Theory of quantum tunneling of the magnetization in magnetic particles

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We study the response of the magnetization to a time-dependent applied magnetic field $H(t)$ in a model for a uniaxial magnet. It is shown that a staircase structure in the magnetization curve results from Landau-Zener tunneling between different pairs of nearly-degenerate energy levels. This mechanism might be relevant to the analysis of the hysteresis of nanoscale magnets at low temperatures, allowing one to extract the energy splittings from the hysteresis curve. We investigate the dependence of the staircase structure on the sweep rate $dH(t)/dt$, and point out some universal features of the staircase in uniaxial magnets. We also study the effect of a step-wise (instead of continuous) increase in the field, and show that the size of the steps depends sensitively on the procedure used to change the applied field. [S0163-1829(97)06042-6]

I. INTRODUCTION

In magnetic particles with uniaxial anisotropy the dynamics of the metastable magnetization due to thermal and quantum fluctuations has attracted interest recently. At sufficiently low temperatures quantum dynamical effects are important and the concept of quantum tunneling of the magnetization (QTM) (Ref. 1) has become a topic of much interest. Theoretical analysis of the response of the magnetization of a quantum spin system at zero temperature to a sudden reversal of the applied magnetic field shows that QTM can only occur for particular values of the applied field, corresponding to the conditions for resonant tunneling.[2–4] On the other hand, quantum dynamical calculations of the change of the magnetization due to a slowly reversing applied field have shown that nonadiabatic transitions between energy levels govern the magnetization dynamics and that the tunneling of the magnetization at $H=0$ can be modeled by the Landau-Zener mechanism.[5–8]

In view of the generic character of the findings of Refs. 2–6, it appears logical to search for a unified description of the quantum dynamics of the magnetization at zero temperature. In this paper we show that the Landau-Zener tunneling picture correctly describes the dependence of the magnetization of a uniaxial magnet on a slowly changing applied field. This in turn suggests the possibility of extracting information on the energy-level scheme from the dependence of the magnetization on the sweep rate of the applied field.

Our results may be of direct relevance to recent experiments on high-spin ($S=10$) molecules (Mn_{12}-Ac) in which steps in the magnetization as a function of the time-dependent magnetic field have been observed.[9–12] These steps are characteristic of the resonant tunneling of the magnetization.[4] Furthermore the resonant tunneling may be thermally assisted.[9–14] As we will show, several features observed in experiment such as the dependence of the steps on the sweep rate of the applied field and the absence of steps in half of the hysterisis curve, follow quite naturally from the Landau-Zener picture.

According to the adiabatic theorem a slowly changing external perturbation will keep a system in the eigenstate it started from unless this eigenstate comes close to another eigenstate. Then the adiabatic approximation might break down, allowing the system to escape, via the Landau-Zener tunneling mechanism,[15–17] from its current eigenstate and ''tunnel'' to the other, nearby, eigenstate.

To illustrate the application of the above concepts let us consider the simplest case, a single spin-$\frac{1}{2}$ system described by the Hamiltonian

$$\mathcal{H}=-\Gamma\sigma_x-ct\sigma_z, \tag{1}$$

where σ_i^x and σ_i^z denote the x, respectively, z component of the Pauli-spin matrices, Γ sets the scale of the energy-level splitting, and c is the sweep rate of the applied field, i.e., $H(t)=-ct$. For large negative times t and $|H(t)|\gg|\Gamma|$, the ground-state consists primarily of the spin-down state. As t goes to infinity the probability for the system to end up in the spin-up state (i.e., the probability to change its magnetization) is given by[15]

$$p=1-\exp\left(-\frac{\pi\Gamma^2}{c}\right). \tag{2}$$

The tunneling occurs for $H\approx 0$ and the magnetization exhibits a step at $H=0$ proportional to p. From Eq. (2) it follows that the step in the magnetization at $H(t)=0$ not only depends on the energy splitting $\Delta E = 2\Gamma$ but also on the rate of change c of the field, as observed in experiment.[12]

It is useful to interpret Eq. (2) in terms of scattering events. The probability for scattering from the ground state to the excited state is $1-p$. In the adiabatic limit $(c \to 0)$ there is no scattering: $p=1$. For very fast sweeps $c \to \infty$ the scattering is complete, i.e., $p=0$.

In the next section we demonstrate that with some minor modifications the above picture correctly describes, on a quantitative level, the steps in the magnetization, not only at $H \approx 0$ but for all H at which the energy levels are nearly degenerate.

II. MODEL

The Ising model in a transverse field is perhaps the simplest microscopic model to describe a uniaxial magnet. Its Hamiltonian reads

$$\mathcal{H} = -J \sum_{i,j \in \mathcal{C}} \sigma_i^z \sigma_j^z - \Gamma \sum_i \sigma_i^x - H(t) \sum_i \sigma_i^z, \qquad (3)$$

where J and Γ are the exchange interaction and transverse field, respectively, and $H(t)$ represents the time-dependent magnetic field. The set $\mathcal{C}$ defines the interactions between pairs of spins in the cluster. As the qualitative features of the results do not depend on the particular choice of $\mathcal{C}$ we will, in this paper, only present results for rings of L spins. Consequently, we set $\mathcal{C} = \{(1,2),(2,3),\ldots,(L-1,L),(L,1)\}$.

The time dependence of the magnetization at zero temperature is obtained from the solution of the time-dependent Schrödinger equation (TDSE)

$$i\hbar \frac{\partial}{\partial t} |\Psi(t)\rangle = \mathcal{H} |\Psi(t)\rangle, \qquad (4)$$

where $|\Psi(t)\rangle$ denotes the wave function of the spin system at time t. We solve the TDSE (4) using three different algorithms: Exact diagonalization, a fourth-order fractal decomposition[18] of $e^{-it\mathcal{H}}$ using $\mathcal{H} = \mathcal{H}_z + \mathcal{H}_x$ with $\mathcal{H}_z = -J\Sigma_{i,j\in\mathcal{C}} \sigma_i^z\sigma_j^z - H(t)\Sigma_i \sigma_i^z$ and $\mathcal{H}_x = -\Gamma\Sigma_i \sigma_i^x$, and another fourth-order fractal decomposition[18] of $e^{-it\mathcal{H}}$ using $\mathcal{H} = \Sigma_{i,j\in\mathcal{C}} \mathcal{H}_{i,j}$ with $\mathcal{H}_{i,j} = -J\sigma_i^z\sigma_j^z - \Gamma(\sigma_i^x + \sigma_j^x)/2 - H(t)(\sigma_i^z + \sigma_j^z)/2$. For the present problem product-formula-based algorithms are more suited for solving the TDSE (4) than the standard exact diagonalization technique. This is because $\mathcal{H}$ changes with time through the time-dependent applied field, requiring the exact diagonalization of $\mathcal{H}$ for each value of the time t. Solving Eq. (4) by one of the product-formula algorithms is more efficient (in terms of CPU time) than doing the same calculation through exact diagonalization. For many simple cases we have checked that the results of these three different algorithms agree, eliminating the possibility that the observed phenomena are due to spurious effects in the numerics.

In practice we solve the TDSE (4) as follows. First we set the applied field to its minimum value $H(t_0) = -H_0$ and put the system in the corresponding ground state, i.e., $|\Psi(t_0)\rangle = |\phi_0\rangle$ where $\mathcal{H}|\phi_0\rangle = E_0(-H_0)|\phi_0\rangle$. Our convention is such that for large H_0, $|\phi\rangle$ is very close to the state with all spins down. The time evolution of the wave function is obtained from $|\Psi(t+\tau)\rangle = e^{-i\tau\mathcal{H}}|\Psi(t)\rangle$, where τ denotes the (small) time step used to integrate the TDSE.

During the integration of the TDSE, the applied field changes with time, from $-H_0$ to $+H_0$. It is convenient to introduce two parameters to characterize this change: The field step $\Delta H = 2H_0/m_f$ and the sweep rate $c = \Delta H/\tau m$. The former controls, through m_f, the number of times the field changes as it increases from $-H_0$ to $+H_0$. The latter fixes the amount of time τm during which the system "feels" a constant applied field. If ΔH is sufficiently small, we recover the Landau-Zener case in which $H(t)$ increases linearly with time. The energy and magnetization at a particular value of the field are given by

$$E(t) = E(H) = E[H(t)] = \langle \Psi(t)|\mathcal{H}|\Psi(t)\rangle, \qquad (5a)$$

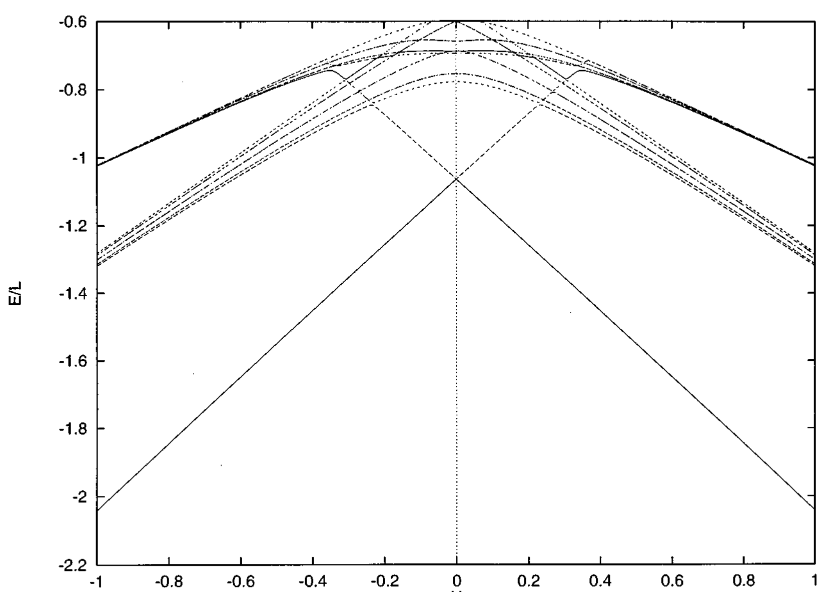

FIG. 1. Energy spectrum of a ring of $L=8$ sites for $J=1$ and $\Gamma=0.5$ plotted as a function of the applied magnetic field H.

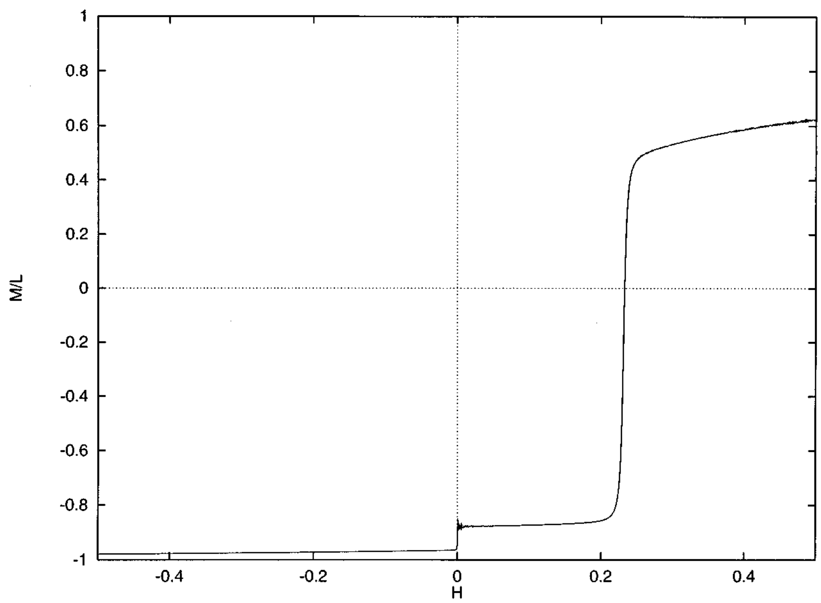

FIG. 2. Magnetization as a function of increasing applied field, for $J=1$ and $\Gamma=0.5$. The sweep rate $c=4.8\times10^{-6}$ and the step in the applied field $\Delta H=1.5\times10^{-5}$.

and

$$M(t)=M(H)=M[H(t)]=\langle\Psi(t)|\sum_i \sigma_i^z|\Psi(t)\rangle, \quad (5b)$$

respectively. Estimates of the Landau-Zener transition probabilities are obtained by projecting the wave function onto the eigenstates, i.e.,

$$\widetilde{p}_i=\lim_{t\to\infty}|\langle\phi_i(t)|\Psi(t)\rangle|^2, \quad (6)$$

where $\phi_i(t)=\phi_i[H(t)]$, $i=0,1,\ldots,$ denotes an eigenstate of the Hamiltonian for a fixed value of the applied field $H(t)$.

III. RESULTS

Previous work has demonstrated the generic character of QTM.[2–6] Therefore we will present results for representative cases only. In our numerical work we set $\hbar=1$ and $J=1$ and express all physical quantities in dimensionless units. In Fig. 1 we show the 16 lowest energy levels of the Ising model in a transverse field as a function of the applied field H, for a ring of 8 sites and $\Gamma=0.5$. From Fig. 1 it is clear that there are at least five instances at which the energy levels are nearly degenerate. Closer inspection shows that for the model parameters of Fig. 1, Landau-Zener transition can occur at $H=0$, $H=\pm0.23$, $H=\pm0.31$, etc. In particular at $H=0$ a transition occurs involving the ground state $(i=0)$ and the first excited state $(i=1)$ whereas the transition at $H=0.23$ involves levels $i=1$ and $i=2$. Note that not all level crossings qualify as candidates for Landau-Zener transition. For example, in the interval $0.23<H<0.31$ the scattered state ($i=2$ at $H=0.23$) crosses several levels but no Landau-Zener transitions take place until the field reaches the value $H=0.31$ where a transition involving states $i=8$ and $i=9$ becomes possible.

In all the models that we have studied the asymmetry of the energy-level splitting as a function of the field is small so that formula (2) applies.

In Fig. 2 we depict the results for $M(H)$ obtained from a simulation of the system with the energy spectrum shown in Fig. 1. The field step ΔH is very small so that the Landau-Zener theory applies. As the field increases from its minimum value ($H_0=-0.5$ in this case), the magnetization increases very little until $H(t)\approx0$ where it exhibits a step. Further increasing the field leads to a second step at $H(t)\approx0.23$, after which the magnetization $M\approx0.6$. In this case we do not observe more than two steps, for reasons that will become clear soon.

According to the Landau-Zener transition picture, the probability for staying in the same eigenstate is given by[5,6]

$$p_i=1-\exp\left(-\frac{\pi(\Delta E_i)^2}{4|M_i|c}\right); \quad i=0,1,\ldots, \quad (7a)$$

TABLE I. Comparison between the transition probabilities obtained from the Landau-Zener tunneling model and those obtained from simulation. The parameters are the same as for Fig. 2. The sweep rate $c=4.8\times10^{-6}$.

Step n	Eigenstate i	Field H_i	Energy splitting ΔE_i	Theory p_i	Eq. (8) $\hat{p}_i$	Simulation $\widetilde{p}_i$
1	0	0	1.456×10^{-3}	0.0424	0.0424	0.044
2	1	0.231	5.424×10^{-2}	1.0000	0.9576	0.96
3	8	0.307	3.672×10^{-4}	0.0000	0.0000	0.00

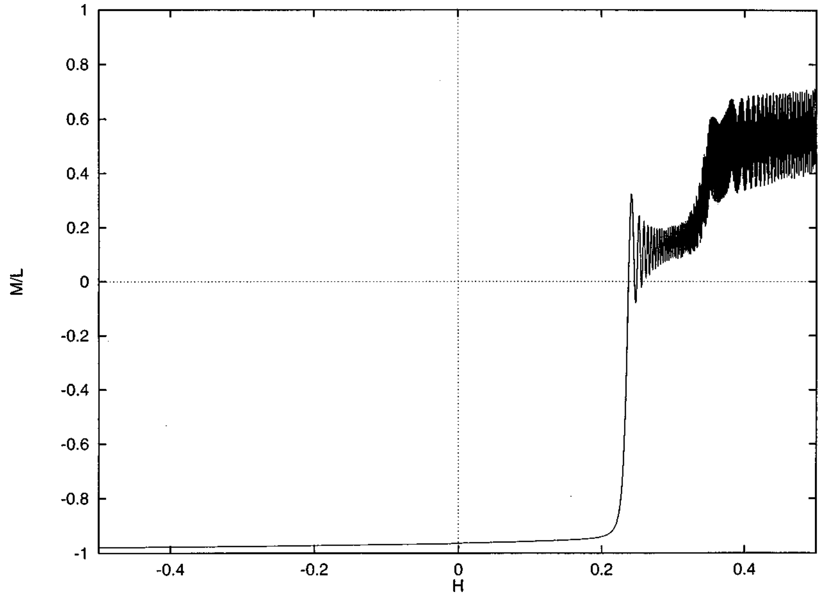

FIG. 3. Magnetization as a function of increasing applied field, for $J=1$ and $\Gamma=0.5$. The sweep rate $c=3.1\times10^{-4}$ and the step in the applied field $\Delta H=1.2\times10^{-4}$.

where

$$\Delta E_i=E_{i+1}-E_i, \tag{7b}$$

and M_i is the magnetization of the ith state. For practical purposes we replace the magnetization per site M_i by its asymptotic value for large H, i.e., $M_0\approx L$ and $M_1\approx L-2$, etc.[5,6] This approximation is not essential but simplifies the reasoning considerably. The probability $\hat{p}_i$ for the system to end up in the ith eigenstate is given by

$$\hat{p}_i=\begin{cases} p_0; & i=0 \\ (1-p_0)\ldots(1-p_{i-1})p_i; & i>0, \end{cases} \tag{8}$$

where it has been assumed that the energy levels are numbered in ascending order. In the simulation we can compute the probabilities $\tilde{p}_i$ directly [see Eq. (6)]. Therefore a comparison of $\tilde{p}_i$ and Eq. (8) should tell us whether or not the response of the magnetization can be understood in terms of successive Landau-Zener transitions.

In Table I we compare the probabilities computed from Eq. (8) with those obtained from the simulation. The agreement is excellent. In this case we do not observe more than two steps because $p_0+(1-p_0)p_1\approx1$, i.e., the probability for a third step to occur is very small (see Table I). The interpretation in terms of Landau-Zener transitions also explains why there are no steps for $H\approx-0.23$ or $H\approx-0.31$ in this case. As long as the field increases from its minimum value but remains negative, the ground state does not come close to one of the excited states (see Fig. 1) so that the scattering probability is extremely small. However, at $H=0$ the energy splitting ΔE_0 is small (see Table I) and the tunneling probability can become appreciable if the sweep rate c is low. The state of the system is, to a good approximation, a linear combination of the ground states for $H<0$ and $H>0$. Further increase of the field then fixes the weight of the ground state in this linear combination until the next resonant field is approached. The magnetization is given by

$$M\approx-L;\quad H<0, \tag{9a}$$

and

$$M\approx Lp_0+(-L)(1-p_0);\quad 0<H, \tag{9b}$$

hence the step of the magnetization at $H=0$ is estimated to be

$$\Delta M_0\approx2p_0\approx0.09, \tag{10}$$

in good agreement with the simulation data.

The same reasoning applies to the second step: Before H reaches its value for the second step ($H\approx0.23$), the state of the system is, to a good approximation, a linear combina-

TABLE II. Comparison between the transition probabilities obtained from the Landau-Zener tunneling model and those obtained from simulation. The parameters are the same as for Fig. 3. The sweep rate $c=3.1\times10^{-4}$. The dashes indicate that the state of the system adiabatically follows the 9th eigenstate.

Step n	Eigenstate i	Field H_i	Energy splitting $\Delta E_i/L$	Theory p_i	Eq. (8) $\hat{p}_i$	Simulation $\tilde{p}_i$
1	0	0	1.456×10^{-3}	0.0007	0.0007	0.0007
2	1	0.231	5.424×10^{-2}	0.7113	0.7108	0.74
3	8	0.307	3.672×10^{-4}	0	0	0
-	9	-	-	1.0000	0.2885	0.26

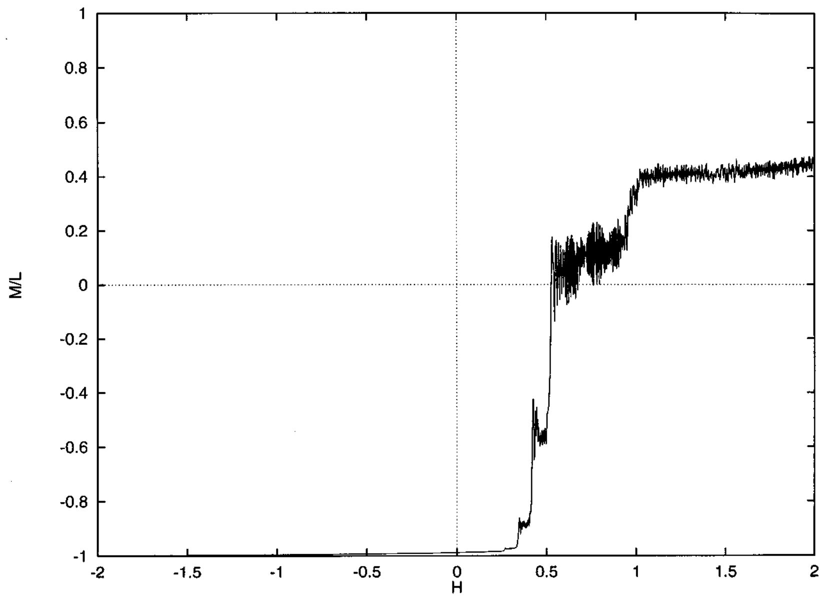

FIG. 4. Magnetization as a function of increasing applied field, for $J=1$ and $\Gamma=0.3$. The sweep rate $c=3.1\times10^{-4}$ and the step in the applied field $\Delta H=1.2\times10^{-4}$.

tion of the ground state and the first excited state with most weight, namely, $1-p_0$, in the latter. At the second transition ($H\approx0.23$) the probability to tunnel from the first excited state to the second is $p_1\approx0.956$, i.e., relatively large (see Table I). Therefore

$$M\approx Lp_0+(-L)(1-p_0);\quad 0<H<0.23, \tag{11a}$$

and

$$M\approx Lp_0+(-L)(1-p_0)(1-p_1)+(L-2)(1-p_0)p_1;\quad 0.23<H<0.31, \tag{11b}$$

yielding for the step in the magnetization

$$\frac{\Delta M_1}{L}=2p_1(1-p_0)\left(1-\frac{1}{L}\right)\approx1.7, \tag{12}$$

again in good agreement with the simulation. The difference between the simulation result for ΔM_1 and Eq. (12) can be traced back to the approximation made in replacing the magnetization of the nearly-degenerate states by its asymptotic value.

Note that in deriving Eq. (12) we assumed that the renormalization of the magnetization is small. In general this is not necessarily the case but in the case at hand it is a reasonable approximation. From this example we conclude that the response of the magnetization to slowly changing field can, to a good approximation, be described in terms of successive Landau-Zener transitions events.

Further support for this point of view comes from considering more examples. In Fig. 3 we show results for a case where the sweep rate is larger than in the previous case. According to the Landau-Zener theory increasing the sweep rate should yield larger scattering probabilities $1-p_i$. This is confirmed by the results reported in Fig. 3 and Table II. From Table II it is clear that the tunneling probability for $H=0$ is much smaller than for the parameters used in Fig. 2 and Table I. We find that $p_0\approx0.0007$, much smaller than in the previous case, explaining why the step in the magnetization is hardly visible. The next step occurs at $H=0.23$ and a third step appears at $H=0.31$. After the third Landau-Zener transition the state of the system adiabatically follows the 9th eigenstate, hence all the remaining weight (≈0.26) is carried by this state. From Table II it is clear that also in this case there is excellent agreement with the Landau-Zener based theory.

In Fig. 4 we present results for the same model except that now $\Gamma=0.3$ instead of $\Gamma=0.5$. The energy splitting at H

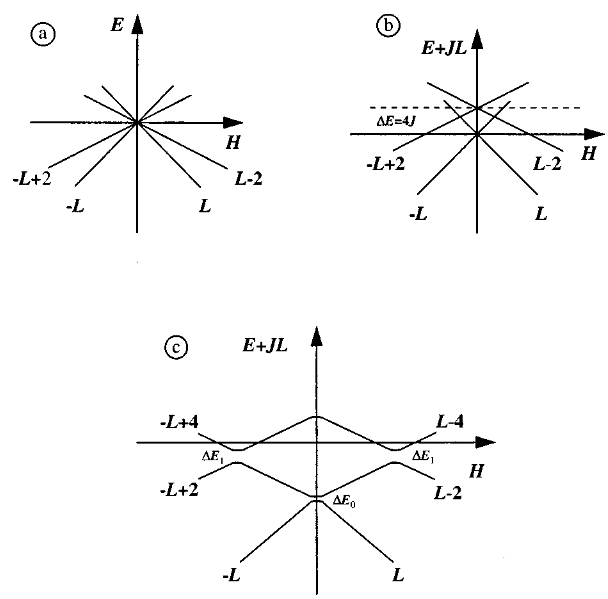

FIG. 5. Schematic diagram of the few lowest energy levels of (a) noninteracting spins in an applied field, (b) a uniaxial Ising spin system in an applied field, and (c) a generic uniaxial quantum spin system. In (a), (b), and (c) the energy levels are labeled by their magnetization in the noninteracting case.

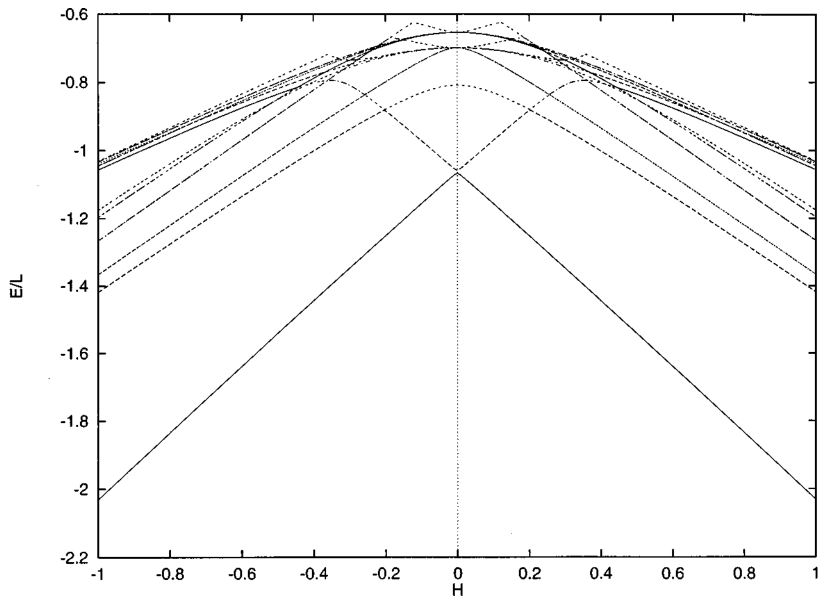

FIG. 6. Energy spectrum of XZ model (13) for a ring of $L=8$ sites for $J_x=0.5$, and $J_z=1$, plotted as a function of the applied magnetic field H.

$=0$ is smaller than in the previous cases so that the step at $H=0$ is too small to be seen. Four steps are clearly visible in this case.

IV. UNIVERSAL BEHAVIOR

In this section we discuss the universal features of the magnetization dynamics. The main features of response of the magnetization to a time-dependent field are intimately related to the structure of the energy-level scheme. In Fig. 5 we show schematic diagrams of the lowest energy levels for, respectively, noninteracting spins [Fig. 5(a)], the Ising model [Fig. 5(b)], and a typical uniaxial quantum spin system [Fig. 5(c)].

For noninteracting spins the energy is a bilinear function of the magnetization and the applied field. All levels cross at $H=0$ [see Fig. 5(a)]. The magnetization is conserved and hence there are no transitions between states of different magnetization. The presence of a uniaxial exchange interaction J changes the energy-level diagram: Flipping one spin not only changes the magnetization but also costs an additional amount of energy $\Delta E=4J$ for the rings studied in this paper. At $H=0$ only levels with opposite magnetization cross [see Fig. 5(b)]. In this case the magnetization is still conserved. Adding quantum fluctuations results in the diagram shown in Fig. 5(c). The magnetization is no longer conserved and energy gaps ΔE_i appear at $H=0$, $H_{\pm 1}$, ...

The magnetization dynamics can be understood in terms of successive Landau-Zener transitions. In general, $\Delta E_0 \ll \Delta E_{\pm 1}$, implying that for a wide range of sweep rates the

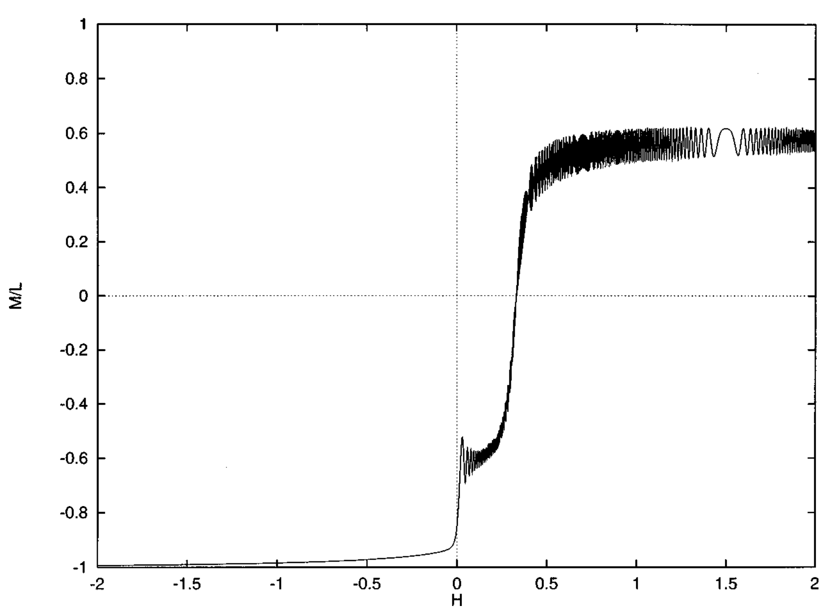

FIG. 7. Magnetization as a function of increasing applied field, for $J_x=0.5$, and $J_z=1$. The sweep rate $c=3.1\times 10^{-3}$ and the step in the applied field $\Delta H=1.2\times 10^{-4}$.

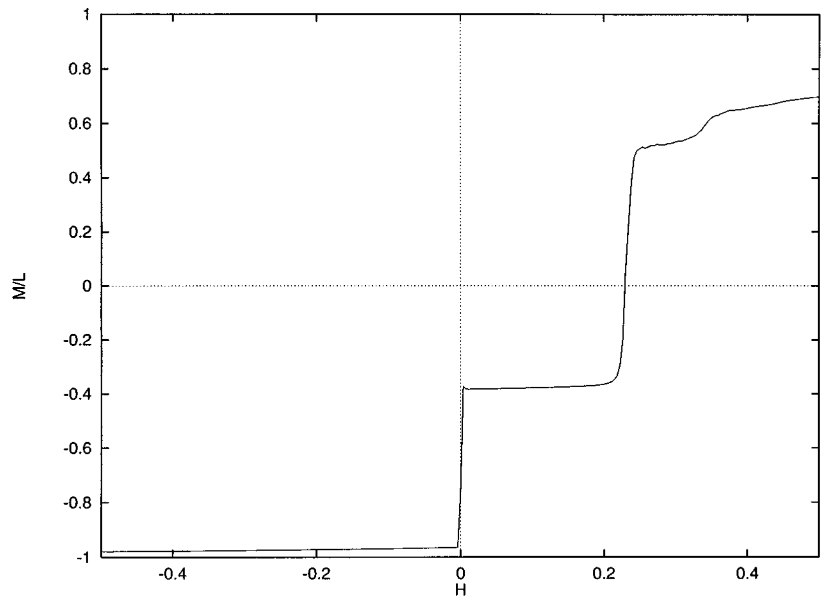

FIG. 8. Magnetization as a function of increasing applied field, for $J=1$ and $\Gamma=0.5$. The sweep rate $c=4.8\times10^{-6}$ and the step in the applied field $\Delta H=3.9\times10^{-3}$.

magnetization will exhibit a small step at $H=0$ and a much larger step at $H=H_1$. This is the most common case. In the extreme case where the sweep rate $c\rightarrow0$ there is no scattering at $H=0$, the magnetization will reverse on crossing $H=0$ and only one step (at $H=0$) is observed. If, on the other hand, c is too large (sudden limit), the magnetization will show no steps at all.

As the schematic energy-level diagram [see Fig. 5(c)] contains the salient features of the low-energy spectrum of uniaxial magnets, we expect the sweep-rate dependence of the magnetization found in this paper to be a universal property of low-temperature magnetization dynamics in nanomagnets, independent of the origin of the quantum fluctuations.

As another illustration of the universal character we show, in Figs. 6 and 7, the energy-level scheme, respectively, magnetization curve for the XZ model described by the Hamiltonian

$$\mathcal{H}=-J_x\sum_{i,j\in\mathcal{C}}\sigma_i^x\sigma_j^x-J_z\sum_{i,j\in\mathcal{C}}\sigma_i^z\sigma_j^z-H(t)\sum_i\sigma_i^z, \quad (13)$$

for $J_x=0.5$ and $J_z=1$. Although the spectrum of this model clearly differs from that of the Ising model in a transverse field, the salient features of it are the same as for the Ising model in a transverse field and consequently the magnetization curve looks very similar.

The steps-like behavior is the main characteristic feature of the quantum hysteresis exhibited by the uniaxial spin systems.

Hysteresis phenomena in classical spin systems at finite temperatures have already been investigated in detail. As a function of the applied field the metastable state becomes unstable at the spinodal field and various types of metastabilities have been discussed.[19] For quantum spin systems the concept of metastability is no longer adequate because the system evolves according to a simplectic equation of motion and even unstable states do not relax. However, as we have shown above, in most cases (excluding those where c is relatively small, corresponding to the equilibrium case in classical systems) we find that the largest change of the magnetization as a function of the field occurs at $H\approx H_1$. On general grounds we may expect that $H_2,H_3,\ldots$ are close to H_1 so that the largest step in the magnetization will occur near H_1 even for fast sweeps of the field. Therefore we may call the field H_1 the ''quantum spinodal field''.

V. DISCUSSION

Two aspects of the magnetization dynamics in uniaxial magnets still need to be addressed: the effect of increasing the field step ΔH and the role of dissipation. The former may be of relevance to experiment because in practice it may be difficult to sweep the field sufficiently slow. The latter may be important for a description of the temperature dependence of the staircase structure observed in the experiments.[9–12]

The effect of increasing the field step ΔH is illustrated in Fig. 8. For sufficiently large ΔH we cannot expect the Landau-Zener picture to yield a correct description. Nevertheless the qualitive features of the magnetization curves remain the same. Steps in the magnetization are found at the same values of the field, the size of the steps depending in a nontrivial manner on the sweep speed c and the field step ΔH. Comparison of Figs. 2, 3, and 8 show that changing both these parameters can have a drastic effect on the size of a particular magnetization step. In general, if the field step is not sufficiently small, the magnetization curve depends in a rather complicated manner on the energy-level scheme, the sweep rate c and ΔH itself. Our results suggest that in order to extract energy gaps from the steps in the magnetization curve the best strategy is to take ΔH as small as possible and to change the sweep rate c.

It is evident from Figs. 2–4, 7, and 8 (and from many other similar calculations not shown) that the present calculations do not yield a magnetization that approaches its saturation value if $H \rightarrow \infty$, except for the case that there is no scattering at $H=0$ (not shown). The reason for this is clear: The system studied in this paper can only absorb or release energy through the time-dependent applied field and not through interaction with other degrees of freedom. The latter is required if the spin system is to relax to the ground state, after a scattering event has taken place. Including this coupling into a TDSE calculation of the kind described in this paper is a challenging problem for future research.

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