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

D. García-Pablos and N. García
Laboratorio de Física de Sistemas Pequeños C.S.I.C.-U.A.M., Facultad de Ciencias, S.I.D.I., C-IX, Universidad Autónoma de Madrid, Cantoblanco, 28049 Madrid, Spain

P. A. Serena
Dept. Física de la Materia Condensada, Facultad de Ciencias, C-III, Universidad Autónoma de Madrid, Cantoblanco, 28049 Madrid, Spain

H. De Raedt
Institute for Theoretical Physics, University of Groningen, Nijenborgh 4, 9747 AG Groningen, The Netherlands

We investigate the reversal of magnetization and the coherence of tunneling when an external magnetic field is rotated instantaneously in systems of a few (N) spin 1/2 particles described by an anisotropic Heisenberg Hamiltonian at $T=0$. The temporal evolution is calculated by a numerically exact solution of the time-dependent Schrödinger equation, and the mean value in time of each spin component is computed as a function of the magnetic field. The correlation function and the spectrum are analyzed in terms of the macroscopic quantum coherence. Our calculations demonstrate that this model for small ($N<11$) magnetic particles exhibit collective tunneling of the magnetization only for some specific resonant values of the applied magnetic field, at variance with the Stoner–Wohlfarth model. [S0021-8979(96)76708-5]

The ability to miniaturize magnetic materials and study the magnetic properties of a single isolated particle has revealed classical and quantum phenomena[1–3] that questions the present understanding of the fundamentals of magnetism. The phenomenon of macroscopic quantum tunneling (MQT)[4,5] has received a lot of attention and 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,[1,6–8] these minima correspond to the two states of opposite magnetization. When there is a repeated coherent tunneling back and forth between the two wells we have a case of macroscopic quantum coherence (MQC). The Stone–Wohlfarth (SW) model,[9] due to 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 some details in experimental systems.[3] The quantum mechanical effects have been studied theoretically by the quantization within a path integral formalism of the classical micromagnetic theory of magnetic dynamics.[6–8] Chudnovsky and Gunther[6] showed that in addition to superconducting devices, single-domain magnetic particles represent a rich field for MQT study. In the semiclassical approximation, a uniform and coherent rotation of all spins is imposed, that is, spins are considered to behave dynamically as a single quantum spin. This is known as single spin model (SSM). In experiments with superconductivity quantum interference device microsusceptometers,[10] a well-defined resonance in the frequency-dependent magnetic susceptibility $\chi''(\omega)$ has been found and it is tempting to be associated with a MQC phenomenon although there is some controversy on this interpretation.[11] On the other hand, the process of magnetization reversal in single particles[12,13] is also being studied nowadays with much interest. Recent experimental[13] and theoretical[14] works have studied the mechanism of the nonuniform reversal of the magnetization in this kind of particles.

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 in the reversal of magnetization. The results we have found calculating the exact quantum evolution of the spins show a qualitatively different landscape to what has been explained above: There is essentially a sharp resonance corresponding to coherent quantum tunneling of the magnetization but only for a particular magnetic field, whereas for lower and larger fields this phenomenon does not appear. Notice that in general, the quantum evolution of the spins is noncoherent. This resonant coherent quantum tunneling occurs at fields much lower than the values corresponding to the vanishing of the barrier in the Stoner–Wohlfarth mode.[9] The former model is at variance with the exact calculations presented in this paper.

We need to introduce the two-time correlation function of the magnetization,[4] 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, it has been calculated the symmetrized correlation function $C(t)$ defined as $C(t)=1/2\langle\Psi(0)|S^z(0)\times S^z(t)+S^z(t)S^z(0)|\Psi(0)\rangle$. With negligible dissipation present, coherent tunneling back and forth between the two states (magnetization up and down) leads to a sinusoidal oscillation of $C(t)$ at a frequency twice the off-diagonal matrix element. For two measurements of the magnetization separated by the time interval t, one should have $\langle S(t')S(t'+t)\rangle=S_0^2\cos(2\Gamma t)$. 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 $\chi''(\omega)$.

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

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

where $\sigma_i^\alpha(\alpha=x,y,z)$ 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. We have limited ourselves to systems with uniaxial anisotropy Δ along the z direction $[J_x=J_y<J_z=J$, $\Delta=(J_z-J_x)/J]$, containing N particles $(2<N<11)$ with different geometrical forms and to instantaneous rotations of the magnetic field of several angles. The range of the parameters is $0.01J\leqslant\Delta\leqslant0.1J$ for the anisotropy and $0\leqslant H\leqslant0.2J$ for the magnetic field. The temporal evolution of the system is calculated by a numerically exact solution of the time-dependent Schrödinger equation (TDSE).[15] This requires the computation of all eigenvalues and eigenvectors of the Hamiltonian. For larger systems $(N>8)$ we use Suzuki's fourth-order fractal product formula[15–17] to solve TDSE.

In our particular model, 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 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 (notice that nothing happens in the exact propagation for $\theta_f=0°$). We have studied the dependence of $\bar{S}_i^z$, mean value in time of $\langle S_i^z(t)\rangle$ for each different spin i, on the size of the second magnetic field $\mathbf{H}_2$:

$$\bar{S}_i^z=\lim_{\tau\to\infty}\left[\tau^{-1}\int_0^\tau dt\langle S_i^z(t)\rangle\right]. \quad (2)$$

The correlation function $C(t)$ for the second Hamiltonian is also analyzed, as well as the eigenvalues, eigenstates and system energy for each magnetic field considered. 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) tunneling region when there is a barrier between the two wells and (b) non-tunneling 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 result obtained is the following: clusters with $N\geqslant5$, and with different geometrical forms (chain, ring, and others) present a pronounced resonance in the curve of $\bar{S}_i^z$ in terms of $H_{x2}=H_{z2}$ for a specific magnetic field H_r that clearly falls in the tunneling region (a). In Fig. 1(a) we show this result for an open linear chain of seven spins. We have found that these resonances correspond to pure sinusoidal oscillations in the correlation function $C(t)$ as it must occur when there is MQC. However, for points around these resonances $C(t)$ does not present this sinusoidal shape at all. In

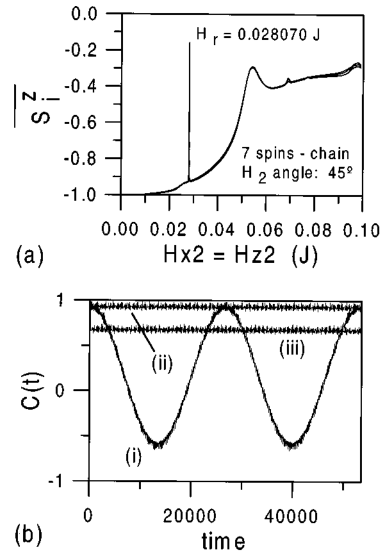

FIG. 1. (a) $\bar{S}_i^z$ for each different spin i as a function of the size of the second magnetic field for a linear chain of seven spins, with $\Delta=0.1$ and $\theta_f=45°$, and (b) symmetrized correlation function for the resonant field $H_r=0.028\,070J$ (i) and two fields around it: (ii) $H=0.0276J$ and (iii) $H=0.0285J$-(iii) curve has been shifted 0.25 in the y axis in order to clarify the picture.

Fig. 1(b) we present $C(t)$ at the resonant field [Fig. 1(b)-(i)] and at two fields around it [Fig. 1(b)-(ii), (iii)] for the same linear cluster. Clusters with $N<5$ do not show this behavior and the reason can be explained in terms of the spectrum and the curve of H_r vs N. Clusters with eight and more spins present more than one peak although only one is really sharp.

In order to understand why a particular magnetic field provokes the resonant MQC we have studied the system spectrum calculating its eigenstates energies for each magnetic field applied. For a complete discussion of the spectrum analysis see Ref. 18. This point of view has also been considered for the study of a SSM.[19] To give a slight idea, the specific field that produces the resonance makes practically equal the energies of the second and third eigenstates of the system, which correspond essentially to all spins in one direction and in the opposite, respectively, and which are the only relevant eigenstates in the system state at that field. The system energy for that field is slightly above these two levels of energy. This fact permits a resonant tunneling of the magnetization for a determined field in each case. 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 $T=2\pi\hbar\Delta E^{-1}$. The values of T and ΔE fit very well to this formula.

Other anisotropy values and other directions have been studied and we have also found sharp resonances corresponding to sinusoidal correlation functions.[17] 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, different values of the anisotropy and for all the directions of the magnetic field $\mathbf{H}_2$ studied.

In Fig. 2 we show the dependence of the resonant fields H_r and the field H_b that makes the barrier disappear in the

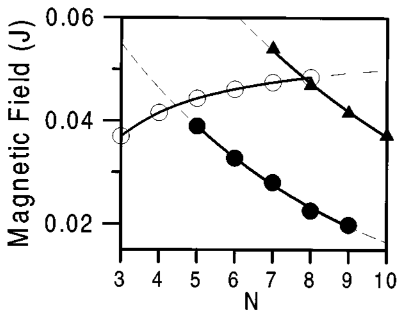

FIG. 2. Dependence of the resonant field H_r (solid circles) and the field needed to vanish the activation barrier H_b (open circles) on the number of spins for a fixed geometrical configuration. The solid triangular symbols correspond to the second peak in the S_i^z curve for clusters with more than seven spins.

single spin model (SSM) on the number of spins ($N<11$) for the same geometrical configuration (chain). As we can see in Fig. 2 $H_r<H_b$ for systems with $N\geqslant 5$. 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.[6] The tendency shown by the two curves can explain why clusters with $N<5$ 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. 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 more than seven spins. The field corresponding to the second peak in eight spins cluster is below H_b whereas in seven spins cluster the second peak field is above H_b (see Fig. 2). The sharpness of the peaks is related to the separation between the levels involved. When the repulsion between the levels involved becomes larger the peak gets less important. As N increases and new resonances appear in the tunneling region, those corresponding to very low fields become smaller.

In conclusion, we have studied the reversal of magnetization and the coherence of tunneling when an external magnetic field is rotated instantaneously in systems for a few spin 1/2 particles described by an anisotropic Heisenberg Hamiltonian at $T=0$. Our calculations demonstrate that the model studied for systems with $4<N<11$, for any geometrical configuration and for different anisotropy values exhibits collective tunneling of the magnetization only for some specific resonant values of the magnetic field, at variance with the Stoner–Wohlfarth model that predicts coherent rotation at all fields.

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