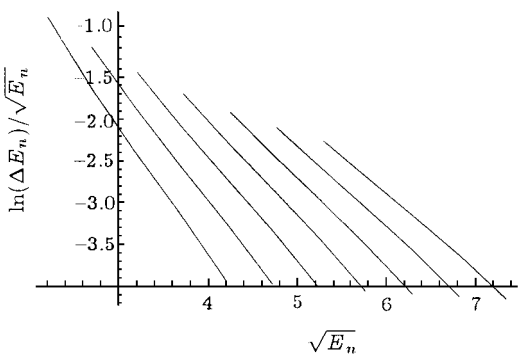


Fig.4. Semi-log plot of $\Delta E_n/\sqrt{E(n)}$ vs $\sqrt{E(n)}$ for integer S(from left to right, S=8,9,$\cdots$,14).

When the field along easy axis is involved (i.e., $h_z \neq 0$), the degenerate states under zero-h_z condition become non-degenerate, and thus one cannot observe the oscillation of the energy splitting with the transverse field. However, by tuning h_z to some specified values such as

$$h_z = m\Delta h, \quad m = 1, 2, 3, \cdots,$$
$$\Delta h = [E(S-1) - E(S)]/g\mu_{\rm B}, \qquad (17)$$

the eigenstates $|-S\rangle$ and $|S-m\rangle$ become "degenerate", and the oscillation of the energy splitting $[E(-S) - E(S-m)]$ with the transverse field is resumed. Such a phenomenon is termed as "resonant tunneling" in experiment.[8] Typical results for "resonant tunneling" of S=10 and 10.5 are shown in Fig.5(a), where one can find the result agrees with the experimental observation qualitatively.[9] The variation of the "resonant tunneling" with the magnitude and the angle of the transverse field is shown in Fig.5(b), the behavior is found to be qualitatively the same as the ground state splitting, but there are two important differences. One is the amount of splitting. The other is the different spin parity effect. It is found that, the spin parity for even m is the same as the ground state, while the spin parity for odd m is opposite to the ground state (say, m=1 for S=10 shows spin parity as m=0 for S=10.5). Such a variation of spin parity with S and m has been observed in experiment and re-produced by numerical result including the fourth-order terms.[9] It should be noted that such a tunneling is qualitatively different from tunneling between two degenerate ground states, and thus is beyond the power of instanton method. U_p to now, a convineing theoretical explanation is still lacking.

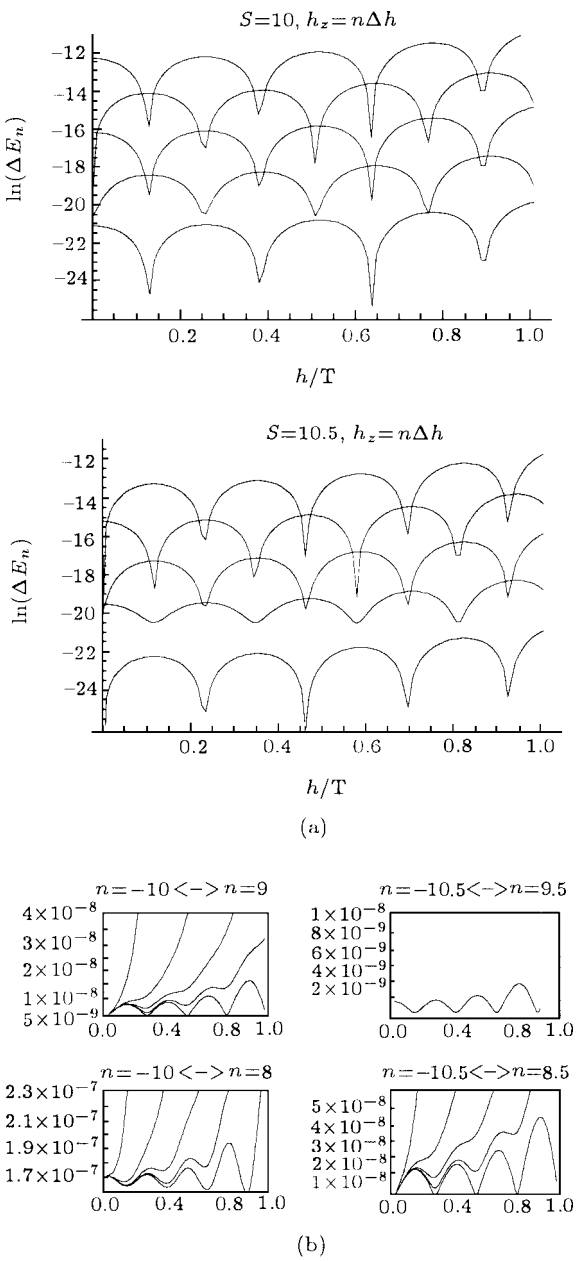


Fig.5. Illustration of resonant tunneling between $|n = S\rangle$ and $|n = S-1, S-2, \cdots,\rangle$ for S=10 and 10.5. One can see different spin parity by comparing with Fig.3. (a) Semi-Log plot for φ=0; (b) Variation of resonant tunneling with the transverse field (from top to bottom: φ=90°, 20°, 10°, 0°).

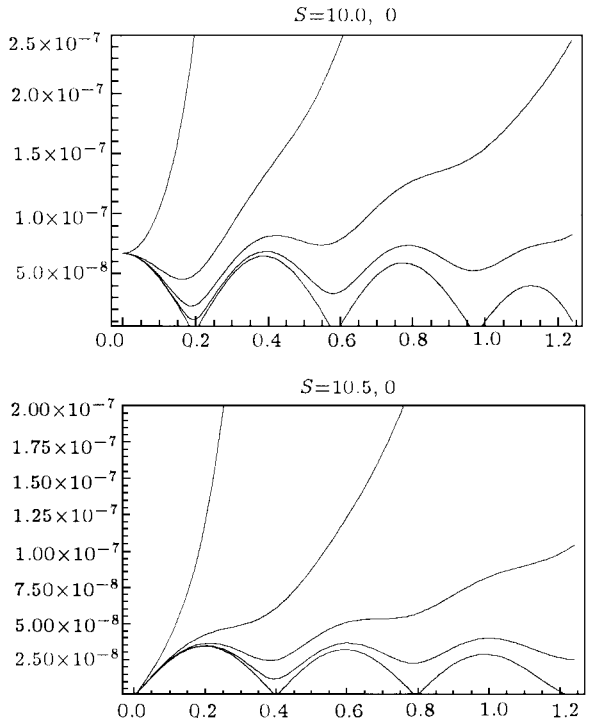


Fig.6. The calculated ground state splitting $\Delta E(n = \pm S)$ including $C(S_+^4 + S_-^4)$ as a function of transverse field for S=10 and S=10.5 (from top to bottom: φ=90°, 20°, 10°, 0°).

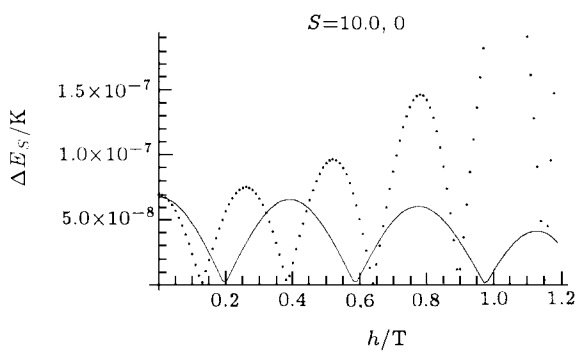


Fig.7. Oscillation of tunneling splitting for S=10 with and without the fourth-order term (solid line: $\Delta E_n(n = S)$ including $C(S_+^4 + S_-^4)$, broken line: $100\times\Delta E_n(n = S)$ without $C(S_+^4 + S_-^4)$).

Although theoretical analysis including the fourth-order term (3) is still unavailable, numerical calculation with the fourth-order term is easy to do. The variation of the tunneling splitting including $C(S_+^4 + S_-^4)$ as a function of transverse field is shown in Fig.6. Our result shows that the global variation looks like the case without fourth-order term(c.f. Fig.2), however, including of $C(S_+^4 + S_-^4)$ improves the calculated result, being closer to the experimental results. It is found that the amount of splitting increases about 2 orders higher and the oscillation periodicity increases to 0.4T, which is close to experimental observation(0.41T). To make it more clear, we present the variation of tunneling splitting with and without $C(S_+^4 + S_-^4)$ as a function of transverse field($h_z = 0$, $\varphi = 0$) in Fig.7, where the splitting without $C(S_+^4 + S_-^4)$ has been multiplied by 100 in order to match the scale. Another improvement is the suppression of the quick-increasing splitting in high field regions.[9]

IV. CONCLUSION AND DISCUSSION

The first conclusion of our numerical result is that the instanton method works very well, and the discrepancy between theoretical prediction and experimental observation comes from the fourth-order term, namely, one needs to include $C(S_+^4 + S_-^4)$ in Hamiltonian to re-cover experimental observation quantitatively. The core of the instanton method is the calculation of the instanton action, which is the main contribution to tunneling splitting.[10,14–17] Up to now, the instanton action has been calculated with or without mapping technique and slightly different S-dependence were found.[3,6,10] The mapping technique predicts the zero-field instanton action as[10]

$$S_{c1} = \sqrt{S(S+1)}\log\frac{1+\sqrt{(D+E)/(D-E)}}{1-\sqrt{(D+E)/(D-E)}} \simeq 2.474\sqrt{S(S+1)}, \quad (18)$$

while the case without mapping technique[3,6] predicted a zero-field instanton action given by Eq.(14). To verify the S-dependence of zero-field instanton action, a semi-log plot of zero-field splitting for integer S(ranging from 3 to 12) versus S is shown in Fig.8. It is found that the curve can be best fitted by either $Y = -2.3S + 1.18133$ or $Y = -2.994\sqrt{S(S+1)} + 2.94$. By ignoring the S-dependence of the prefactor(which is of order $\log S$),[10,17] numerical result seems to suggest that Eq. (14) is better.

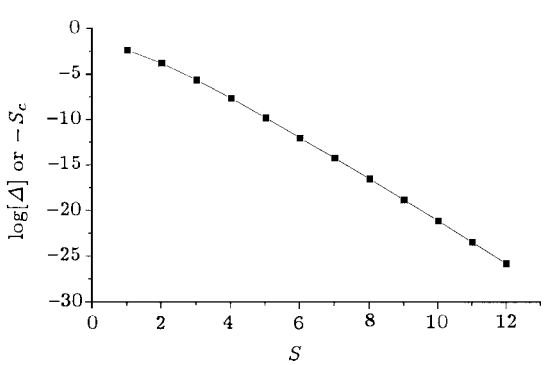


Fig.8. Semi-log plot for zero-field splitting versus S.

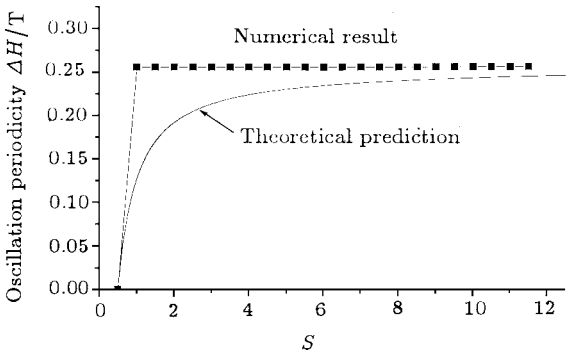


Fig.9. Illustration for the S-dependent oscillation periodicity.

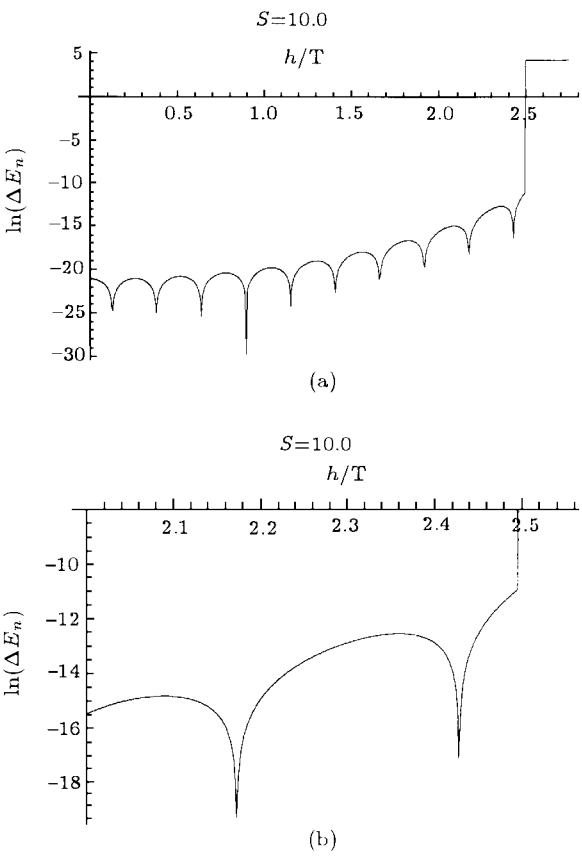


Fig.10. Tunneling splitting in high field range of S=10 in semi-log plot. (a) global behavior; (b) details for the field closed to h_2.

Up to now, theoretical analysis is based on the large spin limit, made by either using the classical energy to replace the "quantum energy",[6,17] or by the mapping technique.[10] Such a theoretical analysis is not a perfect one since it gives incorrect prediction for S=1/2. Recently theoretical analysis by employing the quantum energy without large spin limit is developed and an S-dependent periodicity is found[18]

$$\Delta H_1 = \frac{S-1/2}{S}\Delta H. \tag{19}$$

Here ΔH is given by Eq.(15). Numerical result for the S-dependence of the oscillation periodicity is shown in Fig.9. Surprisingly the oscillation periodicity keeps fixed for S ranging from 1 to 14 but drops to zero for S=1/2 as it should be. This result shows that the large spin limit works well for S around 10.

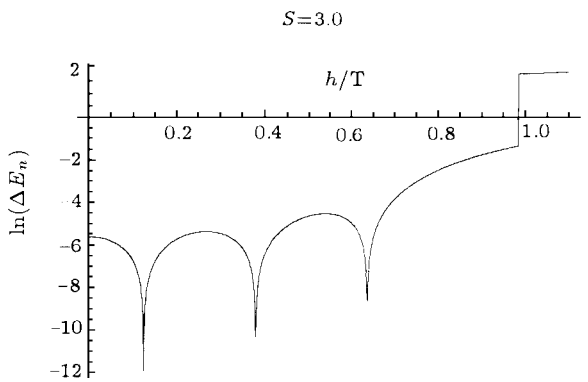


Fig.11. Tunneling splitting in high field range of S=3 in semi-log plot.

Recently, theoretical study on tunneling splitting in high field range predicted[11] a new quantum interference phenomenon associated with it: at a certain field,

$$h_2 = (1-\lambda)^{1/2} h_c, \quad h_c = 2S(D+E)/(g\mu_B),$$

the dependence of tunneling splitting switches from oscillation to a "monotonic growth". This prediction can be tested by numerical calculation. It is found that, for $S > 6.5$, there is no "monotonic growth", instead we find a sharp increase of energy splitting followed by a level off. Typical result for S=10 is shown in Fig.10. For $S \leq 6$, we find "monotonic growth" beginning at the field very close to h_2, but the range of the field for "monotonic growth" is much narrower than the predicted(i.e., $h_2 < h < h_c$) before the sharp increase appears. Typical result is shown in Fig.11 for S=3. Further study is needed for the explanation of this high-field behavior.

ACKNOWLEDGMENTS

We are grateful to Prof. Pu Fu-Cho and Prof. Liang Jiu-Qing for the fruitful discussion.

REFERENCES

[1] L. Gunther, B. Barbara. Quantum Tunneling of Magnetization –QTM'94 (Kluwer, Dordrecht, 1995).

[2] A. J. Leggett, S. Chakraverty, A. T. Dorsey, M. P. A. Fisher, A. Garg, W. Zwerger, *Rev. Mod. Phys.,* **59** (1987),1.

[3] E. M. Chunovsky, L. Gunther, *Phys. Rev. Lett.,* **60** (1988),661.

[4] D. Loss, D. P. Divincenzo, G. Grinstein, *Phys. Rev. Lett.,* **69**(1992),3232.

[5] J. V. Delft, C. L. Henley, *Phys. Rev. Lett.,* **69**(1992),3236.

[6] A. Garg, *Europhys. Lett.,* **22**(1993),205.

[7] J. -Q. Liang, Y. B. Zhang, H. J. W. Müller-kirstern, Jian-Ge Zhou, Z. Zimmerschied, F. -C. Pu, *Phys. Rev.,* **B57**(1998),529.

[8] E. M. Chudnovsky, D. P. Divincenzo, *Phys. Rev.,* **48** (1993),10548.

[9] W. Wernsdorfer, R. Sessoli, *Science,* **284**(1999),133.

[10] S. P. Kou, J. -Q. Liang, Y. B. Zhang, F. C. Pu, *Phys. Rev.,* **B59**(1999),11792.

[11] E. M. Chundnovsky, X. M. Hidalgo, cond-mat/9902218.

[12] A. Garg, cond-mat/0003114.

[13] J. -Q. Liang, H. J. W. Müller-Kirstern, D. K. Park, F. -C. Pu, *Phys. Rev.,* **B61**(2000),8856.

[14] Y. B. Zhang, J. -Q. Liang, H. J. W. Müller-Kirstern, S. -P. Kou, Z. Zimmerschied, F. -C. Pu, *Phys. Rev.,* **B60**(1999),12886.

[15] M. Enz, R. Schilling, *J. Phys.,* **C19**(1988),1765. M. Enz, R. Schilling, *J. Phys.,* **C19**(1988),L711.

[16] J. -Q. Liang, H. J. W. Müller-Kirsten, J. G. Zhou, Z. Zimmerschied, F. C. Pu, *Phys. Lett.,* **B393**(1997),368.

[17] A. Garg, G-H Kim, *Phys. Rev.,* **B45**(1992),12921.