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Quantum dynamical calculations on the magnetization reversal in clusters of spin-1/2 particles: Resonant coherent quantum tunneling

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In the present work the reversal of magnetization and the coherence of tunneling when an external magnetic field is rotated instantaneously are studied in systems of a few spin-1/2 particles described by an anisotropic Heisenberg Hamiltonian at $T=0$. Our calculations demonstrate that this model for small magnetic particles exhibits collective tunneling of the magnetization only for some specific resonant values of the applied magnetic field. These resonant effects occur at fields much lower than the values corresponding to the vanishing of the barrier in the Stoner-Wohlfarth model. The former model is at variance with the exact calculations presented in this paper.

I. INTRODUCTION

In the last years, the study of macroscopic quantum tunneling[1,2] (MQT) of magnetization has received a lot of attention both for fundamental and technological reasons. Most fundamental aspects are connected with the quantum limit and the quantum theory of measurement, and more applied ones are connected with the magnetization reversal mechanisms and their dynamics.[3,4] The ability to miniaturize magnetic materials and study the magnetic properties of a single isolated particle has revealed new classical and quantum phenomena[5,6] that questions the present understanding of the fundamentals of magnetism. When quantum effects are significant, classical interpretations become useless, and we have to give up what is known as macroscopic realism.[1] In addition, MQT of the magnetization when an external magnetic field is suddenly rotated might be of crucial interest in the future for information storage[5] in magnetic particles.

MQT (Refs. 1 and 2) consists of the tunneling of a macroscopic variable through the barrier between two minima of the effective potential of a macroscopic system. For small single-domain ferromagnetic clusters, these minima correspond to the two states of opposite magnetization, and the barrier is proportional to the anisotropy in the exchange interaction. When there is a repeated coherent tunneling back and forth between the two wells, we have a case of macroscopic quantum coherence (MQC), and all the spins behave in the same way.

The Stone-Wohlfarth (SW) model,[7] because of its success in the explanation of many classical magnetic phenomena, provided the idea that the dynamics of small magnetic particles in the single-domain regime would keep its simplicity. However, the SW model has been found inadequate for explaining many details in experimental systems.[8] The quantum mechanical effects have been studied theoretically by the quantization within a path integral formalism of the classical micromagnetic theory of magnetic dynamics.[9,10] In the semiclassical approximation made by Chudnovsky and Gunther[9] the probability of tunneling of the magnetization in a single-domain particle through an energy barrier between easy directions is calculated for several forms of magnetic anisotropy, and uniform and coherent rotation of all the spins is imposed; that is, spins are considered to behave dynamically as a single quantum spin. They show that in addition to superconducting devices, single-domain magnetic particles represent a rich field for MQT study.

It is generally assumed that the electron spins of a single-domain particle are constrained by the exchange interaction between electron spins to behave dynamically as a single quantum spin. This interaction was first proposed by Heisenberg and would explain that the single-domain particle could tunnel from one macrospin state to another. Then the coherent action of the huge number of degrees of freedom would provide an example of MQT. However, recent experiments[11] have found that the reversal mechanism of the magnetization in single-domain particles could differ from the simple uniform rotation. This behavior has been explained in terms of the different initial magnetization between inner and outermost spins when the external field is reversed.[12] Furthermore, extensive theoretical work has been presented to understand why the magnetization of elongated particles is able to rotate incoherently.[13]

The calculations presented in this work show that for the model considered representing small clusters of spin-1/2 particles at $T=0$, the exact quantum evolution of the spins is, in general, noncoherent, but it is found that each model exhibits collective tunneling of the magnetization only for a specific resonant value of the applied magnetic field. This coherent quantum tunneling occurs at fields much lower than the values corresponding to the vanishing of the barrier in the Stoner-Wohlfarth model. The former model is at variance with the exact calculations presented in this paper, and, at fields close to the disappearance of the barrier where semiclassical treatments[9] are applied, such important coherent resonant tunneling is not found at all.

II. MODEL AND METHOD

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

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

where σ_i^α ($\alpha = x,y,z$) are the Pauli-spin matrices at site i related to the spin operators by

$$\mathbf{S} = \frac{\hbar}{2} \boldsymbol{\sigma}, \quad (2)$$

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. The magnetic fields considered are within the xz plain without any generality loss and anisotropy is taken into account by making J_x, J_y, and J_z different. In the text we speak about magnetization and total spin without distinction; notice that they are related by the Bohr magneton μ_B and the electron-g factor.

In order to know which parameters have decisive influence on the magnetization quantum evolution, we have studied different situations, varying (a) the size of the cluster (number of particles N) and its geometrical configuration, (b) the kind and value of the anisotropy, and (c) the value, direction, and reversal mechanism of the applied magnetic field. Because of the limits of computational resources, we have limited ourselves to systems of just a few particles, with a uniaxial anisotropy Δ in the z direction [$J_x = J_y < J_z = J$, $\Delta = (J_z - J_x)/J$] and to instantaneous rotations of the magnetic field. The range of the parameters is $0.01J \leqslant \Delta \leqslant 0.1J$ for the anisotropy and $0 \leqslant H \leqslant 0.2J$ for the magnetic field, considering in most cases $H < \Delta$. We study how the total magnetization and the expectation values of the spins components evolve when the magnetic field is rotated instantaneously. Tunneling and the coherence in the different spins' evolution are analyzed.

The temporal evolution of the system is calculated by a numerically exact solution of the time-dependent Schrödinger equation.[14] 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

$$|\Psi(t)\rangle = e^{-it\mathscr{H}} |\Psi(t=0)\rangle, \quad (3)$$

and to understand the main features of the system, the temporal evolution of the α ($\alpha = x,y,z$) component of each spin can be obtained as follows:

$$\langle S_i^\alpha \rangle (t) = \langle \Psi(t) | S_i^\alpha | \Psi(t) \rangle. \quad (4)$$

Actually, the expectation value of σ_i^α instead of S_i^α is calculated, but as it has been said they are simply related. The essential information of the problem analyzed is concentrated on the z-component of the spins. In addition, the components of the total spin are obtained by

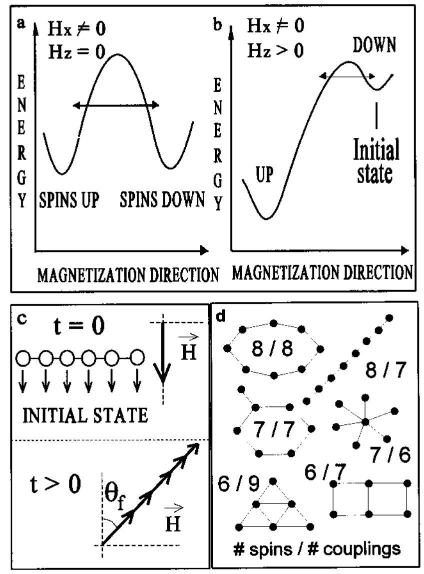

FIG. 1. Schematic representation of the energy barrier for the magnetization in presence of an applied magnetic field with components (a) $Hx \neq 0$, $Hz = 0$, (b) $Hx \neq 0$, $Hz > 0$, (c) the physical situation considered in computation, and (d) examples of clusters with different number of spins and different geometrical forms.

$$\langle S^\alpha \rangle (t) = \left\langle \Psi(t) \middle| \frac{1}{N} \sum_i S_i^\alpha \middle| \Psi(t) \right\rangle, \quad (5)$$

N being the number of particles.

The spin system of a single-domain particle having uniaxial anisotropy in the presence of a field perpendicular to the easy axis has a symmetric, double-well potential [Fig. 1(a)]. The magnetization of this system then exhibits a repeated tunneling back and forth between the two wells in a coherent fashion.[5] This is known as macroscopic quantum coherence (MQC). Usually, it is very difficult to observe MQC, since the energy barrier is too large and the tunneling probability decreases exponentially with the barrier height. By applying a dc magnetic field directed along the easy axis, the magnetization is biased in the direction of the field and the barrier is reduced [Fig. 1(b)]. Obviously, the tunneling rate increases,[5] permitting the MQT phenomenon.

We need to introduce the two-time correlation function of the magnetization,[1] 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 $C(t)$ defined as

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

has been calculated.

There is a negligible probability of finding the magnetization in other than an up or down direction if the energy wells are deep. With negligible dissipation present, coherent tunneling back and forth between the two states leads to a sinusoidal oscillation of $C(t)$ at a frequency twice the MQT tunneling rate Γ. For two measurements of the magnetization separated by the time interval t, one should have[1,2]

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

This equation predicts a resonance for the Fourier transform of $C(t)$ at a frequency $\omega_R = 2\Gamma$. As the fluctuation-dissipation theorem[6] shows that the frequency-dependent magnetic susceptibility $\chi''(\omega)$ is essentially the Fourier transform of the correlation function, the susceptibility should exhibit this resonance, and this will happen for small enough fields so that the energy minima of the two macroscopic states are equivalent. In experiments with superconducting quantum interference device microsusceptometers[15] in which $\chi''(\omega)$ is measured, a well-defined resonance has been found, and it is tempting to be associated with a MQC phenomenon. 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 resonant frequency is very sensitive to small fields. Many research works in MQC have studied the effect of dissipation on this resonance.[1]

In the present work we have assumed that there is no dissipation, $T=0$, and we have considered applied magnetic fields for which the energy barrier is present, giving rise to the appearance of tunneling phenomena. The results we have found show a qualitatively different landscape to what has been explained above: there is essentially a sharp resonance corresponding to collective tunneling of the magnetization back and forth between its two opposite directions but only for a particular magnetic field, whereas for lower and larger fields this phenomenon does not appear.

III. NUMERICAL RESULTS

Following the general method described in the preceding section, the exact spin propagation and the coherence in the magnetization tunneling when the external magnetic field is rotated instantaneously are studied. In our particular model, at $t=0$ there is a field applied along the z direction, $\mathbf{H}_1=(0,0,H_{1z})$ with $H_{1z}<0$. Then the ground state of the ferromagnet has all spins down, and we prepare the system in this 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 [see Fig. 1(c)]. If the magnetic field is reversed, nothing happens in the exact propagation because the ground state of the first Hamiltonian is also an eigenstate of the second one, although not the ground state.

Generally, since not all the particles in the cluster are equivalent (except when they form a ring) the spins evolve in a noncoherent way although the nonuniformity is small. We can appreciate this fact when calculating the spin evolution and its mean value in time. The importance of the noncoherent behavior depends on the magnetic field value and on the anisotropy. It can be said that the spins precess about an effective field, and, since the system state has components with spins up and down, there is some probability of magnetization tunneling. However, we will see that only for certain fields a collective tunneling of the magnetization occurs.

We have calculated the expectation value of S_i^z for each nonequivalent spin i (i.e., spins with different numbers of couplings) averaged over time

$$\bar{S}_i^z = \frac{1}{T}\int_0^T dt \langle S_i^z(t)\rangle \tag{8}$$

for different values of the magnetic field $\mathbf{H}_2$, and the same magnitude for the total spin S^z. In addition, we monitor the time evolution of $\langle S_i^\alpha(t)\rangle$ and $\langle S^\alpha(t)\rangle$, $\alpha=x,y,z$ as well as the correlation function $\langle S^\alpha(0)S^\alpha(t)\rangle$ for $\alpha=x,y,z$. Of course, we also compute the eigenvalues and eigenstates for each value of the magnetic field considered.

In the present calculations, we have thoroughly analyzed clusters with uniaxial anisotropies $\Delta=0.1$, 0.05, and 0.01, with N spins $(2<N<9)$ forming different geometrical configurations and in presence of a wide range of magnetic fields with directions given by angles $\theta_f=45°$, 30°, and 15°. Depending on the value of the magnetic field, the barrier between the two directions of the magnetization can exist or not, and this way we can speak about two regions: (a) a tunneling region when there is a barrier between the two wells and (b) a nontunneling region when that activation barrier has vanished.

Let us concentrate on the results for uniaxial anisotropy $\Delta=0.1$ $(J_x=J_y=0.9J_z)$ and a magnetic field forming an angle $\theta_f=45°$ with the z direction, $\mathbf{H}_2=(H_{x2},0,H_{z2})$ with $H_{x2}=H_{z2}$. The remarkable result obtained is the following: clusters with five, six, seven, and eight particles and with different geometrical forms [chain, ring, and others–see Fig. 1(d)] present a pronounced resonance in the curves of $\bar{S}_i^z$ and $\bar{S}^z$ in terms of $H_{x2}=H_{z2}$ for a specific magnetic field that clearly falls in the tunneling region (a). Evidence of the resonance does not come directly from the time-averaged spin-z component, but this magnitude shows this behavior very clearly. We have found that these resonances correspond to pure sinusoidal oscillations in the correlation function as it must occur when there is MQC. However, for points around these resonances the correlation function does not present this cosine at all. Clusters with less than five spins do not show this behavior, and the reason will be explained later in terms of the spectrum. When the magnetic field is large enough the activation barrier vanishes [region (b)], the behavior of the spins is dominated by the field and it becomes less interesting. Then, the spins seem essentially to precess around the magnetic field direction, although their behavior is not that simple. Later on it will be shown that the resonant coherent quantum tunneling is a general feature of the model, and it is not limited to these particular anisotropy values and orientations of the magnetic field.

Now we will show in detail the results for a cluster of six spins forming a line with $\Delta=0.1$ and $\theta_f=45°$ (see Fig. 2) to illustrate the general behavior just anticipated. $\bar{S}^z$ is shown in Fig. 2(a), and a sharp resonance appears for a specific field falling in the tunneling region. At this resonance, the correlation function presents a sinusoidal oscillation [(i) in Fig. 2(b)] with positive and negative values, whereas for fields just around this specific resonant field this function stays close to the value 1 and exhibits a complex behavior [(ii) and (iii) in Fig. 2(b)]. The sinusoidal oscillation indicates that there is a collective tunneling between the two directions of the magnetization in contrast to the aspect of the correlation

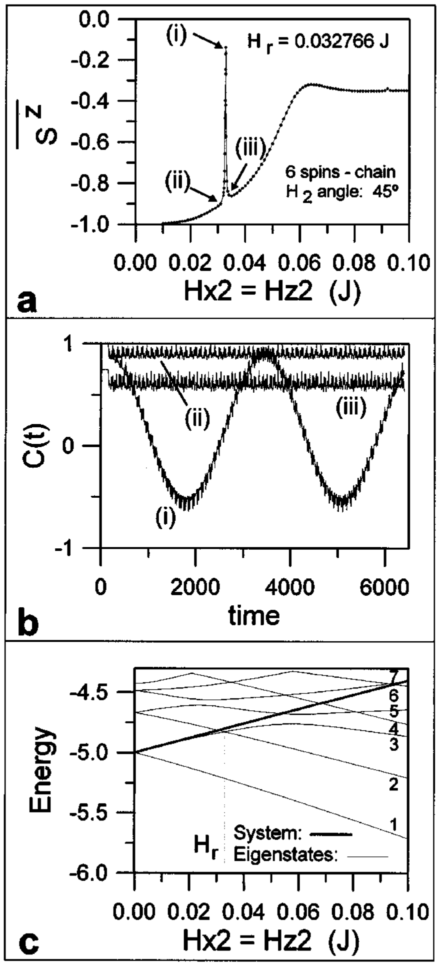

FIG. 2. (a) Dependence of $\overline{S^z}$ for each different spin i on the size of the second magnetic field for a system of six spins forming a chain, with $\Delta=0.1$ and $\theta_f=45°$; (b) symmetrized correlation function for the resonant field $H_r=0.032\,766J$ (i) and two fields around it, (ii) $H=0.031J$ and (iii) $H=0.035J$. Curve (iii) has been shifted 0.25 in the y axis in order to clarify the picture. (c) System energy $\langle\Psi|H|\Psi\rangle$ and energy of the first eigenstates of the system as a function of the magnetic field.

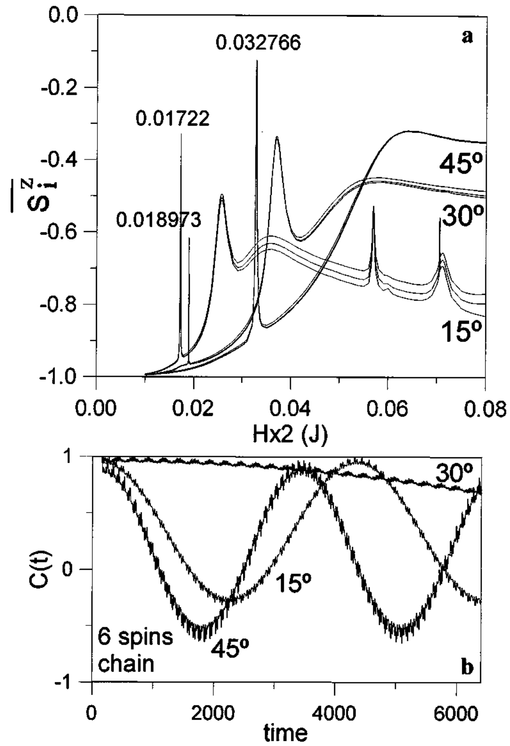

FIG. 3. (a) $\overline{S_i^z}$ for each different spin (i) and (b) symmetrized correlation function $C(t)$ as a function of the second magnetic field for a cluster of six spins forming a chain with $\Delta=0.1$ when the second magnetic field direction forms angles of 45°, 30°, and 15° with the z axis.

function for nonresonant fields, which shows that in these cases the magnetization is not reverted.

In order to understand why a particular magnetic field provokes the resonant coherent quantum tunneling, we have studied the system spectrum calculating its eigenstates energies for each magnetic field applied. It can be seen in Fig. 2(c) that the specific field that produces the resonance makes the energies of the second and third eigenstates of the system practically equal, which correspond essentially to all spins in one direction and in the opposite, respectively. This fact permits a resonant tunneling of the magnetization for a determined field in each case. In Fig. 2(c) the energy of the first eigenstates of the system and the system energy, $\langle\Psi|H|\Psi\rangle$, are plotted as a function of the magnetic field. When the energy of an eigenstate increases with the field H, it means that the eigenstate has mainly all spins against the field, that is spins down, while when the eigenstate energy decreases with H, this fact suggests that the vectors with relevant weight in the eigenstate decomposition are those with all spins up (same direction as the field). For fields below the resonant one, H_r, the second eigenstate corresponds fundamentally to all spins down, and the third to all spins up. At $H=H_r$, their energies coincide, and the two corresponding eigenstates have components with spins up and down, whereas when $H>H_r$, the energy levels seem to be repealed and the opposite to $H<H_r$ occurs, the second level spins up and the third down. The system energy calculated as the expectation value of the Hamiltonian is slightly above the degeneration point at $H=H_r$. The importance of each eigenstate for the system state can be calculated, and this can explain why the resonance occurs: below the resonant field, the system state is essentially the second eigenstate with all spins down; at the resonant field, the second and third eigenstates have an equivalent importance both with up and down spins, and above H_r, the relevant eigenstate is the third one with all the spins down again, as can be corroborated in the representation of S^z, Fig. 2(a). Notice that the first eigenstate does not contribute because we do not take into account dissipation. The behavior shown is general for the cases considered. It must be said that the levels do not cross; there is a small splitting ΔE between their energies that is related to the tunnel frequency and, in consequence, to the oscillating period T of the correlation function $C(t)$ by

$$T=2\pi\hbar\Delta E^{-1}. \tag{9}$$

The values of T and ΔE fit this formula very well.

Until now, the case exhibited corresponded to anisotropy $\Delta=0.1$ and a direction of the second Hamiltonian forming an angle of 45° with the z axis. Other anisotropy values and

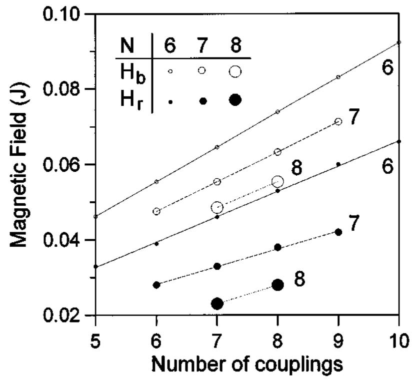

FIG. 4. Dependence of the resonant field (solid circles) on the number of couplings for clusters with six, seven, and eight particles and comparison with the values of the field needed to vanish the activation barrier (open circles).

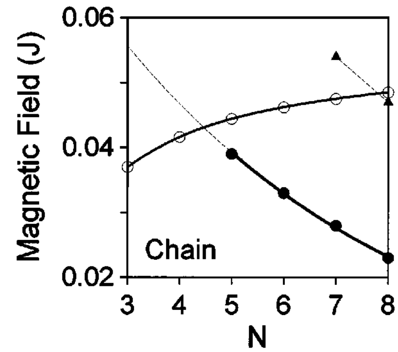

FIG. 5. Dependence of the resonant field H_r (solid circles) and the field needed to cause the disappearance of the activation barrier H_b (open circles) on the number of spins for a fixed geometrical configuration. The solid triangular symbols correspond to the second peak in the $\overline{S_i^z}$ curve for seven and eight spin clusters.

other directions have been studied. In Fig. 3(a) we present the curves of $\overline{S_i^z}$ when the direction of $\mathbf{H}_2$ forms angles θ_f=45°, 30°, and 15° with the z axis and the correlation function corresponding to the leftmost peak of each curve. The equivalent sharp peaks are the leftmost ones. These are resonances whose correlation functions also exhibit a pure cosine [see Fig. 3(b)], whereas the fields around them lead to functions with a completely different behavior. The other sharp points of the 30° and 15° curves do not present that pure cosine; they show more complex oscillating behaviors. It is surprising that the correlation function corresponding to the 30° resonance also presents a perfectly sinusoidal oscillating behavior but with a much larger period [in Fig. 3(b) it only appears at the first part of the cosine]. This long period is because of the very small splitting between the second and third eigenstates when θ_f=30°.

Another important parameter is the value of the anisotropy. The remarkable resonance that has been found with $\Delta=0.1$ ($J_x=J_y=0.9J_z$) is also found when $\Delta=0.05$ ($J_x=J_y=0.95J_z$), and again it corresponds to an oscillating behavior in $C(t)$. The results for $\Delta=0.05$ are similar, and therefore they are not shown. This way it can be said that the resonance found is a general feature of the system considered; it appears for several sizes with any geometrical configuration, for different values of the anisotropy, and for all the directions of the magnetic field $\mathbf{H}_2$ studied.

In Fig. 4 we show the dependence of the resonant field H_r on the number of couplings for clusters with six, seven, and eight particles and the comparison with the values of the field H_b that makes the barrier disappear in a classical model of uniform rotation. Different geometrical configurations have been analyzed [see Fig. 1(d) as an example], N_c/N being the rate between the number of couplings N_c and the number of spins N and in consequence an indication of how compact the cluster is. In our study $H_r<H_b$ for all the systems considered and their dependence on the number of couplings seems to be analogous (same scope). In addition to this, we have also calculated the dependence of H_r and H_b on the number of spins ($N<9$) for the same geometrical configuration (chain). The result obtained does not seem to be trivial at first sight. In Fig. 5 this dependence can be seen, and again $H_r<H_b$ for systems with five spins and more. However H_r and H_b have an opposite dependence on the number of spins: while H_b increases with the number of spins, since the barrier height is proportional to N, H_r decreases with it. The difference between H_r and H_b increases with N, and in consequence the resonance is situated further from the region where the barrier vanishes and the semiclassical approaches are applied.[9] The tendency shown by the two curves can explain why clusters with less than five particles do not present such resonance, as well as the fact that the second and third eigenstates energies do not get close but keep a considerable gap between them. It is of interest to calculate the value of H_r for N larger than 9 in order to know its behavior when N increases. These calculations will be realized shortly using Suzuki's fourth-order fractal product formula[14,16,17] to solve the time-dependent Schrödinger equation. An interesting point is that when N increases and so the separation between H_r and H_b becomes larger, new peaks or resonances appear. We have observed this behavior in clusters with eight spins. The field corresponding to the second peak in the eight-spin curve is below H_b, whereas in the seven-spin curve the second peak field is above H_b (see Fig. 5).

Now we concentrate our attention in a system that exhibits this oscillating behavior in the correlation function $C(t)$. The spins are tunneling between the up and down position, but how does this happen? If we look at the state of the system at different times, we can see that at a time when $C(t)$ is close to 1 [(iii) in Fig. 6], all the spins are down: the only important component of the decomposition of the system state in vectors is that with all spins down. For any minimum of $C(t)$, as (i) in Fig. 6, the relevant components of the system state are those with all spins up, and all spins up except one. This is also seen in Fig. 6. In the resonance case there is a collective tunneling of the magnetization, since at two different times the system state is a linear combination whose relevant terms correspond to vectors with most of the spins down or to vectors with most of the spins up. The sinusoidal aspect of $C(t)$ is a sign of the maximum coherence in the spins reversal. In a nonresonant case, only the terms with all the spins in the direction of the initial situation are important at any time, and so there is not a collective tunneling of the spins.

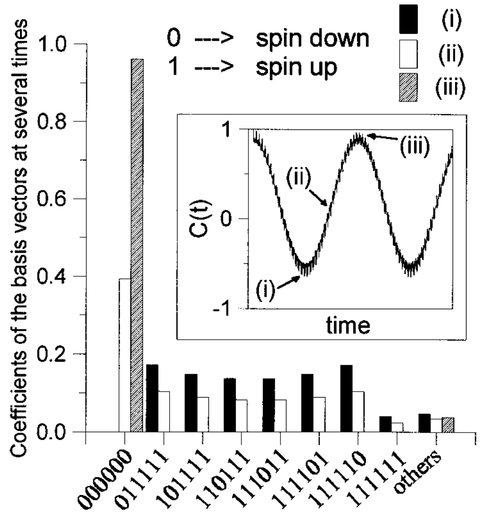

FIG. 6. Decomposition of the system state at several times of the evolution on the basis with vectors with spins up (1) and down (0) for a cluster of six spins forming a chain with $\Delta=0.1$ and $\theta_f=45°$; (i), (ii), and (iii) correspond to three moments of the evolution, namely, the minimum, middle point, and maximum of the symmetrized correlation function, respectively.

IV. CONCLUSIONS

We have studied the reversal of magnetization and the coherence of tunneling when an external magnetic field is rotated instantaneously in systems of a few spin-1/2 particles described by an anisotropic Heisenberg Hamiltonian at $T=0$. We consider clusters in the presence of an applied magnetic field along the easy axis so that the system is prepared with all the spins along that direction. Then the magnetic field is rotated instantaneously, and the exact propagation of the system is calculated. We analyze the mean value in the time of the components of each spin and the total spin as a function of the magnetic field, the correlation function, and also the spectrum in terms of the quantum coherence of the spins, that is, the coherent tunneling back and forth between the two wells of the double-well potential. For systems with five particles and more, for any geometrical configuration and for different anisotropy values a sharp resonance in tunneling of the magnetization direction is found for a fixed and unique resonant magnetic field in each system. We have found that the resonant field is lower than the field needed for the activation barrier to vanish. The correlation function at that remarkable resonance consists of a perfectly sinusoidal oscillation indicative of a collective tunneling of the magnetization. Outside of this resonant field the correlation function does not present a simple behavior. The effect of the magnetic field on the resonant tunneling has been analyzed in terms of the spectrum. The structure of the spectrum explains why the resonance is not found for clusters with less than five particles. Our calculations demonstrate that the model for small magnetic particles studied in the present work exhibits collective tunneling of the magnetization only for some specific resonant values of the magnetic field, at variance with the Stoner-Wohlfarth model, which predicts coherent rotation at all fields. Therefore, we want to make the reader notice the precaution needed in undertaking semiclassical studies of MQC.

Note added in proof. In the case of Fig. 2(b), we have recently calculated up to 200 000 time steps. The result of the coherent oscillation remains present.

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[1] A. J. Legget *et al.*, Rev. Mod. Phys. **59**, 1 (1987).
[2] P. C. E. Stamp, E. M. Chudnovsky, and B. Barbara, Int. J. Mod. Phys. B **6**, 1355 (1992).
[3] W. Wernnsdorfer *et al.* (private communication).
[4] W. Wernnsdorfer *et al.*, J. Magn. Magn. Mater. **145** (1995).
[5] L. Gunther, Phys. World **3**, 28 (1990).
[6] D. D. Awschalom, D. P. Di Vincenzo, and J. F. Smyth, Science **258**, 414 (1992).
[7] E. C. Stoner and E. P. Wohlfarth, Philos. Trans. R. Soc. London, Ser. A **240**, 599 (1948), reprinted in IEEE Trans. Magn. **MAG-27**, 3475 (1991).
[8] D. D. Awschalom and D. P. Di Vincenzo, Physics Today **48** (4), 43–48 (1995).
[9] E. M. Chudnovsky and L. Gunther, Phys. Rev. Lett. **60**, 661 (1988).
[10] M. Enz and R. Schilling, J. Phys. C **19** L-711 (1986); G. Scharf, W. F. Wreszinski, and S. L. Van Hemmen, J. Phys. A **20**, 4309 (1987).
[11] M. Lederman, D. R. Fredkin, R. O'Barr, S. Schultz, and M. Ozaki, J. Appl. Phys. **75**, 6217 (1994).
[12] P. A. Serena and N. García, in *Quantum Tunneling of the Magnetization, NATO ASI Series E*, edited by B. Barbara and L. Gunther (Kluwer Academic, Norwell, MA, in press); D. García-Pablos, P. García-Mochales, N. García, and P. A. Serena, J. Appl. Phys. (to be published).
[13] H. B. Braun, Phys. Rev. Lett. **71**, 3557 (1993).
[14] P. de Vries and H. De Raedt, Phys. Rev. B **47**, 7929 (1993).
[15] D. D. Awschalom *et al.*, Phys. Rev. Lett. **68**, 3092 (1992).
[16] M. Suzuki, Prog. Theor. Phys. **56**, 1454 (1976).
[17] H. De Raedt, Comp. Phys. Rep. **7**, 1 (1987).