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Dynamical calculations on the reversal of single quantum spins: Quantum coherence

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Quantum coherence in small single-domain ferromagnetic particles is studied using dynamical calculations of the reversal of a single quantum spin resulting from a rotation of an external magnetic field, for $\frac{1}{2}\leqslant S\leqslant 75$. The analysis of the time evolution of the spin, the temporal correlation, and the spectrum leads to the conclusion that, for small spin $S\leqslant 15$, coherent tunneling back and forth between the easy directions occurs at some specific resonant fields only. Away from resonance, quantum coherence in the spin temporal evolution is absent. These results are in qualitative accordance with those obtained for a Heisenberg Hamiltonian with exact quantum calculations and are at variance with semiclassical approximations. However, as the spin becomes larger, the number of resonant fields increases, the resonances become very small, and coherence in time evolution is lost for any value of the magnetic field. [S0163-1829(97)08101-0]

Both fundamental[1] and applied[2] questions arise when considering the phenomena of macroscopic quantum tunneling (MQT) and macroscopic quantum coherence (MQC) of the magnetization[3,4] in small magnetic particles, generating great activity in nanoscale investigations over the last few years.[5] The magnetic anisotropy present in single-domain magnetic particles creates two or more energy minima for the total magnetization. Tunneling of the total spin between such low-energy directions is attracting much attention. In particular, the resonance between equivalent easy directions is known as MQC,[3,4] and it implies that all the spins coherently tunnel back and forth between the two minima of the energy. The effect of dissipation in this phenomenon has been actively investigated but will not be considered in this work.[3]

Most theoretical work on quantum tunneling in small magnetic particles is based on the assumption that the semiclassical limit is valid for such particles. In this limit, two approaches, one using a generalization of the conventional WKB method[6,7] and the other[8–11] using the path-integral formalism of quantum mechanics, are applied to a single spin model (SSM). On the other hand, quantum dynamical calculations without applying any assumption have been carried out to investigate the reversal of magnetization in clusters of a few spin-$\frac{1}{2}$ particles represented by a Heisenberg Hamiltonian.[12–14] The results obtained from a numerically exact solution of the time-dependent Schrödinger equation for systems of a few spins are at variance with those obtained with semiclassical approximations for a SSM.

In the present work, the total spin of the single-domain ferromagnetic particle formed by the alignment of the atoms spin is treated as a single quantum spin, adopting the general but sometimes questioned[15–19] assumption that the exchange coupling is large enough to ensure coherent rotation of all spins. However, in contrast to most theoretical work[6–11] we will not use a semiclassical approach but instead we will study the quantum problem by solving exactly the time-dependent Schrödinger equation. We investigate the quantum coherence in small single-domain ferromagnetic particles performing zero-temperature ($T=0$) dynamical calculations of the reversal of quantum single spins up to $S=75$ due to a rotation of an external magnetic field. The calculations presented in this work show that for small spins, there are only some specific resonant fields for which quantum coherence, that is, coherent tunneling back and forth between easy directions, occurs. Out of such resonant fields, quantum coherence in the spin temporal evolution is absent. These results are in qualitative agreement with those obtained for a Heisenberg Hamiltonian[12–14] and are at variance with semiclassical approximations.[10] However, as the spin becomes larger more resonant fields are shown to appear but the corresponding resonances become very small. As a consequence, coherence in time evolution becomes absent for any magnetic field. We consider the following quantum spin Hamiltonian for the single-domain ferromagnetic particle,

$$\mathscr{H}=-J_x\frac{S_x^2}{S^2}-J_y\frac{S_y^2}{S^2}-J_z\frac{S_z^2}{S^2}-2\mu_B(H_xS_x+H_zS_z), \quad (1)$$

where the spin operator $\mathbf{S}$ represents the particle's total spin, $\mathbf{M}=2\mu_B\mathbf{S}$ is the corresponding magnetic moment, μ_B the Bohr magneton, J_x, J_y, and J_z are the anisotropy constants, and H_x, H_z are components of the applied magnetic field. Although this Hamiltonian permits the study of systems with general biaxial anisotropy ($J_x\neq J_y\neq J_z\neq J_x$), in this work we will concentrate on systems with uniaxial anisotropy and choose z along the easy-magnetization axis ($J_z>J_x=J_y$). We will study the situation where initially at $t<0$, there is a magnetic field $\mathbf{H}_1$ applied along the easy axis $\mathbf{H}_1=(0,0,H_{z1})$, $H_{z1}<0$ preparing the system to be in its ground state $\Psi=|SM_S\rangle$ with $M_S=-S$. Here and in the following we denote the eigenstates of the spin operators by $|SM_S\rangle$ where

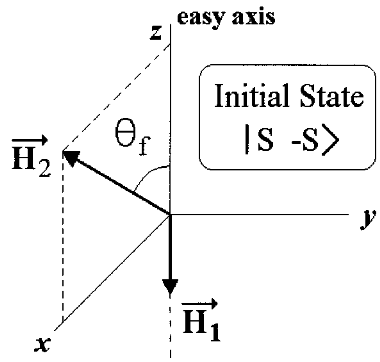

FIG. 1. Single-domain particle in the magnetic field. Initially, the field $\mathbf{H}_1$ is applied along the easy axis preparing the system in the $|S-S\rangle$ state. Subsequently, the magnetic field is rotated, forming an angle $\theta_f=45°$ with the z axis.

$S_z|SM_S\rangle=M_s|SM_S\rangle$. Subsequently, at $t\geqslant 0$ the magnetic field is rotated, forming an angle $\theta_f=45°$ with the z axis (see Fig. 1).

The time evolution of the quantum spin system is calculated from the numerically exact solution of the time-dependent Schrödinger equation (TDSE). In principle this requires the knowledge of all eigenvectors and eigenvalues of the Hamiltonian. In contrast to the Heisenberg-like models where the matrix representing the Hamiltonian (1) is of dimension $2^{2S}\times 2^{2S}$ and dedicated methods[20] are required to solve the problem for $S\geqslant 6$, for the single-spin model (1) the Hamiltonian having only a dimension $(2S+1)\times(2S+1)$ can be diagonalized by standard numerical techniques. The formal solution of the TDSE, given an initial wave function $|\Psi(t=0)\rangle$ is expressed as $|\Psi(t)\rangle=\exp(-it\mathscr{H})|\Psi(\mathrm{t}=0)\rangle$ and the temporal evolution of the αth ($\alpha=x,y,z$) component of the total spin is given by

$$\langle S_\alpha(t)\rangle=\langle\Psi(t)|S_\alpha|\Psi(t)\rangle. \tag{2}$$

The expectation value of S_α averaged over time

$$\overline{S_\alpha}=\lim_{\tau\to\infty}\frac{1}{\tau}\int_0^\tau dt\langle S_\alpha(t)\rangle, \tag{3}$$

is a useful magnitude to study the dependence of the temporal evolution of the total spin on the value of the applied magnetic field. In order to study the quantum coherence in the system evolution, we compute the time-dependent (symmetric) correlation function of the magnetization (total spin)

$$C(t)=\tfrac{1}{2}\langle\psi(0)|S_z(0)S_z(t)+S_z(t)S_z(0)|\Psi(0)\rangle. \tag{4}$$

In the absence of dissipation, coherent tunneling back and forth between easy directions leads to a sinusoidal oscillation of $C(t)$ at a frequency twice the tunneling rate. For two measurements of the magnetization separated by the time interval t, one should have[3,4]

$$\langle S_z(t+t')S_z(t')\rangle\propto\cos 2\Gamma t. \tag{5}$$

As the frequency-dependent magnetic susceptibility $\chi''(\omega)$ is essentially the Fourier transform of the correlation function (fluctuation-dissipation theorem), the former equation predicts a resonance in $\chi''(\omega)$ at $\omega_{\mathcal{R}}=2\Gamma$.

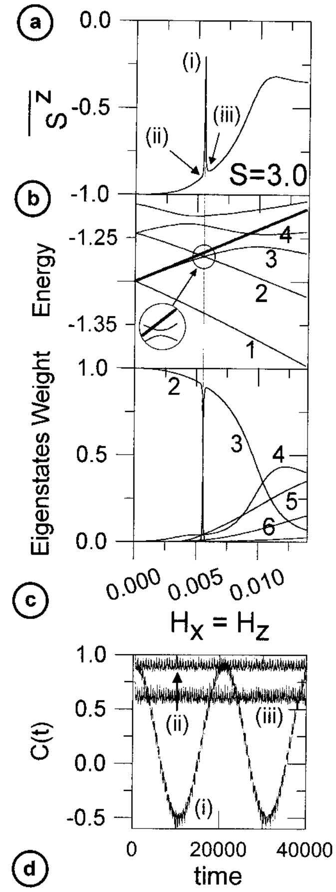

FIG. 2. (a) $\overline{S_z}$ as a function of the applied magnetic field for the model of Eq. (1) with $J_x=J_y=0.9$ and $J_z=1.0$ for spin 3. (b) System energy (thick line) and low-lying eigenenergies numbered in increasing order of energy as a function of the applied magnetic field; the inset shows the energy splitting between levels two and three at the resonant field. (c) Eigenstates weights in the decomposition of the system state, $|\Psi(t=0)\rangle=\Sigma_n c_n|E_n\rangle$, labeled by the number given to the corresponding levels. (d) Symmetrized correlation function $C(t)$ for the resonant field $H_x=H_z=0.005\,55$ (i) and two fields around it, (ii) $H_x=H_z=0.0053$ and $H_x=H_z=0.0059$. Curve (iii) has been shifted by 0.25 along the y axis in order to clarify the picture.

A well-defined resonance in $\chi''(\omega)$ has been found in experiments with superconducting quantum interference device microsusceptometers,[21] and it is tempting to associate this resonance with a MQC phenomenon. Weak dissipation is an important requirement for MQC to be observable and in these experiments[21] the coupling to the environment is thought to be weak, as indicated by the sharpness of the resonance. Many recent investigations have focused on the importance of dissipation in macroscopic quantum phenomena,[3,22] analyzing the possibility that it could prevent[23] the occurrence of MQC.

In the present work we study the reversal of a single quantum spin due to the rotation of an external magnetic field, at zero temperature and in the absence of dissipation. We show results for increasing values of the total spin S, starting from $S=3$. For $S=3$ the model with $J_x=J_y=0.9$, $J_z=1.0$, and $\theta_f=45°$ (these values are kept constant for all calculations in this work) exhibits a peculiar behavior at a

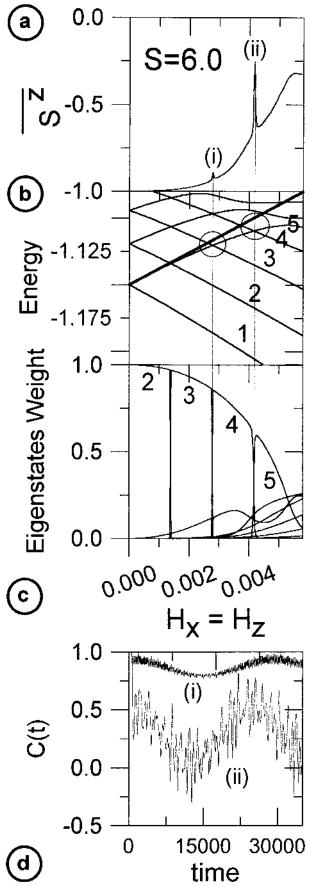

FIG. 3. Same as Fig. 2 for spin $S=6$. Plot (d) shows $C(t)$ for two resonant fields indicated by (i) $H_x=H_z=0.002\,78$ and (ii) $H_x=H_z=0.004\,17$.

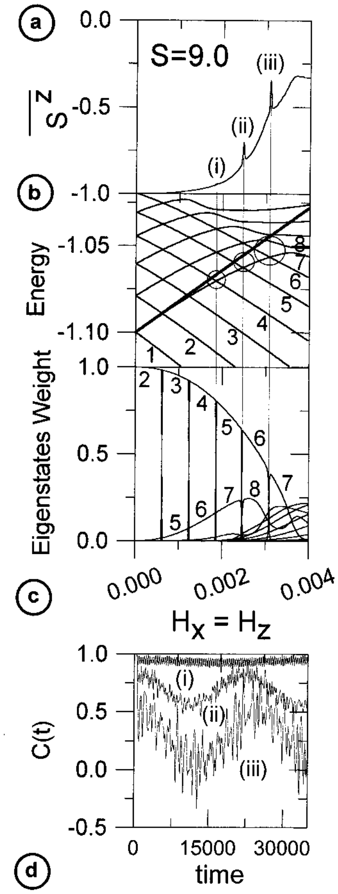

FIG. 4. Same as Fig. 2 for spin $S=9$. Plot (d) shows $C(t)$ for three magnetic fields indicated by (i) $H_x=H_z=0.001\,855\,9$, (ii) $H_x=H_z=0.002\,47$, and (iii) $H_x=H_z=0.003\,085$.

certain value of the magnetic field. The spin goes back and forth between the two easy directions with a coherent evolution. A sharp resonance at $H_x=H_z=0.005\,55$ shows up in the plot of S_z as a function of the applied magnetic field [Fig. 2(a)]. The corresponding correlation function $C(t)$ displays a pure sinusoidal oscillation [Fig. 2(d), curve (i)] as it must be when coherent quantum tunneling occurs. In contrast, for fields different from the resonant value, $C(t)$ [Fig. 2(d) curves (ii,iii)] does not exhibit a clear sinusoidal dependence but instead it looks very noisy.

Analyzing the spectrum [Fig. 2(b)], it follows that at the resonant field, the second and third eigenstates are very close in energy (see inset), both slightly below the energy $E=\langle\Psi(t=0)|H|\Psi(t=0)\rangle$ of the system. Expressing the state of the system in terms of the eigenstates ($|E_n\rangle$) of the Hamiltonian (at $t\geqslant 0$), i.e., $|\Psi(t=0)\rangle=\Sigma_n c_n|E_n\rangle$, the condition for resonant tunneling is that $E\approx E_m$, $E\approx E_{m'}$, $E_m\approx E_{m'}$, $\langle\Psi|E_m\rangle\neq 0$, and $\langle\Psi|E_{m'}\rangle\neq 0$ for some m,m'.

For the case at hand $m=2$ and $m'=3$ (see Fig. 2). At the resonant field, H_r, the weights of the second and third eigenstates are both approximately 0.5, in contrast to the situation at lower and larger fields for which the projection of the initial state onto the eigenstates is dominated by only one eigenstate. For $S=3$, these states are $|3-3\rangle$ and $|3+2\rangle$ resulting in resonant tunneling of the spin at that field. Out of the resonance, the system stays essentially in one eigenstate (the second for $H<H_r$ and the third for $H>H_r$ [Fig. 2(c)]) with a predominant $|3-3\rangle$ component so that no tunneling occurs. In a few words, the anisotropic $S=3$ model exhibits a coherent evolution between easy directions only for a specific resonant field for which the second and third eigenvalues are almost degenerate. The small splitting ΔE between the second and third eigenstates energies is related to the period P in the correlation function by $P\propto\hbar/\Delta E$.

We now consider the case $S=6$ keeping the same values of the anisotropy constants. More ''crossings'' (in fact, strictly speaking there are no crossings[24] because there always are small energy splittings that cannot be resolved on the scale of the figure) between levels below the system energy occur [Fig. 3(b)], leading to the appearance of less pronounced resonances for two resonant values of the applied magnetic field [see Fig. 3(a)]. The correlation functions $C(t)$ at such values of the field are shown in Fig. 3(d). Although not as ''pure'' as in the $S=3$ model, the sinusoidal oscillations found for $S=6$ also indicate that the spin tunnels coherently at these two values of **H**. As Figs. 3(b) and 3(c) show, for increasing **H**, first the system is essentially in the second eigenstate until there is a field for which it switches to a linear combination of the second and third eigenstates, with equal weights. For larger fields, the system is mainly the third eigenstate up to another resonant field for which the third and fourth eigenstates are equally relevant. Further on, the largest component in the system state decomposition is

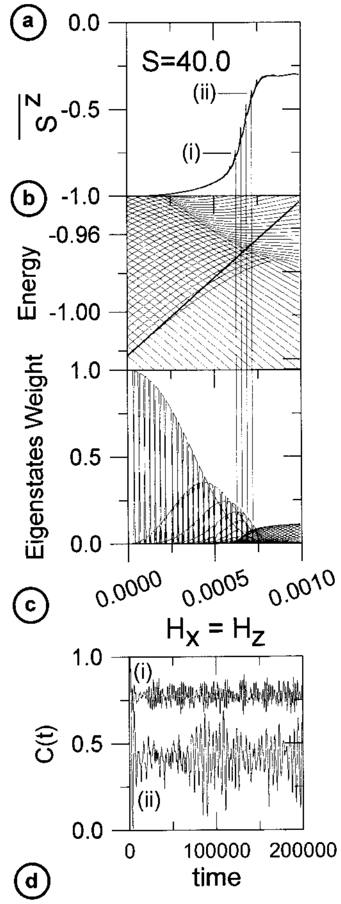

FIG. 5. Same as Fig. 2 for spin $S=40$. Plot (d) shows $C(t)$ for two magnetic fields indicated by (i) $H_x=H_z=0.000\,627$ and (ii) $H_x=H_z=0.000\,721$.

the fourth eigenstate, and so on. At the resonant field, $H_x=H_z=0.004\,17$, the most relevant components of the fourth and third eigenstates (these eigenstates have equal weights in the system state at that field) are $|6-6\rangle$, $|6-5\rangle$, $|6+2\rangle$, and $|6+4\rangle$, resulting in the tunneling phenomenon. Out of the resonance, the system state consists essentially of one eigenstate and no tunneling occurs. An equivalent behavior is shown by larger spins as for instance for $S=9$ (Fig. 4). Three, not very pronounced, resonances can be observed. They correspond to blurred sinusoidal oscillations in the correlation function $C(t)$, again indicating that there is still some coherence in the spin evolution for those specific values of the magnetic field. At these resonant fields the wave function consists, to a very good approximation, of a linear combination of consecutive eigenstates with almost the same weight [see Fig. 4(c)]. When considering larger spins (we present results for $S=40$ in Fig. 5), we find that the spectrum becomes more complicated, and many ''crossings'' between energy levels appear [Fig. 5(b)]. We find that the resonances weaken until they can almost no longer be observed [Fig. 5(a)]. If we look at the correlation function $C(t)$ at those fields corresponding to energy levels crossings [Fig. 5(d)], we do not find sinusoidal oscillations any longer, leading us to the conclusion that the evolution in time is much complicated and there is not quantum coherence at any field, in contrast to what we found for small S.

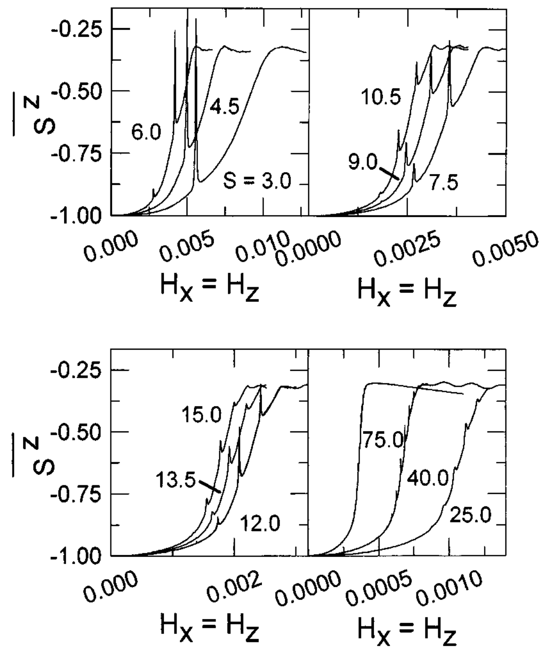

FIG. 6. $\overline{S_z}$ as a function of the applied magnetic field for the model of Eq. (1) with $J_x=J_y=0.9$ and $J_z=1.0$ for several values of the spin $3\leq S\leq 75$.

Quantum spin systems for $S\leq 75$ have been analyzed. We show in Fig. 6 the plots of $\overline{S_z}$ as a function of the magnetic field for several values of the spin S. As S increases, the field needed to overcome the energy barrier becomes smaller and more resonances appear. These resonances for large S are less pronounced until they become unobservable. The coherent evolution exhibited by the system for small S at certain resonant fields disappears with increasing S and more eigenstates become involved, complicating the problem.

In summary, we have performed exact numerical calculations of the dynamics of the reversal of single quantum spins induced by a rotation of an external magnetic field. We have shown that for small spins, there are only some specific resonant fields for which quantum coherence, that is coherent tunneling back and forth between easy directions, is present. Out of such resonant fields, quantum coherence in the spin temporal evolution is absent. These results are in concert with those obtained for a Heisenberg Hamiltonian, showing that systems of a few (up to 12) spin-$\frac{1}{2}$ particles exhibit coherent tunneling of the magnetization only for some specific resonant values of the applied magnetic field,[12,13] and are at variance with semiclassical calculations that fail to predict the phenomenon of resonant tunneling.[10] We have also shown that, as the spin becomes larger, more resonant fields appear, leading to very small resonances and the absence of coherence in the temporal evolution of the quantum spin at any magnetic field. Finally, we want to add that systems with a general biaxial anisotropy ($J_x\neq J_y\neq J_z\neq J_x$ in our model) not only exhibit resonances similar to the ones discussed in this paper, but also show a resonance at zero field. This is due to the presence of an easy direction and an easy plane, leading to an energy splitting between the first two levels at zero field. This splitting appears only for integer spins and is absent for half-integer spins, suggesting the existence of spin-parity effects.[14,25,26] As S increases, large anisotropies are needed for the zero-field resonance to be observable. A more detailed study of this model will be presented elsewhere.

The resonant tunneling results presented here and in our previous work[12–14] are in qualitative agreement with recent experimental data,[27,28] showing that minima in the relaxation time appear at discrete values of the applied magnetic field corresponding to level crossings. We have shown that, exactly for such resonant magnetic fields for which levels cross, coherent quantum tunneling occurs.

Note added in proof. Recently we have carried out quantum dynamical calculations for a single spin model like (1) with $S=10$, biaxial anisotropy ($J_z > J_x > J_y$), and a time-dependent magnetic field along the easy-axis (z) with an angle deviation of 1° which varies from a negative (positive) value to a positive (negative) value in a certain time. We find that the magnetization goes from the initial negative (positive) value to the final positive (negative) value showing several steps at the resonant magnetic fields as those found in recent experiments.[27,28] These steps correspond to resonant tunneling of the magnetization and are obtained in the absence of dissipation. Therefore, magnetization curves with a staircase structure as those reported in Ref. 27 and 28 can be obtained at $T=0$ and without assuming any dissipation mechanism. This indicates that the effect of dissipation is small and temperature is not needed to understand the mechanism of resonant quantum tunneling which is theoretically possible at $T=0$. A detailed study now in progress will be presented elsewhere.

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