Tunneling in a single-molecule magnet via anisotropic exchange interactions

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The model describing two coupled single-molecule magnets is considered by including anisotropic exchange interaction in the spin Hamiltonian. The tunnel splittings are calculated perturbationally for arbitrary spin in several selected cases and estimated numerically by using a Mn_4-dbm as a model system. It is found that the anisotropic exchange interaction plays an important role in the level splittings such as spin-spin cross relaxation and shifts of the resonant field. The results are discussed in comparison with the recent experiment.

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Molecular magnets and the quantum properties they show are an extremely topical area. There are two major reasons for studying molecular magnets: the search for macroscopic quantum phenomena[1] and the possible use as a qubit for quantum computer.[2] Mn_{12} acetate $[Mn_{12}O_{12}(CH_3COO)_{16}(H_2O)_4]$ is the first case of a macroscopic realization of resonant magnetization tunneling in a single-molecule magnet (SMM)[3] and Fe_8 $[[Fe_8O_2(OH)_{12}(tacn)_6]^{8+}]$ is the first one to exhibit quantum phase interference.[4] Many efforts have been made to understand their mechanism by considering crystals of SMM's as consisting of giant spins interacting with environmental degree of freedom such as phonon, dipolar field, and nuclear spin.[5]

Noting that molecular magnets consists of many SMM's, there is the possibility of an exchange interaction that depends on the distance and nonmagnetic atoms in the exchange pathway. Until now, such an intermolecular exchange interaction has been assumed to be negligibly small. Recently, however, it has been reported[6,7] that in SMM's such as $[Mn_4O_3Cl_4(O_2CEt)_3(py)_3]$ (hereafter Mn_4-py) and $[Mn_4O_3(OSiMe_3)(OAc)_3(dbm)]$ (hereafter Mn_4-dbm) exchange interactions lead to a significant influence on the quantum properties of SMM's. This intermolecular exchange interaction was used to couple ferromagnetically or antiferromagnetically two SMM's, each acting as a bias on its neighbor, resulting in quantum behavior different from that of individual SMM's. In this respect it will be very interesting to study a quantum process generated by pairs of SMM's which are coupled by exchange interactions.

Theoretical calculations about magnetization tunneling with exchange interaction have been performed by several groups.[8] Considering a small antiferromagnetic grain having two collinear ferromagnetic sublattices whose magnetization are coupled by the exchange interaction, they obtained the tunneling rate of the magnetic moment due to the noncompensation of two sublattices by using the spin-coherent state path integral method. However, since their results were confined to the ground state tunneling with large spin, they are not considered as sufficient to apply to molecular magnetic systems with exchange interactions. Very recently, the author[9] dealt with the quantum tunneling of magnetization in such molecular magnetic systems by employing the perturbation method[10–12] and presented the level splitting of magnetization tunneling in a system of identical, antiferromagnetically coupled dimers with isotropic exchange interaction, as is likely to be the case in recent experiment.[6] For typical values of the anisotropy constant and the exchange coupling constant in Mn_4-py it was found that the level splitting only induced by the transverse exchange interaction is much smaller than the one induced by the transverse anisotropy and the transverse field and such exchange interaction leads to resonant field shifts. In this work we will extend the situation to the tunnel transitions for two ferromagnetically or antiferromagnetically coupled SMM's biased by the anisotropic exchange interaction and find a more crucial role of the exchange interaction in quantum tunneling of such SMM's. Selecting typical level crossings, the tunnel splittings will be presented in the presence of longitudinal or transverse field. This problem is especially important in the light of recent experimental evidence of a collective quantum process, called spin-spin cross relaxation (SSCR).[7] Focusing on the steplike features in the hysteresis loop of the molecular magnet Mn_4-dbm, we clarify the main sources of the tunnel transitions in the anticrossing points and estimate the order of magnitude of the level splittings in the low temperature regime. Furthermore, a quantitative comparison of the results with the experimental ones allows us to extract intrinsic transverse field and transverse exchange interaction in Mn_4-dbm.

The Hamiltonian which is the simplest model describing the spin system of an isolated SMM is written in the form

$$\mathcal{H}_i = -D\hat{S}_{iz}^2 + \mathcal{H}_i^{\text{trans}} - H_z\hat{S}_{iz}, \tag{1}$$

where the index $i(=1 \text{ or } 2)$ labels different SMM's, D is the longitudinal anisotropy constant, $\mathcal{H}_i^{\text{trans}}$ containing $\hat{S}_{x,i}$ or $\hat{S}_{y,i}$ spin operators, gives the transverse anisotropy or field which is small compared to $D\hat{S}_{z,i}^2$ in SMMs and the last term describes the Zeeman energy associated with an applied field. H_z stands for $g\mu_B H_z$ where g is the electronic g factor and μ_B is the Bohr magneton. Henceforth, we will usually drop the combination $g\mu_B$ for better readability of the formula.

Considering the anisotropic exchange interaction between two coupled SMM's, the spin Hamiltonian is represented as

$$\mathcal{H}=\sum_{i=1}^{2}\mathcal{H}_i+\sum_{\alpha=x,y,z}J_\alpha\hat{S}_{1\alpha}\hat{S}_{2\alpha}, \tag{2}$$

where two SMM's are coupled by anisotropic exchange interaction with $|J_x|\neq|J_y|\neq 0$. Without the transverse terms in the Hamiltonian (2), each state of the dimer is labeled by two quantum numbers (M_1,M_2) for two SMM's, with $M_1=S_1,\ S_1-1,\ldots,-S_1$ and $M_2=S_2,S_2-1,\ldots,-S_2$ and the energy level spectrum with $(2S_1+1)(2S_2+1)$ values becomes

$$E^{(0)}_{M_1,M_2}=-D(M_1^2+M_2^2)+J_zM_1M_2. \tag{3}$$

Therefore, as the field is applied along the easy axis $(\hat{z})$, the energy levels which correspond to the states with (M_1,M_2) and (M_1',M_2') cross at certain values of H_z given by

$$H_z^{(0)}=\frac{E^{(0)}_{M_1',M_2'}-E^{(0)}_{M_1,M_2}}{\sum_{i=1}^{2}(M_i'-M_i)}. \tag{4}$$

When the spin Hamiltonian (2) contains transverse terms, the level crossings can be avoided level crossings. The spins S_1 and S_2 are in resonance between two states when the local longitudinal field is close to an avoided level crossing. The energy gap, i.e., the tunnel splitting is determined by the terms perpendicular to the S_z such as the transverse exchange interaction, the transverse anisotropy and the transverse field. As the transverse terms are much smaller than the longitudinal ones, the tunnel splittings can be calculated by using the perturbation theory. In such cases, the tunnel splitting of the degenerate level pair (M_1,M_2) and (M_1',M_2') is represented as the shortest chain of matrix elements and energy denominators connecting the states $|M_1',M_2'\rangle$ and $|M_1,M_2\rangle$.

The first example,"model I," corresponds to the simplest case of tunnel splitting induced by the anisotropic exchange interaction

$$\mathcal{H}=-\sum_{i=1}^{2}D\hat{S}_{iz}^2+J_z\hat{S}_{1z}\hat{S}_{2z}+J_{+-}(\hat{S}_{1+}\hat{S}_{2-}+\hat{S}_{1-}\hat{S}_{2+})+J_{++}(\hat{S}_{1+}\hat{S}_{2+}+\hat{S}_{1-}\hat{S}_{2-}), \tag{5}$$

where $\hat{S}_{k\pm}=\hat{S}_{kx}\pm i\hat{S}_{ky}$ and $J_{+\pm}=(J_x\mp J_y)/4$. Considering the transverse exchange terms, we divide level splittings into two different types. For the exchange interaction with J_{+-} the tunnel splitting of the degenerate pair (M_1',M_2'), (M_1,M_2) appears only in the chain of matrix elements with connecting the states $|M_1'+k,M_2'-k\rangle$ and $|M_1'+k+1,M_2'-k-1\rangle$, where $M_1'=-M_1$, $M_2'=M_1>0$, $M_2=-M_1$, and k is an integer with $0\leqslant k\leqslant M_1-1-M_1'$. It corresponds to the level splitting of the degenerate pair $(-M_1,M_1)\rightarrow(M_1,-M_1)$. Taking $S_1=S_2(\equiv S)$ in the ensuing discussion, the level splitting of the degenerate pair becomes

$$\Delta E_{-M_1,M_1,M_1,-M_1}=2\prod_{p=-M_1}^{M_1\text{-}1}V^{(J_{+-})}_{p,-p,p+1,-p-1}\times\prod_{q=-M_1+1}^{M_1\text{-}1}(E^{(0)}_{q,-q}-E^{(0)}_{-M_1,M_1})^{-1}, \tag{6}$$

where

$$V^{(J_{+-})}_{p,-p,p+1,-p-1}=\langle p,-p|J_{+-}(\hat{S}_{1+}\hat{S}_{2-}+\hat{S}_{1-}\hat{S}_{2+})\times|p+1,-p-1\rangle=J_{+-}l_{p+1}l_{-p}, \tag{7}$$

with

$$l_M=\sqrt{(S+M)(S-M+1)}, \tag{8}$$

and $E^{(0)}_{q,-q}=-(2D+J_z)q^2$ are the unperturbed energy levels from Eq. (3). Calculating the product (6), we obtain the level splitting given by

$$\Delta E_{-M_1,M_1,M_1,-M_1}=2(2D+J_z)\left(\frac{J_{+-}}{2D+J_z}\right)^{2M_1}\left[\frac{(S+M_1)!}{(S-M_1)!(2M_1-1)!}\right]^2. \tag{9}$$

Next, for the exchange interaction with J_{++} the tunnel splitting consists of the chain of matrix elements with connecting the states $|-M_1+k,-M_1+k\rangle$ and $|-M_1+k+1,-M_1+k+1\rangle$ where k is an integer with $0\leqslant k\leqslant 2M_1-1$. The level splitting of the degenerate pair $(-M_1,-M_1)$ and (M_1,M_1) is calculated from the product

$$\Delta E_{-M_1,-M_1,M_1,M_1}=2\prod_{p=-M_1}^{M_1\text{-}1}V^{(J_{++})}_{p,p,p+1,p+1}\times\prod_{q=-M_1+1}^{M_1\text{-}1}(E^{(0)}_{q,q}-E^{(0)}_{-M_1,-M_1})^{-1}, \tag{10}$$

where

$$V^{(J_{++})}_{p,p,p+1,p+1}=\langle p,p|J_{++}(\hat{S}_{1+}\hat{S}_{2+}+\hat{S}_{1-}\hat{S}_{2-})|p+1,p+1\rangle=J_{++}l_{p+1}^2, \tag{11}$$

and thereby the corresponding tunnel splitting is given by

$$\Delta E_{-M_1,-M_1,M_1,M_1}=2(2D-J_z)\left(\frac{J_{++}}{2D-J_z}\right)^{2M_1}\left[\frac{(S+M_1)!}{(S-M_1)!(2M_1-1)!}\right]^2. \tag{12}$$

In the ground state $(M_1=S)$ the results (9) and (12) simplify to

$$\Delta E_{-S,\pm S,S,\mp S}=2(2D\pm J_z)(2S)^2\left(\frac{J_{+\mp}}{2D\pm J_z}\right)^{2S}. \quad (13)$$

For highly excited states with $S-M_1\gg 1$ and $M_1\gg 1$ we have the splitting given by

$$\Delta E_{-M_1,\pm M_1,M_1,\mp M_1}=\frac{2M_1}{\pi}(2D\pm J_z)\left[\left(\frac{J_{+\mp}}{2D\pm J_z}\right)\sqrt{\frac{S^2-M_1^2}{16M_1^4}}\right]^{2M_1}\times\left(\frac{S+M_1}{S-M_1}\right)^{2S+1}. \quad (14)$$

As a second example, "model II," let us consider the level splitting generated by the transverse exchange interaction with J_{++} and the transverse anisotropy in the xy plane

$$\mathcal{H}_i^{\text{trans}}=B(\hat{S}_{xi}^2-\hat{S}_{yi}^2). \quad (15)$$

Writing $\mathcal{H}_i^{\text{trans}}=\frac{1}{2}B(\hat{S}_{+i}^2+\hat{S}_{-i}^2)$ and choosing $i=2$, the level splitting is expected to appear in the degenerate pair of (M_1',M_2') and (M_1,M_2) where $M_1=M_1'+1$ and $M_2-(M_2'+1)$ is even number. In this situation the transverse exchange interaction should be included in the level splitting through the single perturbation step along the chain connecting the degenerate states. Hence, the corresponding level splitting becomes

$$\begin{aligned}\Delta E_{M_1',M_2',M_1,M_2}&=2V^{(J_{++})}_{M_1',M_2',M_1'+1,M_2'+1}\left(\prod_{p=M_2'+1}^{M_2\text{-}2}{}'\frac{V^{(B)}_{M_1,p,M_1,p+2}}{E_{M_1,p}-E_{M_1,M_2}}\right)\\&+2\sum_{k=M_2'+2}^{M_2\text{-}3}{}'\left[\left(\prod_{p_1=M_2'+2}^{k}{}'\frac{V^{(B)}_{M_1',p_1-2,M_1',p_1}}{E_{M_1',p_1}-E_{M_1M_2}}\right)V^{(J_{++})}_{M_1',k,M_1'+1,k+1}\left(\prod_{p_2=k+1}^{M_2\text{-}2}{}'\frac{V^{(B)}_{M_1,p_2,M_1,p_2+2}}{E_{M_1,p_2}-E_{M_1M_2}}\right)\right]\\&+2\left(\prod_{p=M_2'}^{M_2\text{-}3}{}'\frac{V^{(B)}_{M_1',p,M_1',p+2}}{E_{M_1',p+2}-E_{M_1,M_2}}\right)V^{(J_{++})}_{M_1-1,M_2-1,M_1,M_2},\end{aligned} \quad (16)$$

where Π' and Σ' increase in steps of 2, and the matrix element $V^{(J_{++})}_{M_1',M_2',M_1'+1,M_2'+1}$ is expressed as Eq. (11). Using the unperturbed energy level $E_{M_1,q}=-D(M_1^2+q^2)+J_zM_1q-H_z^{(0)}(M_1+q)$ with Eq. (4) for $H_z^{(0)}$ and

$$V^{(B)}_{M_1,p,M_1,p+2}=\langle M_1p|\frac{B}{2}\hat{S}_{-2}^2|M_1,p+2\rangle=\frac{B}{2}l_{p+1}l_{p+2}, \quad (17)$$

the formula for the level splitting reads

$$\Delta E_{M_1',M_2',M_1,M_2}=2J_{++}\left(\frac{B}{4D}\right)^{(M_2-M_2'-1)/2}g_{++}\sum_{k=M_2'}^{M_2\text{-}1}{}'\left[\frac{1}{(M_2-k-1)!!(k-M_2')!!}\frac{\Gamma\left(\frac{-k-a}{2}\right)\Gamma\left(\frac{k+b+1}{2}\right)}{\Gamma\left(\frac{-M_2'-a}{2}\right)\Gamma\left(\frac{M_2+b}{2}\right)}\right], \quad (18)$$

where $\Gamma(z)$ is the gamma function, $a=M_2'+(H_z^{(0)}-J_zM_1')/D$, $b=M_2+(H_z^{(0)}-J_zM_1)/D$, and

$$g_{++}=\left[(S+M_1'+1)(S-M_1')\frac{(S-M_2')!(S+M_2)!}{(S+M_2')!(S-M_2)!}\right]^{1/2}. \quad (19)$$

Our next example, "model III," is the level splitting generated by the transverse exchange interaction containing J_{+-} and the transverse anisotropy in the xy plane as Eq. (15). In this case, the level splitting is expected to appear in the degenerate pair of (M_1',M_2') and (M_1,M_2) where $M_2'=M_2+1$ and $M_1-(M_1'+1)$ is even number. In the same way as we have done in model II, the transverse exchange interaction containing J_{+-} should be included in the level splitting through the single perturbation step along the chain connecting the degenerate states. Then, the level splitting is given by

$$\Delta E_{M_1',M_2',M_1,M_2}=2V^{(J_{+-})}_{M_1',M_2',M_1'+1,M_2'-1}\left(\prod_{p=M_1'+1}^{M_1\text{-}2}{}'\frac{V^{(B)}_{p,M_2,p+2,M_2}}{E_{p,M_2}-E_{M_1,M_2}}\right)+2\sum_{k=M_1'+2}^{M_1\text{-}3}{}'\left[\left(\prod_{p_1=M_1'+2}^{k}{}'\frac{V^{(B)}_{p_1-2,M_2',p_1,M_2'}}{E_{p_1,M_2'}-E_{M_1M_2}}\right)V^{(J_{+-})}_{k,M_2',k+1,M_2'-1}\left(\prod_{p_2=k+1}^{M_1\text{-}2}{}'\frac{V^{(B)}_{p_2,M_2,p_2+2,M_2}}{E_{p_2,M_2}-E_{M_1M_2}}\right)\right]+2\left(\prod_{p=M_1'}^{M_1\text{-}3}{}'\frac{V^{(B)}_{p,M_2',p+2,M_2'}}{E_{p+2,M_2'}-E_{M_1,M_2}}\right)V^{(J_{+-})}_{M_1-1,M_2+1,M_1,M_2}. \tag{20}$$

Using the matrix elements (7) and (17) and the unperturbed energy level with the resonance field $H_z^{(0)}$, the resulting splitting is represented as

$$\Delta E_{M_1',M_2',M_1,M_2}=2J_{+-}\left(\frac{B}{4D}\right)^{(M_1-M_1'-1)/2}g_{+-}\sum_{k=M_1'}^{M_1\text{-}1}{}'\left[\frac{1}{(M_1-k-1)!!(k-M_1')!!}\frac{\Gamma\left(\frac{-k-d}{2}\right)\Gamma\left(\frac{k+c+1}{2}\right)}{\Gamma\left(\frac{-M_1'-d}{2}\right)\Gamma\left(\frac{M_1+c}{2}\right)}\right], \tag{21}$$

where $c=M_1+(H_z^{(0)}-J_zM_2)/D$, $d=M_1'+(H_z^{(0)}-J_zM_2')/D$, and

$$g_{+-}=\left[(S+M_2+1)(S-M_2)\frac{(S-M_1')!(S+M_1)!}{(S+M_1')!(S-M_1)!}\right]^{1/2}. \tag{22}$$

Let us consider as "model IV" the transverse exchange interaction containing J_{++} and the transverse field given by

$$\mathcal{H}^{\text{trans}}=-H_x(\hat{S}_{x1}+\hat{S}_{x2}), \tag{23}$$

where H_x can be either internal or external magnetic field. Using $\hat{S}_{xi}=(\hat{S}_{+i}+\hat{S}_{-i})/2$ and considering the case at $i=2$, the level splitting exists in the degenerate pair of (M_1',M_2') and (M_1,M_2) where $M_1=M_1'+1$ and $M_2-(M_2'+1)$ is odd number. In this situation the transverse exchange interaction containing J_{++} should be included in the level splitting through the single perturbation step along the chain connecting the degenerate states. Therefore, the corresponding level splitting is

$$\Delta E_{M_1',M_2',M_1,M_2}=2V^{(J_{++})}_{M_1',M_2',M_1'+1,M_2'+1}\left(\prod_{p=M_2'+1}^{M_2\text{-}1}\frac{V^{(H)}_{M_1,p,M_1,p+1}}{E_{M_1,p}-E_{M_1,M_2}}\right)+2\sum_{k=M_2'+1}^{M_2\text{-}2}\left[\left(\prod_{p_1=M_2'+1}^{k}\frac{V^{(H)}_{M_1',p_1-1,M_1',p_1}}{E_{M_1',p_1}-E_{M_1M_2}}\right)V^{(J_{++})}_{M_1',k,M_1'+1,k+1}\left(\prod_{p_2=k+1}^{M_2\text{-}1}\frac{V^{(H)}_{M_1,p_2,M_1,p_2+1}}{E_{M_1,p_2}-E_{M_1M_2}}\right)\right]+2\left(\prod_{p=M_2'}^{M_2\text{-}2}\frac{V^{(H)}_{M_1',p,M_1',p+1}}{E_{M_1',p+1}-E_{M_1,M_2}}\right)V^{(J_{++})}_{M_1-1,M_2-1,M_1,M_2}. \tag{24}$$

Using the matrix element

$$V^{(H)}_{M_1,p,M_1,p+1}=\left\langle M_1,p\left|-\frac{H_x}{2}\hat{S}_{-2}\right|M_1,p+1\right\rangle=-\frac{H_x}{2}l_{p+1}, \tag{25}$$

in the limit of small transverse field we have the level splitting given by

$$\Delta E_{M_1',M_2',M_1,M_2}=2J_{++}\left(\frac{H_x}{2D}\right)^{M_2-M_2'-1}g_{++}\sum_{k=M_2'}^{M_2\text{-}1}\left[\frac{1}{(M_2-k-1)!(k-M_2')!}\frac{\Gamma(-k-a)\Gamma(k+b+1)}{\Gamma(-M_2'-a)\Gamma(M_2+b)}\right]. \tag{26}$$

As a ‘‘model V,’’ we consider the transverse exchange interaction containing J_{+-} and the same form of $\mathcal{H}_i^{\mathrm{trans}}$ as Eq. (23). Taking the case at $i=1$, the tunnel splitting occurs in the degenerate pair of (M_1', M_2') and (M_1, M_2) where $M_2' = M_2 + 1$ and $M_1 - (M_1'+1)$ is odd number. Including the transverse exchange interaction in the level splitting through the single perturbation step along the chain connecting the degenerate states, the level splitting is expressed as

$$\Delta E_{M_1',M_2',M_1,M_2} = 2V^{(J_{+-})}_{M_1',M_2',M_1'+1,M_2'-1}\left(\prod_{p=M_1'+1}^{M_1\text{-}1}\frac{V^{(H)}_{p,M_2,p+1,M_2}}{E_{p,M_2}-E_{M_1,M_2}}\right)$$
$$+2\sum_{k=M_1'+1}^{M_1\text{-}2}\left[\left(\prod_{p_1=M_1'+1}^{k}\frac{V^{(H)}_{p_1-1,M_2',p_1,M_2'}}{E_{p_1,M_2'}-E_{M_1M_2}}\right)V^{(J_{+-})}_{k,M_2',k+1,M_2'-1}\left(\prod_{p_2=k+1}^{M_1\text{-}1}\frac{V^{(H)}_{p_2,M_2,p_2+1,M_2}}{E_{p_2,M_2}-E_{M_1M_2}}\right)\right]$$
$$+2\left(\prod_{p=M_1'}^{M_1\text{-}2}\frac{V^{(H)}_{p,M_2',p+1,M_2'}}{E_{p+1,M_2'}-E_{M_1,M_2}}\right)V^{(J_{+-})}_{M_1-1,M_2+1,M_1,M_2}, \tag{27}$$

which leads to

$$\Delta E_{M_1',M_2',M_1,M_2} = 2J_{+-}\left(\frac{H_x}{2D}\right)^{M_1-M_1'-1} g_{+-}\sum_{k=M_1'}^{M_1\text{-}1}\left[\frac{1}{(M_1-k-1)!(k-M_1')!}\frac{\Gamma(-k-d)\Gamma(k+c+1)}{\Gamma(-M_1'-d)\Gamma(M_1+c)}\right]. \tag{28}$$

Up to now, we have separately considered the transverse anisotropy and the transverse field in models II–V. In some situations both the transverse field and the transverse anisotropy are present with H_x and B being the same order of magnitude, i.e., $H_x \sim B \ll D$. In this case the effect of the transverse field on level splitting is much weaker than that of the transverse anisotropy, as is evident in Eqs. (18), (21), (26), and (28). Thus, for $M_2 - M_2' - 1$ (or $M_1 - M_1' - 1$) with even number one can neglect the field contribution to the level splittings, whereas for $M_2 - M_2' - 1$ (or $M_1 - M_1' - 1$) with odd number the field should be taken into account in the first order only. In the latter case, the main source of the splitting is the transverse anisotropy taken in a high order of a perturbation theory. For such cases both the transverse exchange interactions (J_{++} or J_{+-}) and the transverse field make the missing two perturbation steps along the chain of matrix element. The corresponding matrix elements $V^{(J_{++})}_{M_1',k,M_1'+1,k+1}$ (or $V^{(J_{+-})}_{k,M_2',k+1,M_2'-1}$) and $V^{(H)}_{q,k,q,k+1}$ (or $V^{(H)}_{k,q,k+1,q}$) can be inserted at any place in the chain, $k = M_2', M_2'+2, \ldots, M_2-1$ (or $k = M_1', M_1'+2, \ldots, M_1-1$), where q becomes M_1 or M_1' (M_2 or M_2') depending on the ordering of $V^{(J_{++})}$ (or $V^{(J_{+-})}$) and $V^{(H)}$ in the chain. Keeping these points in mind, we first consider as ‘‘model VI’’ the level splitting generated by the transverse exchange interaction with J_{++}, the transverse anisotropy, and the transverse field. In this situation we divide the contributions of the level splitting into three parts: (i) $\{V^{(H)}V^{(J_{++})}\cdots\} + \{V^{(J_{++})}\cdots V^{(H)}\cdots\} + \{V^{(J_{++})}\cdots V^{(H)}\}$, (ii) $\{V^{(H)}\cdots V^{(J_{++})}\} + \{\cdots V^{(H)}\cdots V^{(J_{++})}\} + \{\cdots V^{(J_{++})}V^{(H)}\}$, and (iii) $\{V^{(H)}\cdots V^{(J_{++})}\cdots\} + \{\cdots V^{(H)}\cdots V^{(J_{++})}\cdots\} + \{\cdots V^{(J_{++})}\cdots V^{(H)}\cdots\} + \{\cdots V^{(J_{++})}\cdots V^{(H)}\}$. Here, $\cdots$ in the curly bracket, e.g., $\{V^{(H)}V^{(J_{++})}\cdots\}$ indicates that all the possible combinations of $V^{(B)}$ should be counted and summarized, whose result will be shown later. Then, the level splittings can be written in the form

$$\Delta E^{(\mathrm{i})}_{M_1',M_2',M_1,M_2} = 2V^{(H)}_{M_1',M_2',M_1',M_2'+1}\frac{V^{(J_{++})}_{M_1',M_2'+1,M_1'+1,M_2'+2}}{E_{M_1',M_2'+1}-E_{M_1,M_2}}\left(\prod_{p=M_2'+2}^{M_2\text{-}2}{}'\frac{V^{(B)}_{M_1'+1,p,M_1'+1,p+2}}{E_{M_1,p}-E_{M_1,M_2}}\right)$$
$$+2V^{(J_{++})}_{M_1',M_2',M_1'+1,M_2'+1}\sum_{l=M_2'+1}^{M_2\text{-}3}{}'\left[\left(\prod_{p_1=M_2'+1}^{l-2}{}'\frac{V^{(B)}_{M_1'+1,p_1,M_1'+1,p_1+2}}{E_{M_1'+1,p_1}-E_{M_1M_2}}\right)\frac{V^{(H)}_{M_1'+1,l,M_1'+1,l+1}}{E_{M_1'+1,l}-E_{M_1M_2}}\right.$$
$$\left.\times\left(\prod_{p_2=l+1}^{M_2\text{-}2}{}'\frac{V^{(B)}_{M_1,p_2,M_1,p_2+2}}{E_{M_1,p_2}-E_{M_1M_2}}\right)\right]+2V^{(J_{++})}_{M_1',M_2',M_1'+1,M_2'+1}\frac{V^{(H)}_{M_1,M_2-1,M_1,M_2}}{E_{M_1,M_2-1}-E_{M_1,M_2}}\left(\prod_{p=M_2'+1}^{M_2\text{-}3}{}'\frac{V^{(B)}_{M_1'+1,p,M_1'+1,p+2}}{E_{M_1'+1,p}-E_{M_1,M_2}}\right), \tag{29}$$

$$\Delta E^{(\mathrm{ii})}_{M_1',M_2',M_1,M_2}=2V^{(H)}_{M_1',M_2',M_1',M_2'+1}\left(\prod_{p=M_2'+1}^{M_2\text{-}3}{}'\frac{V^{(B)}_{M_1',p,M_1',p+2}}{E_{M_1',p}-E_{M_1,M_2}}\right)\frac{V^{(J_{++})}_{M_1-1,M_2-1,M_1,M_2}}{E_{M_1-1,M_2-1}-E_{M_1,M_2}}$$
$$+2\sum_{l=M_2'+2}^{M_2\text{-}2}{}'\left[\left(\prod_{p_1=M_2'}^{l-2}{}'\frac{V^{(B)}_{M_1',p_1,M_1',p_1+2}}{E_{M_1',p_1+2}-E_{M_1M_2}}\right)\frac{V^{(H)}_{M_1',l,M_1',l+1}}{E_{M_1',l+1}-E_{M_1M_2}}\left(\prod_{p_2=l+1}^{M_2\text{-}3}{}'\frac{V^{(B)}_{M_1',p_2,M_1',p_2+2}}{E_{M_1',p_2+2}-E_{M_1M_2}}\right)\right]$$
$$\times V^{(J_{++})}_{M_1-1,M_2-1,M_1,M_2}+2\left(\prod_{p=M_2'}^{M_2\text{-}4}{}'\frac{V^{(B)}_{M_1',p,M_1',p+2}}{E_{M_1',p+2}-E_{M_1,M_2}}\right)\frac{V^{(J_{++})}_{M_1-1,M_2-2,M_1,M_2-1}}{E_{M_1,M_2-1}-E_{M_1,M_2}}V^{(H)}_{M_1,M_2-1,M_1,M_2}, \tag{30}$$

and

$$\Delta E^{(\mathrm{iii})}_{M_1',M_2',M_1,M_2}=2V^{(H)}_{M_1',M_2',M_1',M_2'+1}\sum_{k=M_2'+3}^{M_2\text{-}3}{}'\left[\left(\prod_{p_1=M_2'+1}^{k-2}{}'\frac{V^{(B)}_{M_1',p_1,M_1',p_1+2}}{E_{M_1',p_1}-E_{M_1,M_2}}\right)\frac{V^{(J_{++})}_{M_1',k,M_1'+1,k+1}}{E_{M_1',k}-E_{M_1,M_2}}\right.$$
$$\left.\times\left(\prod_{p_2=k+1}^{M_2\text{-}2}{}'\frac{V^{(B)}_{M_1,p_2,M_1,p_2+2}}{E_{M_1,p_2}-E_{M_1,M_2}}\right)\right]+2\sum_{k=M_2'+3}^{M_2\text{-}3}{}'\sum_{l=M_2'+2}^{k-1}{}'\left[\left(\prod_{p_1=M_2'}^{l-2}{}'\frac{V^{(B)}_{M_1',p_1,M_1',p_1+2}}{E_{M_1',p_1+2}-E_{M_1M_2}}\right)V^{(H)}_{M_1',l,M_1',l+1}\right.$$
$$\left.\times\left(\prod_{p_2=l+1}^{k-2}{}'\frac{V^{(B)}_{M_1',p_2,M_1',p_2+2}}{E_{M_1',p_2}-E_{M_1M_2}}\right)\frac{V^{(J_{++})}_{M_1',k,M_1'+1,k+1}}{E_{M_1',k}-E_{M_1M_2}}\left(\prod_{p_3=k+1}^{M_2\text{-}2}{}'\frac{V^{(B)}_{M_1,p_3,M_1,p_3+2}}{E_{M_1,p_3}-E_{M_1M_2}}\right)\right]$$
$$+2\sum_{l=M_2'+3}^{M_2\text{-}3}{}'\sum_{k=M_2'+2}^{l-1}{}'\left[\left(\prod_{p_1=M_2'}^{k-2}{}'\frac{V^{(B)}_{M_1',p_1,M_1',p_1+2}}{E_{M_1',p_1+2}-E_{M_1M_2}}\right)\frac{V^{(J_{++})}_{M_1',k,M_1'+1,k+1}}{E_{M_1'+1,k+1}-E_{M_1M_2}}\right.$$
$$\left.\times\left(\prod_{p_2=k+1}^{l-2}{}'\frac{V^{(B)}_{M_1,p_2,M_1,p_2+2}}{E_{M_1,p_2+2}-E_{M_1M_2}}\right)V^{(H)}_{M_1,l,M_1,l+1}\left(\prod_{p_3=l+1}^{M_2\text{-}2}{}'\frac{V^{(B)}_{M_1,p_3,M_1,p_3+2}}{E_{M_1,p_3}-E_{M_1M_2}}\right)\right]$$
$$+2\sum_{k=M_2'+2}^{M_2\text{-}4}{}'\left[\left(\prod_{p_1=M_2'}^{k-2}{}'\frac{V^{(B)}_{M_1',p_1,M_1',p_1+2}}{E_{M_1',p+2}-E_{M_1,M_2}}\right)\frac{V^{(J_{++})}_{M_1',k,M_1'+1,k+1}}{E_{M_1'+1,k+1}-E_{M_1,M_2}}\right.$$
$$\left.\times\left(\prod_{p_2=k+1}^{M_2\text{-}3}{}'\frac{V^{(B)}_{M_1,p_2,M_1,p_2+2}}{E_{M_1,p_2+2}-E_{M_1,M_2}}\right)\right]V^{(H)}_{M_1,M_2-1,M_1,M_2}, \tag{31}$$

where it is noted that $M_1=M_1'+1$ and M_2-M_2' is even number. Now, summing these three contributions, one obtains

$$\Delta E_{M_1',M_2',M_1,M_2}=2J_{++}\left(\frac{H_x}{4D}\right)\left(\frac{B}{4D}\right)^{(M_2-M_2'-2)/2}g_{++}$$
$$\times\sum_{k=M_2'+1}^{M_2\text{-}1}{}'\sum_{l=M_2'}^{k-1}{}'\left\{\frac{1}{(l-M_2')!!(M_2-k-1)!!}\frac{\Gamma\left(\frac{-l-a}{2}\right)\Gamma\left(\frac{k+b+1}{2}\right)}{\Gamma\left(\frac{-M_2'-a}{2}\right)\Gamma\left(\frac{M_2+b}{2}\right)}\right.$$
$$\left.\times\left[\frac{(l-M_2'-1)!!}{(k-M_2')!!}\frac{\Gamma\left(\frac{-k-a}{2}\right)}{\Gamma\left(\frac{-l-a+1}{2}\right)}+\frac{(M_2-k-2)!!}{(M_2-l-1)!!}\frac{\Gamma\left(\frac{l+b+1}{2}\right)}{\Gamma\left(\frac{k+b+2}{2}\right)}\right]\right\}. \tag{32}$$

Our final example, model VII, is the level splitting generated by the transverse anisotropy and the transverse field as well as the transverse exchange anisotropy with J_{+-}. Taking the case at $i=1$ as in model V, the tunnel splitting exists in the degenerate pair satisfying the condition that $M_2'=M_2+1$ and M_1-M_1' is even number. In the same way as we have done in model VI, we consider three types which contribute the level splitting. Then, the resulting level splitting has the form

$$\Delta E_{M_1',M_2',M_1,M_2}=2J_{+-}\left(\frac{H_x}{4D}\right)\left(\frac{B}{4D}\right)^{(M_1-M_1'-2)/2} g_{+-} \times \sum_{k=M_1'+1}^{M_1\text{-}1}{}' \sum_{l=M_1'}^{k-1}{}' \left\{ \frac{1}{(l-M_1')!!(M_1-k-1)!!} \frac{\Gamma\left(\frac{-l-d}{2}\right)\Gamma\left(\frac{k+c+1}{2}\right)}{\Gamma\left(\frac{-M_1'-d}{2}\right)\Gamma\left(\frac{M_1+c}{2}\right)} \times \left[\frac{(l-M_1'-1)!!}{(k-M_1')!!} \frac{\Gamma\left(\frac{-k-d}{2}\right)}{\Gamma\left(\frac{-l-d+1}{2}\right)} + \frac{(M_1-k-2)!!}{(M_1-l-1)!!} \frac{\Gamma\left(\frac{l+c+1}{2}\right)}{\Gamma\left(\frac{k+c+2}{2}\right)} \right] \right\}. \quad (33)$$

To illustrate the results with a concrete example, we have considered a supramolecular dimer Mn_4-dbm. This compound has a distorted cubanelike core geometry and contains three Mn^{3+} ions and one Mn^{4+} ion with axial anisotropy constant ($D\simeq0.72$ K) and a transverse anisotropy constant ($B\simeq0.033$ K), and exchange coupling along the $\hat{z}$ axis ($J_z \simeq -0.01$ K) between them leads to the $[Mn_4]_2$ dimer having a ground-state spin of $S_1=S_2=9/2$.[7] Many energy level crossings can be possible quantum transitions depending on the magnitude of transverse terms including B, H_x, J_{+-}, and J_{++} and the type of the transitions. Among them we have selected 13 level crossings and plotted the corresponding energy levels at resonant fields ($H_z^{(0)}$) (Fig. 1). In Table I, we have divided 13 such level crossings into different types and clarified the main sources of the level splitting.

Before proceeding to the detailed discussion of the transitions, it is important to obtain the magnitude of the intrinsic transverse field (H_x^{int}) and the transverse exchange constants (J_{++} and J_{+-}). Comparison of the level splittings ΔE in 1 and 7 with the results in experiment (Fig. 5 in Ref. 7) gives $H_x^{\text{int}}\sim0.146$ K and $J_{++}\sim5.85\times10^{-3}$ K. Taking $J_{++}\sim J_{+-}$, we obtain the tunnel splitting with the order of magnitude $10^{-3}-10^{-7}$ by using Eqs. (18), (21), (32), (33). Transitions 1, 8, 9, and 12 are identical to those of the model describing the spin system of an isolated SMM. In these situations the resonant fields are shifted due to the exchange interaction between two SMM's. Transitions 2–6 and 10 correspond to thermal activation to excited state (IS) from the ground state in one of two coupled spins $[(-9/2,-9/2)$ or $(-9/2,9/2)]$ and tunneling from the excited state (IS) to another excited state (FS). At extremely low temperature (e.g., $T\sim40$ mK) such transitions are negligibly small, as is shown in Table I. Thus, only a few of transitions (1, 7, 8, 9, 11, 12, 13) are relevant, which is consistent with hysteresis loop measurements of a single crystal of Mn_4-dbm at very low temperature. Among them transitions 7, 11, and 13 are collective quantum process, called SSCR, involving pairs of SMM's which are coupled by J_{++} or J_{+-}. For example, transition 7 corresponds to tunneling from the ground state $(-9/2,-9/2)$ to the excited state $(-7/2,9/2)$. Indeed, for transition 7 the change of the quantum number $\delta M(=M_2-M_2')$ is even, whereas it $(=M_1-M_1'$ or $M_2-M_2')$ is odd for transitions 11 and 13. As will become apparent below, even for $H_x^{\text{int}}(\sim0.146$ K) much greater than $B(\sim0.033$ K) the effect of the transverse field on level splitting is much weaker than that of the transverse anisotropy. Thus, for the even δM one can neglect the field contribution to the splittings, whereas for the odd δM the field should be taken into

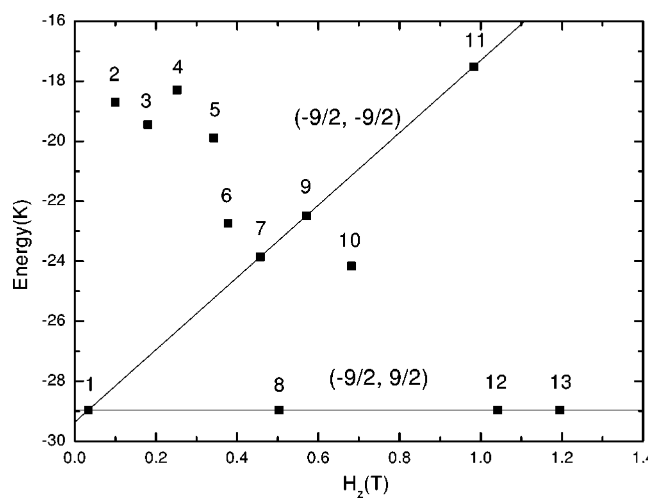

FIG. 1. Low lying energy level diagram of two coupled spins $S=9/2$ for the resonant magnetic field [Eq. (4)] in Mn_4-dbm. The numbers, labeled from 1 to 13, indicate the states claimed as the tunnel transitions in Ref. 7. Two straight lines are the energy levels with the states $(-9/2,-9/2)$ and $(-9/2,9/2)$ which are ground states of the Mn_4-dbm dimer. Note that initial states for transitions 1, 7, 9, 11 and 8, 12, 13 are $(-9/2,-9/2)$ and $(-9/2,9/2)$, respectively.

TABLE I. The 13 tunnel transitions, which are labeled from 1 to 13 in Fig. 1, for dimer with $S=9/2$. The level splitting (ΔE) [$B=0.033$ K (Ref. 13) and $H_x \sim 0.108$ T, $J_{+\pm} \sim 5.85\times 10^{-3}$ K for illustration] of the low-lying degenerate pair in Mn_4-dbm, the rate of thermally assisted tunneling (TAT) from ground state with $\Delta E\ \exp(-\beta U_{G_i^\pm \to \mathrm{IS}})$ [$T\sim 40$ mK, $U_{G_i^\pm \to \mathrm{IS}} \simeq -D(M_i'^2 - S^2)$, where $G_i^\pm : (-9/2, \pm 9/2)$, and i denotes one of two single molecules with index 1 or 2], the resonant field ($H_z^{(0)}$), and the physical origins which induce level splittings. For clarity, degenerate states such as (M_1, M_2) and (M_2, M_1) are not both listed. IS: initial state (M_1', M_2'), FS: final state (M_1, M_2), SSM: single-spin model (Ref. 9).

	IS	FS	$H_z^{(0)}$(T)	ΔE(K)	$G_i^\pm$	TAT	main source	Type
1	$(-\frac{9}{2},-\frac{9}{2})$	$(-\frac{9}{2},\frac{9}{2})$	0.0336	2.0×10^{-7}			B, H_x	SSM
2	$(-\frac{9}{2},\frac{5}{2})$	$(\frac{7}{2},\frac{7}{2})$	0.0999	2.38×10^{-5}	G_2^+	8.58×10^{-115}	J_{++}, B, H_x	VI
3	$(-\frac{5}{2},\frac{9}{2})$	$(\frac{7}{2},\frac{7}{2})$	0.180	1.39×10^{-3}	G_1^+	5.01×10^{-113}	J_{+-}, B, H_x	VII
4	$(-\frac{9}{2},\frac{5}{2})$	$(\frac{9}{2},\frac{3}{2})$	0.252	5.89×10^{-7}	G_2^+	2.13×10^{-116}	J_{+-}, B	III
5	$(-\frac{9}{2},-\frac{7}{2})$	$(\frac{9}{2},-\frac{5}{2})$	0.343	5.34×10^{-7}	G_2^-	1.55×10^{-69}	J_{++}, B	II
6	$(-\frac{9}{2},\frac{7}{2})$	$(\frac{9}{2},\frac{5}{2})$	0.378	6.72×10^{-7}	G_2^+	1.95×10^{-69}	J_{+-}, B	III
7	$(-\frac{9}{2},-\frac{9}{2})$	$(-\frac{7}{2},\frac{9}{2})$	0.457	5.0×10^{-7}			J_{++}, B	II
8	$(-\frac{9}{2},\frac{9}{2})$	$(\frac{7}{2},\frac{9}{2})$	0.504	2.09×10^{-5}			B	SSM
9	$(-\frac{9}{2},-\frac{9}{2})$	$(-\frac{9}{2},\frac{7}{2})$	0.571	2.09×10^{-5}			B	SSM
10	$(-\frac{7}{2},\frac{9}{2})$	$(\frac{7}{2},\frac{7}{2})$	0.682	4.84×10^{-3}	G_1^+	1.4×10^{-65}	J_{+-}, B	III
11	$(-\frac{9}{2},-\frac{9}{2})$	$(-\frac{7}{2},\frac{7}{2})$	0.982	5.86×10^{-6}			J_{++}, B, H_x	VI
12	$(-\frac{9}{2},\frac{9}{2})$	$(\frac{5}{2},\frac{9}{2})$	1.04	3.56×10^{-5}			B, H_x	SSM
13	$(-\frac{9}{2},\frac{9}{2})$	$(\frac{7}{2},\frac{7}{2})$	1.19	1.22×10^{-5}			J_{+-}, B, H_x	VII

account in the first order only (models VI or VII). Thus, for transition 7 the transverse anisotropy is the dominant mechanism causing tunneling, with the exchange interaction being of the first order. For transitions 11 and 13 the tunnel splitting is primarily due to transverse anisotropy with a small admixture of tunneling due to a transverse field and an exchange interaction. It is also noted that, in the case that the transverse field is considered as the dominant mechanism causing quantum transitions, the order of magnitude of the tunnel splittings are estimated to be about 10^{-10} for transitions 11 (model V) and 13 (model IV), which are much smaller than about 10^{-5} or 10^{-6} in Table I. Finally, it is meaningful to estimate the tunnel splittings only induced by the transverse exchange interactions which correspond to the transitions either from $(-9/2,9/2)$ to $(9/2,-9/2)$ or from $(-9/2,-9/2)$ to $(9/2,9/2)$. Using the results (9) and (12) in model I, the level splittings are of the order of 10^{-20} which is much smaller than those induced by the transverse anisotropy or the transverse field, as is shown in Table I. As a result, the steplike features in the hysteresis loops of the Mn_4-dbm are generated by the combination of the transverse anisotropy, the transverse field and the transverse exchange interactions.

In conclusion, we have considered the level splittings in two coupled single-molecule magnets with the anisotropic exchange interaction. A perturbation approach allows us to estimate the level splittings of the states, degenerate pairwise in Mn_4-dbm. It is found that the anisotropic exchange interaction plays a crucial role in the level splitting and shifts of the resonant field. In comparison with recent experimental results, the intrinsic transverse field and the transverse exchange interactions have been extracted. Using these values the level splittings of the low-lying degenerate pair have been estimated for 13 cases and the main sources of each resonance have been clarified. Among them seven cases which include SSCR and SSM have been found to be dominant at very low temperature.

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