Resonant coherent quantum tunneling of the magnetization of spin-$\frac{1}{2}$ systems: Spin-parity effects

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We perform quantum dynamical calculations to study the reversal of the magnetization for systems of a few spin-$\frac{1}{2}$ particles with a general biaxial anisotropy in the presence of an external magnetic field at $T=0$ and with no dissipation. Collective quantum tunneling of the magnetization is demonstrated to occur only for some specific resonant values of the magnetic field proving the invalidity of semiclassical approximations for clusters of a few spin-$\frac{1}{2}$ particles. Quantum tunneling of the magnetization direction at zero field is shown to be spin-parity dependent: whereas clusters with an odd number of particles do not exhibit quantum tunneling of the magnetization due to the degeneration between the first two levels, clusters with an even number of spins present resonant coherent quantum tunneling of the magnetization. On the contrary, resonant coherent quantum tunneling of the magnetization at finite magnetic field has no detectable parity dependence. We study the dependence of such collective tunneling of the magnetization on the magnetic field, the number of spins, and the anisotropy. [S0163-1829(97)08201-5]

Quantum tunneling of macroscopic variables provides one of the most striking manifestations of quantum mechanics.[1] The phenomena of macroscopic quantum tunneling (MQT) and coherence (MQC) of the magnetization in small magnetic particles have been the subject of active research over the last few years.[2–4] For a single-domain ferromagnetic particle the magnetic anisotropy creates easy directions for the total magnetization which correspond to the local minima of the energy. For such particles, MQT consists of the tunneling of the magnetization out of metastable easy directions in the presence of external fields, and MQC consists of the resonance between equivalent easy directions, or in other words, MQC occurs when all the spins coherently tunnel back and forth between the two minima of the energy.[5,6]

Quantum tunneling in spins systems has been theoretically studied in the semiclassical limit and for a single spin model (SSM) by means of two approaches, one using a generalization of the conventional WKB method[7,8] and the other[9–12] based on Feynman's path-integral formulation of quantum mechanics. In such semiclassical approximations, all spins are considered to behave dynamically as a single-quantum spin, and in consequence, uniform and coherent rotation of all the spins is imposed. However, recent experiments[13,14] and theoretical works[15–17] have found that the reversal mechanism of the magnetization in single-domain particles could differ from the simple uniform rotation. A theoretical approach to the problem has been recently made by the authors performing quantum dynamical calculations on the magnetization reversal in systems of a few spins by calculating a numerically exact solution of the time-dependent Schrödinger equation.[18,19] These calculations demonstrate that the model considered (clusters of spin-$\frac{1}{2}$ particles described by an anisotropic Heisenberg Hamiltonian at $T=0$) for small magnetic particles exhibits collective tunneling of the magnetization only for some specific resonant values of the magnetic field, at variance with the Stoner-Wohlfarth model which fails to predict the phenomenon of resonant tunneling. Similar conclusions have been reached performing full quantum mechanical calculations for the single-spin quantum model,[20] demonstrating that coherent tunneling of the magnetization occurs at specific values of the resonant magnetic field only. In addition, exact quantum calculations of MQC on the two-dimensional anisotropic Heisenberg antiferromagnetic have been presented in a recent paper.[21]

For a general biaxial anisotropy (which leads to the existence of easy, medium, and hard axes) spin-parity effects in quantum tunneling have been found for uniform spin systems using path-integral analysis.[22,23] These calculations have shown the suppression of tunneling for half-integer spins at zero field. In this formalism, this phenomena is caused by destructive interference between different tunneling paths, these interference effects being much related to the symmetries of the spin Hamiltonian. For some Hamiltonians, Kramers' degeneracy[24] in half-integer spins has been shown to imply absence of tunneling at zero field. Later works[25,26] considering external magnetic fields have reached the conclusion that this quenching of tunneling is far more general than Kramers' theorem, finding that the tunnel splitting oscillates with the field and vanishes at certain values (vanishing at zero field for half-integer spins also follows from Kramers' theorem). A recent paper[27] has associated these spin-parity effects with a selection rule due to an underlying rotational symmetry.

Since semiclassical methods have been shown inadequate to examine the quantum tunneling phenomenon for clusters of a few spins with uniaxial anisotropy,[18,19] we have also considered systems with general biaxial anisotropy and in-

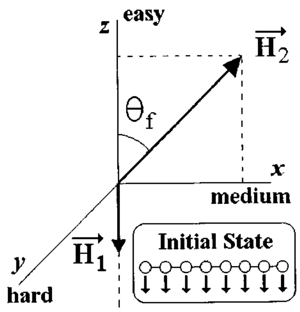

FIG. 1. System of N spin-$\frac{1}{2}$ particles with easy, medium, and hard axes of magnetization along z, x, and y, respectively, in the presence of magnetic fields. Initially, the field $\mathbf{H}_1$ is applied along the easy z axis preparing the system with all spins down. Subsequently, the magnetic field is rotated forming an angle $\theta_f = 45°$ with the z axis.

vestigated with quantum dynamical exact calculations such total spin-parity effects. We are interested in studying models of small magnetic particles without assuming that all the spins must behave as a rigid single one and not employing semiclassical approximations. As pointed out previously there are experimental and theoretical works that question these assumptions. Performing quantum dynamical calculations for systems of a few spin-$\frac{1}{2}$ particles described by a Heisenberg Hamiltonian with general biaxial anisotropy at $T=0$, quantum tunneling of the magnetization direction at zero field is shown to be spin-parity dependent: whereas clusters with an odd number of particles do not exhibit quantum tunneling of the magnetization due to the degeneration between the first two levels, clusters with an even number of spins present resonant coherent quantum tunneling of the magnetization. On the other hand, the phenomenon of resonant coherent quantum tunneling is shown to be spin-parity independent at finite magnetic fields: both clusters with even and odd numbers of spins can present off-zero-field resonances corresponding to coherent tunneling.

We have represented a system containing N spin-$\frac{1}{2}$ particles in the presence of an applied magnetic field through its Heisenberg Hamiltonian:

$$\mathcal{H} = -J_x \sum_{\langle ij \rangle} \sigma_i^x \sigma_j^x - J_y \sum_{\langle ij \rangle} \sigma_i^y \sigma_j^y - J_z \sum_{\langle ij \rangle} \sigma_i^z \sigma_j^z - H_x \sum_i \sigma_i^x - H_z \sum_i \sigma_i^z, \quad (1)$$

where σ_i^α $(\alpha = x,y,z)$ are the Pauli-spin matrices at site i related to the spin operators by $\mathbf{S} = \hbar \boldsymbol{\sigma}/2$, the sum $\langle ij \rangle$ is over nearest-neighbor pairs, J_x, J_y, J_z are the exchange constants, and H_x, H_z are the components of the external magnetic field. This model can represent a single-domain ferromagnetic particle with general biaxial anisotropy by making different the exchange constants J_x, J_y, and J_z, so that if $J_z > J_x > J_y$ then the system has its easy, medium, and hard axes of magnetization along z, x, and y, respectively (Fig. 1). (In other words, in that case z is the easy-magnetization axis and xz the easy-magnetization plane.) We consider instantaneous rotations of the external magnetic field and calculate the temporal evolution of the system to study the magnetization reversal for these systems. At $t=0$ there is a field applied along the z direction, $\mathbf{H}_1 = (0,0,H_{z1})$ with $H_{z1} < 0$. Then the ground state of the ferromagnet has all spins down and we prepare the system in such state. At $t>0$, the magnetic field is rotated instantaneously about the y axis so that $\mathbf{H}_2 = (H_{x2}, 0, H_{z2})$ with $H_{x2}, H_{z2} > 0$ forms an angle θ_f with the z axis and the dynamical evolution is computed.

The temporal evolution of the system is calculated by a numerically exact solution of the time-dependent Schrödinger equation.[28] This requires the computation of all eigenvalues and eigenvectors of the Hamiltonian. The limiting factor of this approach is the amount of memory needed to store all eigenvectors, which scales as 2^{2N}. The formal solution of the time-dependent Schrödinger equation, given an initial wave function $|\Psi(t=0)\rangle$ is expressed as

$$|\Psi(t)\rangle = e^{-it\mathcal{H}} |\Psi(t=0)\rangle, \quad (2)$$

and the temporal evolution of the α $(\alpha = x,y,z)$ component of each spin and the total spin can be obtained, respectively, as

$$\langle S_i^\alpha \rangle(t) = \langle \Psi(t) | S_i^\alpha | \Psi(t) \rangle \quad (3)$$

and

$$\langle S^\alpha \rangle(t) = \left\langle \Psi(t) \left| \frac{1}{N} \sum_i S_i^\alpha \right| \Psi(t) \right\rangle, \quad (4)$$

N being the number of particles. Actually, the expectation value of σ_i^α instead of S_i^α is calculated. In addition, we have calculated the expectation value of S_i^α for each nonequivalent spin i (i.e., spins with different numbers of couplings) averaged over time

$$\overline{S_i^\alpha} = \lim_{\tau \to \infty} \left[\tau^{-1} \int_0^\tau dt \langle S_i^\alpha(t) \rangle \right] \quad (5)$$

for different values of the magnetic field $\mathbf{H}_2$, and the same quantity for the total spin, $\overline{S^\alpha}$.

It is essential to introduce the two-time correlation function of the magnetization,[5] which compares the z component of S at one time with its value at a time later: $\langle S^z(t') S^z(t'+t) \rangle$. In the present work, the symmetrized correlation function $C(t)$ is calculated using the definition

$$C(t) = \tfrac{1}{2} \langle \Psi(0) | S^z(0) S^z(t) + S^z(t) S^z(0) | \Psi(0) \rangle. \quad (6)$$

With negligible dissipation present, coherent tunneling back and forth between easy directions leads to a sinusoidal oscillation of $C(t)$ at a frequency twice the tunneling rate Γ. For two measurements of the magnetization separated by the time interval t, one should have[5,6]

$$\langle S(t') S(t'+t) \rangle = S_0^2 \cos(2\Gamma t). \quad (7)$$

As the fluctuation-dissipation theorem shows that the frequency-dependent magnetic susceptibility $\chi''(\omega)$ is essentially the Fourier transform of the correlation function, the former equation predicts a resonance at $\omega_R = 2\Gamma$ for $\chi''(\omega)$. In experiments with superconducting quantum inter-

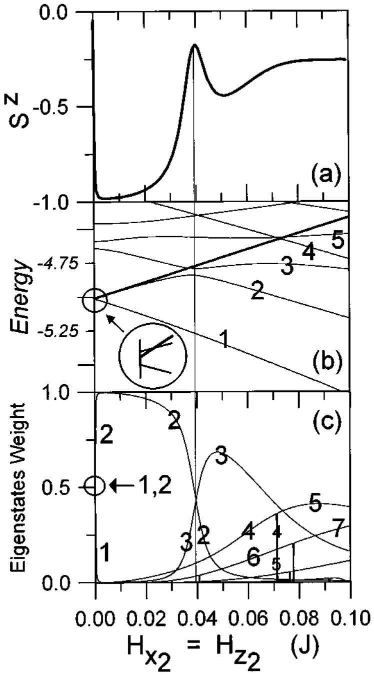

FIG. 2. (a) $\overline{S^z}$ as a function of the applied magnetic field for the model of Eq. (1) with $J_x=0.9$, $J_y=0.85$, and $J_z=1.0$ (easy axis along z and xz as easy plane) for a cluster of six spins. (b) System energy $\langle\Psi|H|\Psi\rangle$ (thick line) and low-lying eigenenergies numbered in order of increasing energy as a function of the applied magnetic field; an inset shows the energy splitting between the first two levels at zero field. (c) Eigenstates weights in the decomposition of the system state, $|\Psi\rangle=\Sigma c_n|E_n\rangle$, labeled by the number given to the corresponding energy levels.

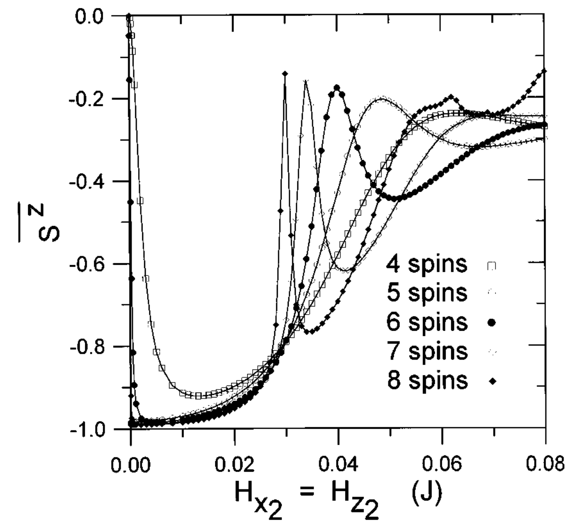

FIG. 3. $\overline{S^z}$ as a function of the applied magnetic field for the model of Eq. (1) with $J_x=0.9$, $J_y=0.85$, and $J_z=1.0$ for systems with 4 (squares), 5 (circles), 6 (solid circles), 7 (rhombus), and 8 (solid rhombus) spins.

ference device microsusceptometers,[29] a well-defined resonance in $\chi''(\omega)$ has been found and it is tempting to be associated with a MQC phenomenon although there is some controversy on this interpretation.[30] The sharpness of the resonance indicates that the coupling to the environment is weak, which is an important requirement for MQC, and it is also found that the resonance frequency is very sensitive to small fields. Many research works in MQC have studied the effect of dissipation on this resonance.[5]

According to the method previously described, the temporal evolution of the system of spins is calculated when an external magnetic field is rotated instantaneously. This evolution is not coherent in general, the nonuniformity depending on the values of the anisotropy and the magnetic field. As it was demonstrated in Ref. 18, only for some specific resonant fields coherent tunneling of the magnetization occurs in systems of spins with uniaxial anisotropy. We will now show that for clusters with a general biaxial anisotropy ($J_x\neq J_y \neq J_z$ in our Hamiltonian), in addition to resonances for off-zero-magnetic fields, a zero-field resonance corresponding to coherent quantum tunneling can be present or not depending on the even or odd character of the number N of spin-$\frac{1}{2}$ particles.

Let us analyze a cluster of six spins with easy axis along z and xz as easy plane ($J_x=0.9$, $J_y=0.85$, and $J_z=1.0$) that is initially prepared with all spins down by applying a magnetic field along the easy direction [$\mathbf{H}_1=(0,0,-0.06)$]. Subsequently, the magnetic field is rotated forming a 45° angle with the z axis and the dynamical evolution is computed for different values of the magnetic field. Two peaks can be seen in the curve of $\overline{S^z}$ versus the magnetic field [Fig. 2(a)]: a sharp one at zero field and a broader one at a field with H_x and H_z close to 0.04. These resonances correspond to coherent reversal of all the spins. The correlation function for the resonance of the six spins cluster at $H=0$ can be seen in Fig. 4, it being a pure sinusoidal oscillation as it must be in the case of quantum coherence phenomena. On the contrary, for fields out of resonances the correlation function does not present this sinusoidal aspect but a complicated shape close to the value 1,[18] which means that the total spin is not reversed and the spins do not evolve coherently.

The off-zero-field resonance although not sharp is one of the type shown in Ref. 18 and it appears since at that specific field, the system state is essentially a superposition of the second and third eigenstates [see in Fig. 2(c) that for $H_x=H_z\simeq0.04$, eigenstates 2 and 3 have equal importance in the system state decomposition $|\Psi\rangle=\Sigma c_n|E_n\rangle$, where c_n represents the weight of the nth eigenstate $|E_n\rangle$ in the system state] which are both combinations of vectors with spins up and down. The system energy $\langle\Psi|H|\Psi\rangle$ is slightly above this approach between levels 2 and 3 [Fig. 2(b)]. This fact permits a resonant tunneling of the magnetization. However, for fields lower than the resonant one, the system state is essentially the second eigenstate [Fig. 2(c)], which corresponds to all spins down (this can be understood realizing that the second eigenstate energy increases with the magnetic field, which implies that the eigenstate has mainly all spins against the field, in the same way that when the energy of an eigenstate decreases with the field, such behavior suggests that the important vectors in the eigenstate decomposition are those with all the spins in the field direction), and for fields above the resonant field the system state is fundamentally the third eigenstate [Fig. 2(c)], which corresponds again to spins down. The role played by the total-energy curve can be understood in a qualitative way: if it is very close to an eigenvalue curve, then most of the weight on the state is in

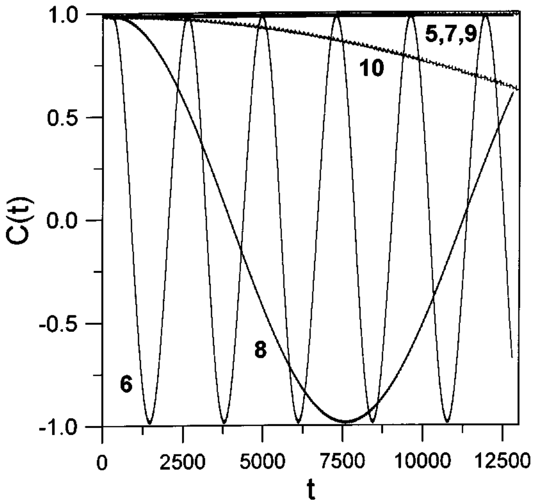

FIG. 4. Symmetrized correlation function $C(t)$ at zero field for clusters containing from five up to ten spins, with $J_x=0.9$, $J_y=0.85$, and $J_z=1.0$. $C(t)$ for ten-spins cluster oscillates between $+1$ and -1, but due to its large period, only part of the oscillation is shown in the figure.

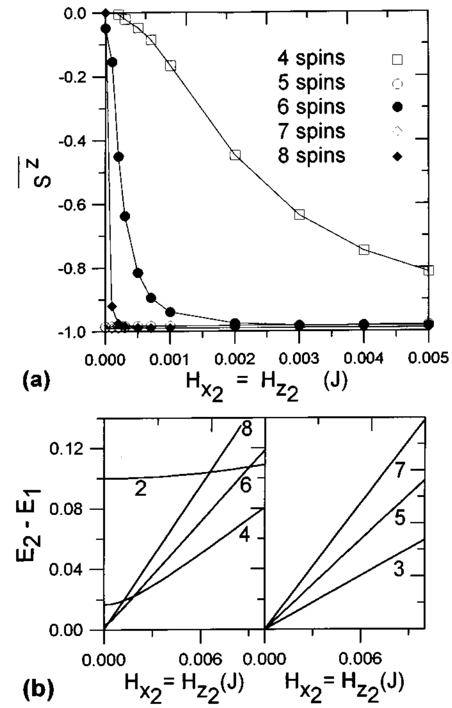

FIG. 5. (a) Same as Fig. 3 but for small magnetic fields. (b) Energy splitting between the first two eigenstates as a function of the applied magnetic field for systems with N spins, $2\leqslant N\leqslant 8$. Systems with an even number of spins have a nonzero splitting. The small splitting for $N=8$ cannot be resolved on the scale of the figure.

that eigenvalue. At the resonant field, the system energy is very close to the involved eigenvalues and the system state is essentially a superposition of both levels. The above explanation fits more dramatically to sharper resonances like the one shown in Ref. 18 or that appearing in the cluster of eight spins for the same parameters (Fig. 3), but we have chosen this picture since we are specially interested in discussing the zero-field resonance. We want to point out that although the cases presented in this paper do not exhibit more than one off-zero-field resonance, unpublished calculations for larger clusters with uniaxial anisotropy show more than one resonance corresponding to coherent quantum tunneling, as is the case of, for instance, a cluster with eleven spins forming an open line which exhibits six resonances.

In the $\overline{S^z}$ curve of Fig. 2(a) a sharp peak at $H=0$ can be observed which corresponds to resonant coherent quantum tunneling of the magnetization since only at such a field does the correlation function present a pure sinusoidal oscillation between $+1$ and -1 (Fig. 4). All the spins coherently tunnel back and forth between the two easy directions. It happens that the system state at zero field is a combination of just the first two eigenstates of the system [see their energies in Fig. 2(b) and their weights in Fig. 2(c), both indicated with a circle] and these eigenstates consist essentially of the symmetric and antisymmetric combination of the vectors $|all\ spins\ up\rangle$ and $|all\ spins\ down\rangle$. This fact as well as the proximity of the system energy to the involved eigenvalues permits the phenomenon of coherent tunneling of the spins. As the magnetic field becomes present the system state turns out to be essentially the second eigenstate [Fig. 2(c)], which does not anymore correspond to an antisymmetric combination with equal weights of the vectors $|all\ spins\ up\rangle$ and $|all\ spins\ down\rangle$ but to a state with all spins down (the energy of the second eigenstate increases with H). Similarly, the first eigenstate becomes essentially a state with all spins up when a small magnetic field forming an angle of 45° with the z axis is applied. Let us remind the reader that the system does not escape to the ground state when a resonant field is applied (zero field in this case) since no dissipation is taken into account in this work.

It has been shown that the resonant coherent quantum tunneling of all the spins occurs at zero field in a cluster of six spins forming a line. Studying clusters with different numbers of spins, we reach the conclusion that this resonant coherent quantum tunneling at zero field occurs only for systems with a general biaxial anisotropy that have an even number of spin-$\frac{1}{2}$ particles whereas systems with an odd number do not exhibit the above-mentioned phenomenon. In Fig. 3, the $\overline{S^z}$ curves for $N=4$, 6, and 8 show a peak at $H=0$ in contrast to the curves for $N=5$ and 7, which do not present any peak at $H=0$. To the contrary, both clusters with even and odd numbers of spins can present off-zero-field resonances corresponding to coherent tunneling as the clear ones shown in Fig. 3 for $N=8$ and 7. This fact means that there is no parity effect at finite magnetic field. The correlation function at zero field for clusters with a number of spins from five to ten is presented in Fig. 4 showing a pure sinusoidal oscillation in the case of even N (six, eight, and ten—for $N=10$ the period is so large than only a small part of the oscillation appears on that scale) and a different behavior for odd N (five, seven, and nine). The period T of the oscillations of $C(t)$ for even N depends on the splitting ΔE between the two resonant states (values obtained fit very well to $T=2\pi\hbar\Delta E^{-1}$) and is related to the resonance width in the $\overline{S^z}$ curve. In Fig. 5, a zoom of the $\overline{S^z}$ curves for small fields as well as the energy splitting between the first two

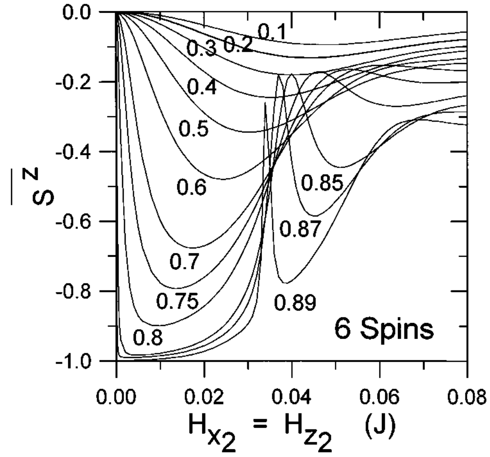

FIG. 6. $\overline{S^z}$ as a function of the applied magnetic field for a system of six spins forming an open line for different values of J_y, with $J_x=0.9$ and $J_z=1.0$ (easy axis along z and xz as easy plane).

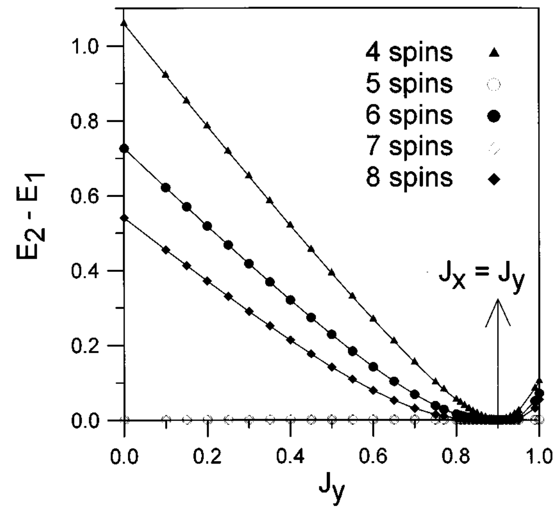

FIG. 7. Energy splitting between the first two eigenstates as a function of J_y ($J_x=0.9$, $J_z=1.0$) for systems with N spins, $4\leqslant N\leqslant 8$. $J_y=J_x=0.9$ corresponds to uniaxial anisotropy.

eigenstates are shown. Figure 5(b) explains why in the case of even N the period of the oscillations in $C(t)$ increases with the number of spins since the energy splitting between the resonant states becomes smaller [Fig. 5(b)]. As a consequence, the zero-field resonance becomes narrower for larger clusters with an even number of spins [Fig. 5(a)].

The reason why there is a total spin-parity-dependent behavior can be found in the spectrum. Clusters with an odd number of spins present at zero field a degeneration between the first two eigenstates [Fig. 5(b)], which consist of all spins up and all spins down, respectively, so that no tunneling can occur. On the other hand, clusters with an even number of spins have a splitting in energy between the first two eigenstates [Fig. 5(b)], which correspond, as previously mentioned, to symmetric and antisymmetric combinations of all spins down and all spins up so that tunneling between the two states can take place. The sinusoidal aspect of the correlation function for the resonances indicates that the tunneling is coherent.

Finally, we analyze the role played by the anisotropy in the phenomenon of resonant coherent quantum tunneling at zero field (for clusters with an even number of spins). All cases mentioned until this moment have considered the values $J_x=0.9$, $J_y=0.85$, and $J_z=1.0$ (z easy axis, xz easy plane) for the exchange constants. Decreasing the value of J_y we can reach larger transverse anisotropies for comparatively smaller anisotropies in the easy plane xz, that is, as J_y decreases the spins are more strongly forced to lie in the easy plane. It can be seen in Fig. 6 where $\overline{S^z}$ is plotted versus the magnetic field for different values of J_y ($J_x=0.9$ and $J_z=1.0$) that, in a cluster of six spins forming a line, the zero-field resonance becomes broader and shorter as the transverse anisotropy increases (J_y decreases). In the presence of applied magnetic fields the reversal of the magnetization becomes easier (although not coherent) as the transverse anisotropy gets larger. For instance, at $H_x=H_z=0.02$ the mean value in time of the total spin z component for a cluster of six spins with $J_y=0.85$ is $\overline{S^z}=-0.940\,25$ whereas for $J_y=0.6$, $\overline{S^z}=-04651$. The first value corresponds to an evolution of the spin's z component between approximately -1 and -0.88 whereas the second one corresponds to an evolution between -1 and 0.07.

At zero field, the correlation function remains sinusoidal with decreasing J_y although the period of the oscillations becomes smaller and the oscillations do not appear so pure. In Fig. 7 the energy splitting between the first two eigenstates at $H=0$ as a function of J_y for clusters with N spins from 4 up to 8 is presented. It is confirmed again that clusters with an odd number of spins have a degeneration between the first two levels. For even N it can be observed that the splitting increases with decreasing J_y, which explains the increasing width of the resonance and the smaller period of the oscillations in $C(t)$ at $H=0$. Also it is appreciated again that the splitting decreases for larger systems of spins.

In the limit when $J_y=J_x$ ($=0.9$) there is uniaxial anisotropy and the first two levels are degenerate corresponding, respectively, to all spins up and all spins down: no tunneling occurs. When J_y decreases, the degeneration is broken by tunneling and the first two eigenstates at $H=0$ consist of symmetric and antisymmetric combinations of all spins down and all spins up. The weight of the ground state becomes more important in the system state as the anisotropy increases (J_y decreases) and although the first two levels remain symmetric and antisymmetric, respectively, the weights of the vectors $|all\ spins\ up\rangle$, and $|all\ spins\ down\rangle$ become lower (still the largest in the system states decomposition) and new vectors with couples and groups of spins up and down become relevant.

As a conclusion, we have performed quantum dynamical calculations to analyze the reversal of magnetization for systems of a few spin-$\frac{1}{2}$ particles with a general biaxial anisotropy in the presence of an external field at $T=0$ and with no dissipation. The reversal of the spins in small clusters is shown to be coherent only for some specific resonant fields, which in the case of systems with general biaxial anisotropy includes the absence of applied magnetic field if the total number of spin-$\frac{1}{2}$ particles is even. Quantum tunneling of the magnetization direction at zero field is shown to be spin-parity dependent: whereas clusters with an odd number of particles do not exhibit quantum tunneling of the magnetization due to the degeneration between the first two levels, clusters with an even number of spins present resonant coherent quantum tunneling of the magnetization. Contrary to the zero-field resonance, the phenomenon of resonant coherent quantum tunneling of the magnetization at finite magnetic field is shown not to have any detectable parity dependence: both clusters with even and odd numbers of spins can present off-zero-field resonances corresponding to coherent quantum tunneling.

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