Vol 17 No 4, April 2008
1674-1056/2008/17(04)/1436-07

Chinese Physics B

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Dephasing of quantum tunnelling in molecular nanomagnets*

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(Received 14 September 2007; revised manuscript received 14 October 2007)

Dephasing mechanism of quantum tunnelling in molecular magnets has been studied by means of the spin-coherent-state path integral in a mean field approximation. It is found that the fluctuating uncompensated transverse field from the dipolar-dipolar interaction between molecular magnets contributes a random phase to the quantum interference phase. The resulting transition rate is determined by the average tunnel splitting over the random phase. Such a dephasing process leads to the suppression of quenching due to the quantum phase interference, and to the steps due to odd resonances in hysteresis loop survived, which is in good agreement with experimental observations in molecular nanomagnets Fe_8 and Mn_{12}.

Keywords: single molecule magnet, quantum tunnelling, dephasing
PACC: 7550X, 7340E, 7535C

1. Introduction

Macroscopic quantum coherence (MQC) in molecular nanomagnets has become an attractive subject in recent years.[1−11] MQC of quantum tunnelling due to interference of symmetric tunnelling paths was predicted for isolated magnetic molecules with different symmetries.[3,4,6,9] One characteristic of MQC is the oscillation of tunnel splitting with respect to the transverse magnetic field along the hard axis. Such a kind of oscillation was observed in several molecular nanomagnets such as Fe_8[3] and Mn_{12}.[3,10] Nevertheless, it is found that theoretically, quenching of quantum phase interference is not complete in oscillation experiments.[3,10,11] Steps due to the odd resonances are observed in hysteresis measurements of Fe_8 and Mn_{12},[3,9] which is in contradiction with the theoretical prediction. This indicates that there exists a dephasing mechanism to suppress the quenching from the quantum phase interference. In fact, decoherence from the coupling between the isolated magnetic molecule and its environment is regarded as an important subject, especially as molecular magnets may be used as possible elements to develop quantum computer.[12−14]

Essentially, there are two kinds of coupling relevant for decoherence in molecular magnets. One of them, the spin-phonon interaction with dissipation,[8] can be safely ignored at very low temperatures since the spin-lattice relaxation time becomes extremely long.[15] Another one, the spin-spin interaction, including the dipolar interaction between the spin and the nuclear spin and dipolar interaction between spins, is responsible for dephasing at low temperatures.[3,10,12] Although the dephase mechanism is discussed extensively, a concrete example or calculation still lacks. Since the tunnel splitting in Fe_8 and Mn_{12} is very small, the dipolar interaction between spins will force most of spins off resonance[16] and the spins cannot relax continuously without a 'fluctuating field' Such a 'fluctuating field' can either be the fast-fluctuating field of hyperfine origin to broaden the resonance effectively,[16] or a sweeping field along the easy axis.[3] Physically the effect of the 'fluctuating field' is to compensate the biased field on the easy axis by the dipolar interaction between spins and to bring more spins into resonance. However, the transverse field component from the dipolar interaction is not compensated during resonant tunnelling. We know that the quantum interference phase depends on the transverse field experienced by the spin in resonance,[4−6] and the uncompensated fluctuating transverse field from the dipolar interaction can be the source of dephasing. This is the key issue to be

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addressed in the present paper. In the following, we shall take the Fe_8 system as an example to illustrate the dephasing mechanism due to the uncompensated fluctuating transverse field. The mechanism can be generalized to other molecular magnets.

2. Dephasing in biaxial spin models

We start with the biaxial model for Fe_8 systems[4−6]

$$H = K_1 S_z^2 + K_2 S_y^2 - g\mu_{\rm B}(B_z + h_z)S_z, \qquad (1)$$

where $K_1 > K_2 > 0$ and B_z is the magnetic field along the hard axis and h_z is the uncompensated fluctuating transverse field. Both experimental and the Monte Carlo study[17−19] show that the internal field h_z created by the dipolar interaction has a random distribution. Here h_z is assumed to have a Gaussian distribution,

$$p(h_z) = \frac{1}{(2\pi\sigma^2)^{1/2}} \exp\left[-\frac{(h_z - h_0)^2}{2\sigma^2}\right], \qquad (2)$$

where both σ and h_0 depend on the way of preparing the initial state.[17−19] A typical value of the distribution width for Fe_8 is about 0.05T.[16,17] Accordingly, in what follows we assume that h_z is too small to affect the tunnelling barrier (and thus the instanton action and pre-factor). To illustrate how such a weak transverse field leads to dephasing in quantum tunnelling, the tunnel splitting in the presence of fluctuating transverse field will be calculated by the instanton method, and then the resulting transition rate will be presented.

An efficient way to deal with the quantum tunnelling in spin systems is the spin-coherent-state path integral.[4−6] The tunnelling amplitude from the initial state $|n_{\rm i}\rangle$ to the terminal state $|n_{\rm f}\rangle$ is written as

$$\begin{aligned}\mathcal{K}(n_{\rm f}, T/2; n_{\rm i}, -T/2) &= \langle n_{\rm f}|\, {\rm e}^{{\rm i}HT/\hbar}\, |n_{\rm i}\rangle \\ &= \int {\rm d}\Omega \exp\left[\frac{\rm i}{\hbar}\int_{-T/2}^{T/2} L\ {\rm d}t\right], \qquad (3)\end{aligned}$$

where the Lagrangian in the large spin approximation is given by

$$L = -s\hbar(1 - \cos\theta)\dot{\phi} - E(\theta, \phi). \qquad (4)$$

$E(\theta, \phi)$ is the anisotropic energy. By utilizing the mapping technique, $(\phi, \hbar\, s\cos\theta)$ are treated as canonical variables.[6] The propagator in Eq.(3) is equivalent to a propagator of a particle in a double-well potential with an effective Lagrangian

$$L_{\rm eff}(\phi, \dot{\phi}) = \frac{1}{2} m(\phi)\dot{\phi}^2 - V(\phi) - \Theta(\phi)\dot{\phi}, \qquad (5)$$

where

$$V(\phi) = K_2 s^2 \sin^2\phi - \frac{[g\mu_{\rm B}(B_z + h_z)]^2 \lambda\ \sin^2\phi}{4K_1(1 - \lambda\ \sin^2\phi)}, \qquad (6)$$

$\Theta(\phi) = s - g\mu_{\rm B}(B_z + h_z)m(\phi)$, and $m(\phi) = 1/2K_1(1 - \lambda\sin^2\phi)$ with $\lambda = K_2/K_1$. Following the instanton method,[4−6] the ground state tunnel splitting is found to be

$$\Delta_0 = Q{\rm e}^{-S_{\rm c}} |\cos[s\pi - d(B_z + h_z)]|, \qquad (7)$$

where $d = g\mu_{\rm B}\pi/(2K_1\sqrt{1-\lambda})$, the instanton action $S_{\rm c}$

$$S_{\rm c} = s\lambda^{1/2}\int_0^{\pi} \frac{\sin\phi(1 - u^2 - \lambda\sin^2\phi)^{1/2}}{1 - \lambda\sin^2\phi}{\rm d}\phi, \qquad (8)$$

and the pre-factor Q

$$\begin{aligned}Q =& 16s\sqrt{K_1 K_2}\left(\frac{\sqrt{1-u^2} - \sqrt{\lambda}}{\sqrt{1-u^2} + \sqrt{\lambda}}\right)^c \\ &\times (1 - u^2)\sqrt{1 - u^2 - \lambda}, \qquad (9)\end{aligned}$$

with $c = \frac{1}{2}\sqrt{\lambda(1-u^2)}$ and $u = g\mu_{\rm B}(B_z + h_z)/(2K_1 s)$.

The calculation of tunnel splitting in the excited states and resonant tunnelling by using the instanton method is more complicated than that in the ground state.[6] One needs to use the Bohr–Summerfeld quantization rule $\oint p{\rm d}x = \oint s\cos\theta{\rm d}\phi = nh$ (n is a non-negative integer) to define the classical degenerate excited states (i.e. the classical periodic orbits). Then the total tunnelling amplitude can be obtained by integrating over the classical degenerate orbits.[6] Tunnel splitting for the nth excited state is found to be

$$\Delta E_n = Q_n {\rm e}^{-S_{\rm cn}} |\cos[(s - n)\pi - d(B_z + h_z)]|, \qquad (10)$$

where

$$Q_n^{-1} = \frac{1}{4}\int_{-\phi_n}^{\phi_n} {\rm d}\phi \left\{\frac{1}{2} m(\phi)/[E_n - V(\phi)]\right\}^{1/2} \qquad (11)$$

and

$$S_{\rm cn} = \int_{\phi_n}^{\pi - \phi_n} {\rm d}\phi \sqrt{2m(\phi)[V(\phi) - E_n]}. \qquad (12)$$

E_n and ϕ_n are the energy and the turning point of the quantized periodic orbits, respectively. In the case of resonant tunnelling, the Hamiltonian is written as

$$H_{\rm r} = K_1 S_z^2 + K_2 S_y^2 - g\mu_{\rm B}(B_z + h_z)S_z - g\mu_{\rm B} B_x S_x, \qquad (13)$$

where B_x is the applied field along the easy axis. In this model the spin almost aligns within the $x-y$ plane provided that the transverse field B_z is not very strong. Thus it is safe to take approximately $\sin\theta = \sqrt{1-\cos^2\theta} \simeq 1 - \frac{1}{2}\cos^2\theta$. In this way one can follow the mapping and quantization scheme as that for the excited state.[6] The effective Lagrangian has the same expression as that in Eq.(5), while the mass and the potential are now given by

$$m_1(\phi) = \left[2K_1(1-\lambda\sin^2\phi) + (g\mu_{\rm B}B_x/s)\cos\phi\right]^{-1},$$
$$V_1(\phi) = V(\phi) - g\mu_{\rm B}B_x s\cos\phi. \quad (14)$$

By tuning B_x to satisfy the resonance condition $B_x = n_{\rm r}\Delta B$ where $\Delta B \approx (K_1+K_2)/2g\mu_{\rm B}$ is the field interval between two neighbouring resonant tunnelling, the classical resonance happens between the mth quantized excited state of one well and the nth level in the other well, namely $E_m^1 = E_n^2$ with $|m-n| = n_{\rm r}$. Following the calculation for excited state tunnelling, the tunnel splitting in this case is obtained:

$$\Delta_{n_{\rm r}} = 4q^2{\rm e}^{-S_{\rm cr}}|\cos[(s-n_{\rm r}/2)\pi - d_{n_{\rm r}}(B_z+h_z)]|, \quad (15)$$

where

$$d_{n_{\rm r}} = \frac{g\mu_{\rm B}}{2K_1}\int_0^{\pi}\frac{{\rm d}\phi}{1-\lambda\sin^2\phi - \dfrac{g\mu_{\rm B}}{2K_2 s}\, n_{\rm r}\Delta B\cos\phi}, \quad (16)$$

$$S_{\rm cr} = \int_{\phi_m}^{\pi-\phi_n}{\rm d}\phi\left\{2m_1(\phi)[V_1(\phi)-E_m^1]\right\}^{1/2}$$

and

$$q^{-2} = \int_{-\phi_m}^{\phi_m}{\rm d}\phi\left\{\frac{1}{2}m_1(\phi)/[E_m^1 - V_1(\phi)]\right\}^{1/2};$$

$\pm\phi_m$ and $\pi\pm\phi_n$ are the turning points of the mth quantized classical orbits in one well and of the nth quantized classical orbits in the other well, respectively. Equation (15) tells us that there is a $\pi/2$ shift for the oscillation of odd and even resonance, and for an integer s, $\Delta_{n_{\rm r}} = 0$ for $n_{\rm r} = 1,3,5,\cdots$ when $B_z = h_z = 0$. Namely, the odd resonance is quenched due to the quantum phase interference.

As one can see from Eqs.(7), (10) and (15), the main contribution from the fluctuating uncompensated transverse field h_z is a random phase dh_z in the quantum interference phase. This implies that the quantum interference phases of the tunnelling spins inside the resonance window are not the same. Instead, there is a distribution. Such a distribution of random phases is responsible for dephasing as we shall illustrate in the following. The relation between the tunnel splitting and the transition rate depends on the nature of the 'fluctuating field' which maintains the continuous relaxation. If the 'fluctuating field' is the sweeping field on the easy axis, the coherent spin transition rate will depend on tunnel splitting by the Landau–Zener transition formula[3,8]

$$P_{\rm LZ} = 1 - \exp\left[-\frac{\pi\Delta_{n_{\rm r}}^2}{2g\mu_{\rm B}\hbar(2s-n_{\rm r}){\rm d}B_x/{\rm d}t}\right]$$
$$\simeq \frac{\pi\Delta_{n_{\rm r}}^2}{2g\mu_{\rm B}\hbar(2s-n_{\rm r}){\rm d}B_x/{\rm d}t}, \quad (17)$$

where ${\rm d}B_x/{\rm d}t$ is the sweeping rate; and we have used the low transition rate approximation, namely $P_{\rm LZ} \ll 1$ as for Fe_8 system.[22] On the other hand, due to the dipolar interaction of hyperfine origin, the incoherent transition rate is given by[16]

$$P_{\rm i} = \Delta_{n_{\rm r}}^2\Gamma, \quad (18)$$

where Γ is determined by the nuclear spin dynamics. In both cases, a quenching of tunnel splitting means zero transition rate, $P_{\rm LZ}(\Delta_{n_{\rm r}}=0) = P_{\rm i}(\Delta_{n_{\rm r}}=0) = 0$. Nevertheless, the above formula are based on the assumption that all the spins tunnel with the same tunnel splitting inside the resonance window. This is obviously not true when the fluctuating uncompensated transverse field is included. As one can see from the above calculation, the tunnel splitting always depends on h_z. This implies that the spins will tunnel with different tunnel splitting inside the resonance window. Accordingly, the resulting transition rate should be (at low transition rate)

$$\langle P_{\rm LZ}\rangle \simeq \frac{\pi\langle\Delta_{n_{\rm r}}^2\rangle}{2g\mu_{\rm B}\hbar(2s-n_{\rm r}){\rm d}B_x/{\rm d}t} \quad (19)$$

or

$$\langle P_{\rm i}\rangle = \left\langle\Delta_{n_{\rm r}}^2\right\rangle_{h_z}\Gamma, \quad (20)$$

where $\langle\cdots\rangle_{h_z}$ represents the average over the distribution of h_z. Consequently, the resulting transition rate depends on $\left\langle\Delta_{n_{\rm r}}^2\right\rangle_{h_z}$ rather than $\Delta_{n_{\rm r}}$. Using the result we have obtained, it is easy to show that there is no quenching point of $\left\langle\Delta_{n_{\rm r}}^2\right\rangle_{h_z}$. Taking the ground state tunnelling as example, one can find that

$$\left\langle\Delta_0^2\right\rangle_{h_z} \simeq Q_0^2{\rm e}^{-2S_{\rm c}^0}\left\langle\cos^2[s\pi - {\rm d}(B_z+h_z)]\right\rangle_{h_z}$$
$$= \frac{1}{2}Q_0^2{\rm e}^{-2S_{\rm c}^0}\{{\rm e}^{-2\sigma^2 d^2}\cos[2(s\pi - dh_0 - dB_z)] + 1\}, \quad (21)$$

where we have made use of the assumption that h_z is too small to affect the tunnelling amplitude. $Q_0 = Q(h_z=0)$, and $S_{\rm c}^0 = S_{\rm c}(h_z=0)$. In the case of

$\sigma = h_0 = 0$, the above result is reduced to the original one[4−6]

$$\Delta_0^2 = Q_0^2 \mathrm{e}^{-2S_{\mathrm{c}}^0} \cos^2(s\pi - dB_z). \tag{22}$$

Qualitative difference between Δ_0^2 and $\langle \Delta_0^2 \rangle_{h_z}$ becomes obvious by comparing Eq.(21) with Eq.(22). From Eq.(22), one can see the quenching points of Δ_0^2 are determined by $s\pi - dB_z = k\pi + \pi/2$ ($k = 0, 1, 2, \cdots$), which represents the spin parity effect in the case of $B_z = 0$. Nevertheless, Eq.(22) tells us that at the quenching points (we set $h_0 = 0$ for simplicity)

$$\langle \Delta_0^2 \rangle_{h_z}^q \simeq \frac{1}{2} Q_0^2 \mathrm{e}^{-2S_{\mathrm{c}}^0} (1 - \mathrm{e}^{-2\sigma^2 d^2}) \neq 0. \tag{23}$$

This implies that, under the zero-field condition, the transition rate for molecular magnets with half-integer spin is not zero, and thus the spin parity effect cannot be directly observed in a relaxation experiment.[11] How does this happen? The answer is the dephasing. As we have mentioned, the fluctuating uncompensated transverse field h_z leads to a random phase, while Eq.(21) shows that $\langle \Delta_0^2 \rangle_{h_z}$ is a mean value over the random phase. We can say that the phase-averaging process is a dephasing in quantum tunnelling.[20]

The situation for excited state tunnelling and resonant tunnelling is almost the same. One can find that

$$\langle \Delta_n^2 \rangle_{h_z} \simeq \frac{1}{2} Q_{n0}^2 \mathrm{e}^{-2S_{\mathrm{cn}}^0} \left\{ \mathrm{e}^{-2\sigma^2 d^2} \cos 2\phi_n + 1 \right\}, \tag{24}$$

where $\phi_n = s\pi - n\pi - dh_0 - dB_z$, $Q_{n0} = Q_n(h_z = 0)$, $S_{\mathrm{cn}}^0 = S_{\mathrm{cn}}(h_z = 0)$ and

$$\langle \Delta_{n_{\mathrm{r}}}^2 \rangle_{h_z} \simeq 8 q_0^4 \mathrm{e}^{-2S_{\mathrm{cr}}^0} \left\{ \mathrm{e}^{-2\sigma^2 d^2} \cos 2\phi_{n_{\mathrm{r}}} + 1 \right\}, \tag{25}$$

with $\phi_{n_{\mathrm{r}}} = s\pi - n_{\mathrm{r}}\pi/2 - dh_0 - dB_z$, $q_0 = q(h_z = 0$, $S_{\mathrm{cr}}^0 = S_{\mathrm{cr}}(h_z = 0)$. For an integer-spin molecular magnet, Eq.(25) shows that for zero-field condition and odd-resonance we have

$$\langle \Delta_{n_{\mathrm{r}}}^2 \rangle_{h_z}^q = 8 c_0^4 \mathrm{e}^{-2S_{\mathrm{cr}}^0} (1 - \mathrm{e}^{-2\sigma^2 d^2}), \tag{26}$$

which leads to non-zero transition rate, and thus the steps due to the odd resonances survive in the hysteresis loops as observed in the experiments.[3,9]

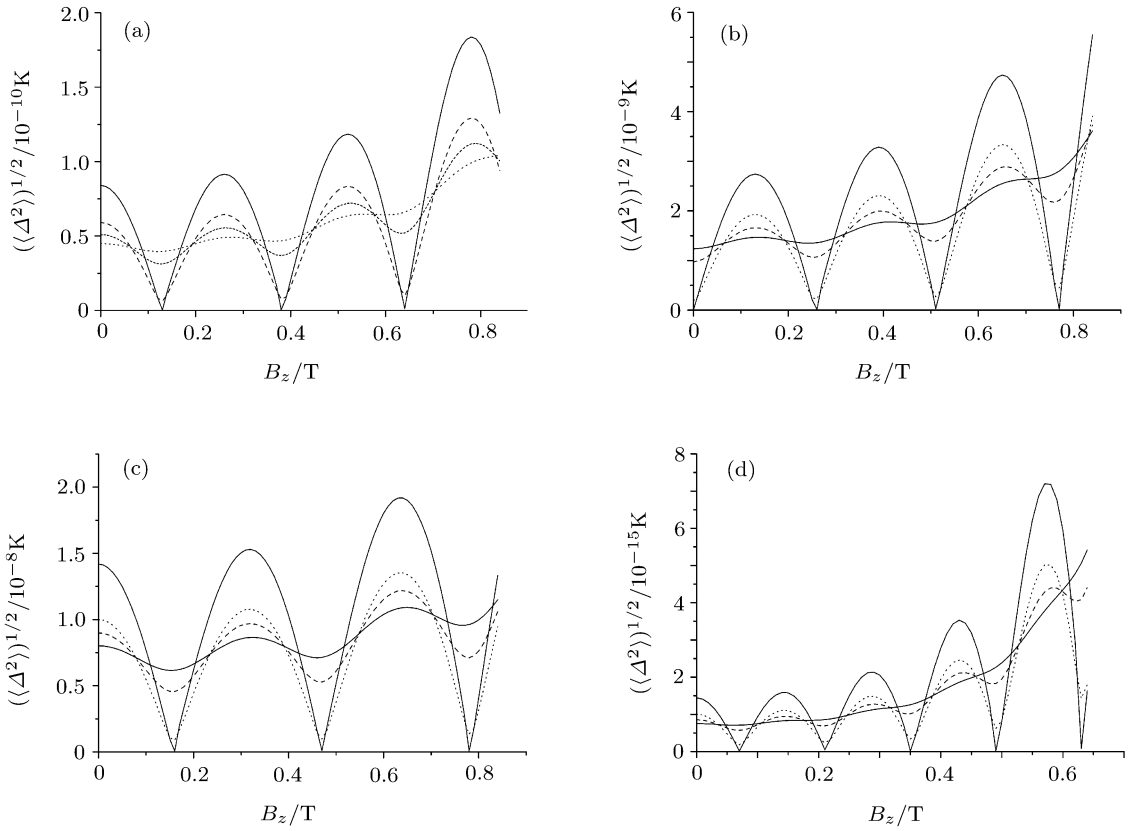


Fig.1. Illustration of dephasing effect on the oscillation of tunnel splitting. Anisotropic parameters are $K_1 = 0.321$K, $K_2 = 0.229$K, and $\lambda_0 = K_2/K_1$ for Fe_8 system. (a) s=10 and $\lambda = \lambda_0$; (b) s=9.5 and $\lambda = \lambda_0$; (c) s=10 and $\lambda = 0.8\lambda_0$; (d) s=10 and $\lambda = 1.28\lambda_0$.

Taking the anisotropic parameters for Fe_8, the dephasing effect due to the fluctuating random field is plotted in Fig.1. One can see that even for the fluctuating field with a distribution width as small as 0.8 mT (typically for the dipolar interaction of nuclear spins in Fe_8),[16,17] the quenching is not complete. The oscillation is suppressed further as the distribution width increases. When the distribution width approaches half of the oscillation period, the oscillation is suppressed totally, and the classical behaviour that tunnel splitting increases with the transverse field monotonously, is resumed. As one can see from Fig.1(b), the suppression of the oscillation also occurs for half-integer spin system. It is interesting to see that the suppression of the oscillation depends on λ critically. For Fe_8 systems ($\lambda_0 \simeq 0.739$) a distribution width as large as 0.11 T is needed to suppress the oscillation completely, while a distribution width of 0.05 T is enough to suppress the oscillation for a system with $\lambda = 1.28\lambda_0$. The averaged energy gap $\left\langle \Delta_n^2 \right\rangle^{1/2}$ decreases drastically with the increase of λ. It can be seen from Figs.1(a), (c), and (d), that the gap decreases about 7 to 8 orders of magnitude as λ increases from $\lambda = 0.8\lambda_0$ to $1.28\lambda_0$. This result predicts that the oscillation can be easily observed in biaxial systems with a smaller λ.

3. Dephasing in other systems

In this section, we show that the analytic theory of dephasing for biaxial spin model can be generalized to other molecular magnets with more complicated Hamiltonians. The first example will be the biaxial spin model with higher order terms[3,9]

$$H_1 = K_1 S_z^2 + K_2 S_y^2 + C(S_-^4 + S_+^4) - g\mu_{\rm B} S_z (B_z + h_z). \tag{27}$$

The analytic expression of tunnel splitting for this Hamiltonian has not yet been obtained. However, numerical diagonalization shows that tunnel splitting oscillates with the transverse field.[3,9] This implies that the tunnel splitting in this case can be expressed as

$$\Delta E = \Delta E_0 \{\cos[s\pi - D(B_z + h_z)]\}, \tag{28}$$

where $D \simeq 2\pi/0.41$.[3] In the same way, the magnitude of the fluctuating uncompensated transverse field h_z is too small to affect the tunnelling barrier, and the main contribution of h_z to the interference quantum phase is a random phase. The resulting transition rate is determined by

$$\left\langle \Delta E^2 \right\rangle_{h_z} \simeq \Delta E_0^2 \{ {\rm e}^{-2\sigma^2 d^2} \cos[2(s\pi - Dh_0 - DB_z)] + 1\}, \tag{29}$$

which is averaged over the random phase. Again, such a dephasing process will suppress the quenching due to quantum phase interference. As a matter of fact, the dephasing mechanism is expected to work in molecular magnets in which the quantum phase interference will lead to the oscillation of tunnel splitting with respect to the transverse field. Another known example is the uniaxial spin model for Mn_{12},[9,21]

$$H_2 = -ES_z^2 - C_1 S_z^4 + C_2(S_-^4 + S_+^4) - g\mu_{\rm B} S_x (B_x + h_x), \tag{30}$$

where E and $C_{1,2}$ are anisotropic parameters. Numerical solution[9] shows that tunnel splitting will oscillate with the transverse field B_x in absence of h_x. The key point is that once the MQC leads to oscillation in isolated molecular magnets, the expression of tunnel splitting will be of the form in Eq.(28) which means the quantum interference phase depends on the transverse field explicitly. Inevitably the inclusion of the fluctuating uncompensated transverse field $h_{\rm t}$ leads to a random phase in the interference phase, and then the transition rate is given in Eqs.(19) or (20) rather than Eqs.(17) or (18). In other word, ΔE^2 will be replaced by $\left\langle \Delta E^2 \right\rangle_{h_{\rm t}}$ which can be simply expressed as the following replacement:

$$\cos^2(s\pi - DB_{\rm t}) \to \left\langle \cos^2[s\pi - D(B_{\rm t} + h_{\rm t})] \right\rangle_{h_{\rm t}}. \tag{31}$$

Such a replacement leads to an average over the random phase. Thus the dephasing happens and quenching due to the quantum phase interference is suppressed.

4. Conclusion and discussion

In the previous sections, we explore the dephasing mechanism of quantum tunnelling in molecular magnets, and conclude that the fluctuating uncompensated transverse field from the dipolar-dipolar interaction between molecular magnets contributes a random phase to the quantum interference phase. The resulting transition rate is determined by the average tunnel splitting over the random phase. Such a dephasing process leads to the suppression of quenching from quantum phase interference, and thus the steps

due to odd resonance survive in the hysteresis measurements. The dephasing mechanism may work in molecular magnets having MQC.

In the present paper, the effect of the uncompensated transverse field is treated in the mean field approximation, and hence the dynamics of the environment is not included.[12] Although the transverse field is regarded as 'fluctuating' inside the resonance window, the distribution of the transverse field is frozen. This implies that our theory is valid within the time scale when the relaxation due to tunnelling begins to affect the distribution of the internal field from the dipolar-dipolar interaction. For the relaxation using a sweeping field, this is also the basis for the Landau–Zener model.[22] If the spin begins to experience the change of the internal field of dipolar-dipolar interaction, the Landau–Zener model is not valid any more. One needs to consider other effects such as the nonlinear Landau–Zener model[23] or the 'hole-digging' mechanism.[16,17] For the relaxation mediated by fluctuating nuclear spins, our theory is expected to be valid in a short time scale. However, it should be noted that the essence of our theory is the spins tunnel with different transverse fields inside the resonance window. In the calculation we choose a Gaussian distribution for the random field. The main conclusion is not modified by choosing other distribution forms of the random field. Consequently, the dephasing mechanism is valid for all time scale, however the transition rate will be modified slightly. On the other hand, there may exist some kind of interactions which will lead to the transverse field fluctuation in the time scale shorter than the tunnelling time scale. In this case, we should make the average over the tunnelling propagator. Take the ground state tunnelling in the biaxial spin model in Eq.(1) as an example. The one instanton (or anti-instanton) propagator can be written as

$$\mathcal{K}_1^{\pm} = I\mathrm{e}^{S_\mathrm{c}\pm\mathrm{i}[s\pi-d(B_z+h_z)]}, \tag{32}$$

where I is the fluctuation functional integral.[4–6] By taking the same assumption, one finds

$$\left\langle \mathcal{K}_1^{\pm} \right\rangle_{h_z} \simeq I\mathrm{e}^{-S_\mathrm{c}^0-d^2\sigma^2/2}\mathrm{e}^{\mathrm{i}\pm[s\pi-d(B_z+h_0)]}. \tag{33}$$

The corresponding tunnel splitting is

$$\langle \Delta_0 \rangle = Q_0\mathrm{e}^{-S_\mathrm{c}^0-d^2\sigma^2/2}|\cos[s\pi - d(B_z+h_0)]|. \tag{34}$$

By using the typical parameters for Fe_8, the modification is omissible for unbiased fluctuation (i.e. $h_0 = 0$). Of course, the complete treatment of the transverse field needs to include the component of the field on the medium axis. As we have known, the component on the medium axis will destroy the symmetry of the tunnelling paths and thus suppress the interference.[5] Although the inclusion of the component on the medium axis will make the solution more complicated, it will not modify the key conclusion. Further analysis will be presented elsewhere.

Before ending this paper, we draw an analogy between the dephasing process and the Young's double-slit interference experiment in order to make the dephasing picture more clear. Physically the two symmetric tunnelling paths can be regarded as the two paths through the double slits. Under an ideal condition, the molecular magnet is isolated, and hence the interference phase from the source (here it is the initial state $|n_\mathrm{i}\rangle$) to the point in the screen (here it is the terminal state $|n_\mathrm{f}\rangle$) is unique. This corresponds to the ideal condition when two slits are geometric lines. However, when the fluctuating uncompensated transverse field is involved, the interference phase is not unique. Instead, there exists a distribution for the interference phases. Correspondingly the two slits are not geometric lines but with a finite width. The finite-width of the double-slit smears the interference pattern just like the finite distribution of transverse field will suppress the oscillation due to quantum interference.

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