Nontrivial Response of Nanoscale Uniaxial Magnets to an Alternating Field

Seiji Miyashita and Keiji Saito
Department of Earth and Space Science, Faculty of Science, Osaka University, Toyonaka, Osaka 560

Hans De Raedt
Institute for Theoretical Physics and Materials Science Centre, University of Groningen, Nijenborgh 4, NL-9747 AG, Groningen, The Netherlands
(Received 30 June 1997)

The response of nanoscale uniaxial magnets to an alternating field is studied by direct numerical calculation. A nontrivial oscillation of the magnetization is found and subsequently analyzed in terms of the nonadiabatic transition due to the time dependent field. A new method to estimate the tunneling gap of the magnet is proposed. [S0031-9007(98)05296-X]

PACS numbers: 75.40.Gb, 76.20.+q

The quantum dynamical motion of metastable states has attracted interest recently. If thermal fluctuations allow for excitations over the energy barrier, the relaxation of metastable states is described by Arrhenius's law. In this case the nucleation process plays an essential role. Various field and temperature dependencies of the relaxation rate have been investigated [1], and also the relaxation of the magnetization of a single-domain particle has been studied [2]. The relaxation time resulting from this mechanism is expected to become very long at low temperatures. However, in many cases it has been found that the relaxation time saturates at some temperature. Below this temperature, quantum fluctuations become important and tunneling phenomena may cause the system to relax. The tunneling rate and the tunneling gap are the quantities of interest [3]. The effects of quantum fluctuation on the nucleation process is an interesting problem. However, for small particles, the total magnetization evolves coherently, and the quantum mechanical motion can be studied more explicitly. In fact, such observations have been reported recently [4,5]. Therefore it becomes very interesting to investigate the various features of quantum dynamics in situations where the quantum mechanical motion is directly observed. We have studied the relationship between the tunneling phenomena and the nonadiabatic transition (NAT), a process of purely quantum mechanical origin. In particular, making use of the Landau-Zener-Stückelberg (LZS) [6–8] mechanism, we have proposed a method to estimate the energy gaps from the change of magnetization when the field is swept from H_0 to $-H_0$ during a finite time interval [9,10]. This estimate would provide a good check of the value of the energy gap obtained by other methods.

Recently experimental observations of quantum dynamical phenomena in nanoscale, molecular, magnets such as Mn_{12}-Ac or Fe_8 have been reported [11–15]. Steplike magnetization curves have been found, and the importance of level crossing has been pointed out [16–18]. We also showed that successive NAT's lead to a steplike magnetization curve which is very sensitive to the speed with which the the field changes [19]. To fully describe the experimental situation, the temperature [20,21] and also the effect of random modulations due to the environment [22] need to be taken into account. However, even without including these effects, we may expect to uncover characteristics of NAT, such as sweeping rate dependence of the shape of the magnetization process.

In this Letter, in order to uncover additional characteristics of NAT, we study the quantum mechanical response of the magnetization to an alternating field, adopting a simple model of a uniaxial magnet. We will study the transverse-field Ising model,

$$\mathcal{H} = -J \sum_{\langle i,j \rangle} \sigma_i^z \sigma_j^z - \Gamma \sum_{i=1}^{L} \sigma_i^x - H(t) \sum_{i=1}^{L} \sigma_i^z, \quad (1)$$

where

$$H(t) = H_0 \cos \omega t \,. \quad (2)$$

Throughout this Letter we take J as a unit of energy and put it equal to one. We will only show results for a system of four spins ($L = 4$) subject to periodic boundary conditions and $\Gamma = 0.5$. For other choices of model parameters, the results are qualitatively similar.

In Fig. 1 we present the eigenvalues of the model of Eq. (1) as a function of fixed value of the external field H, which can be regarded as the adiabatic potential of the present model. Only the eight lowest states, some of which are degenerate, are shown. When the energy gap at $H = 0$ is small, the lowest two levels are located far below the other levels. When we take the initial state to be the ground state, the system can be regarded as a two level system, at least if H_0 is small, and there is no second scattering to the higher levels. Successive nonadiabatic transitions to higher levels have been studied in Ref. [19].

If H_0 is very small, we can use Kubo's formula to study the linear response, where the Zeeman term $H(t)M_z$ is treated as a perturbation, and relevant frequencies are only those due to the energy gaps at $H = 0$. On the other hand, in the present paper we are interested in phenomena resulting from nonadiabatic transitions in which $H(t)M_z$

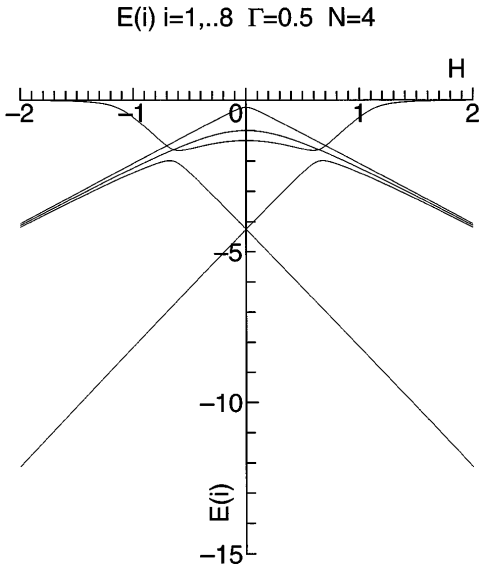

FIG. 1. Energy levels for $\Gamma = 0.5$.

cannot be treated as a perturbation: H_0 is $O(1)$ and not small enough to be treated as a perturbation.

The probability for the system to remain in the ground state when the field changes sign is given by [10]

$$p = 1 - \exp\left(-\frac{\pi(\Delta E)^2}{4c|M_0|}\right), \tag{3}$$

where c is the sweep velocity, $c = |\frac{\mathrm{d}}{\mathrm{d}t} H(t)|_{H(t)=0} = H_0\omega$, and M_0 is the ground-state magnetization near $H = 0$. For $\Gamma = 0.5$, the energy gap at $H = 0$ between the ground state and the first excited state $\Delta E \sim 0.035\,49$ and $|M_0| \sim L = 4$.

The time evolution of the system is given by

$$|t\rangle = e^{-i\int_0^t \mathcal{H}(s)\,ds/\hbar}|0\rangle, \tag{4}$$

where $|0\rangle$ is an initial state which is chosen to be the ground state of the model for $H = H(0)$ and the exponential denotes the time-ordered exponential. We solve Eq. (4) making use of the fourth order decomposition proposed by Suzuki [23,24]. Hereafter we put $\hbar = 1$ for simplicity.

As in our previous studies, the validity of Eq. (3) is confirmed by the simulation results. From Eq. (3), $p = 0.0062$ for $\omega = 0.2$ and $H_0 = 0.2$. In the simulation we calculate the overlap between the ground state and the time dependent state [9]

$$x(t) = |\langle G(t)|t\rangle|^2, \tag{5}$$

where $|G(t)\rangle$ is the ground state for $H = H(t)$. After a half period, $t = \pi/\omega$, we find $x(t = \pi/\omega) = |\langle G(t)|t\rangle|^2 = 0.0063$, which confirms the LZS prediction.

In Fig. 2 we show the time dependence of the magnetization, $M(t) = \langle t|\sum_i \sigma_i^z|t\rangle$, and observe a gradual relaxation due to successive nonadiabatic transitions. When

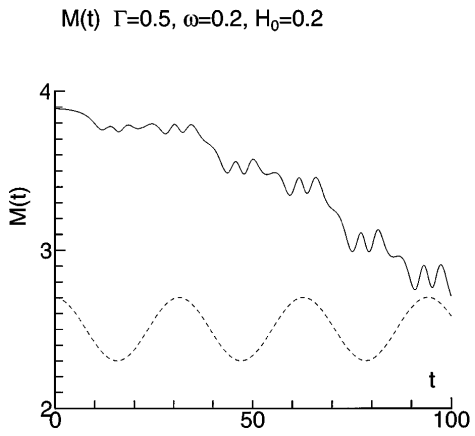

FIG. 2. Time dependence of magnetization. $M(t)$ is shown by a solid line. $H(t) + 2.5$ is shown by a dashed line.

we continue the simulation, a sinusoidal motion,

$$M(t) \sim \cos(\Omega t), \tag{6}$$

is found, as illustrated by Fig. 3 in which we also include $x(t)$ and $H(t)$. The frequency Ω of this sinusoidal motion does not correspond to an eigenfrequency of the system or to the period of the external field. Actually, when we change the amplitude of the field H_0, the period of the magnetization changes as is shown in Fig. 4 and it also depends on ω. Although the dependence of Ω on H_0 in Fig. 4 seems irregular, we find a rather regular dependence when we plot the frequency Ω as a function of H_0, as in Fig. 5.

The nontrivial resonance phenomenon discussed above can be analyzed from the viewpoint of the Floquet theorem: In the presence of a periodic external field the state $|t\rangle$ takes the form $|t\rangle = U(t)\,|t\rangle'$, where $U(t)$ is a matrix of periodic functions of period $2\pi/\omega$. The time evolution of $|t\rangle'$ does not explicitly depend on time. In order to study the dependence of Ω on H_0 and ω, let us

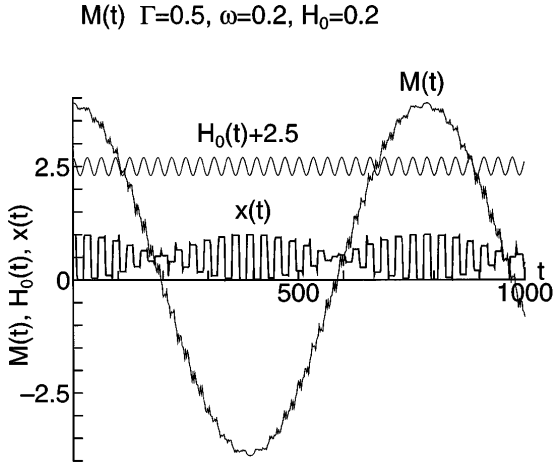

FIG. 3. Nontrivial oscillation of magnetization $M(t)$. $x(t)$ and $H(t)$ are also shown.

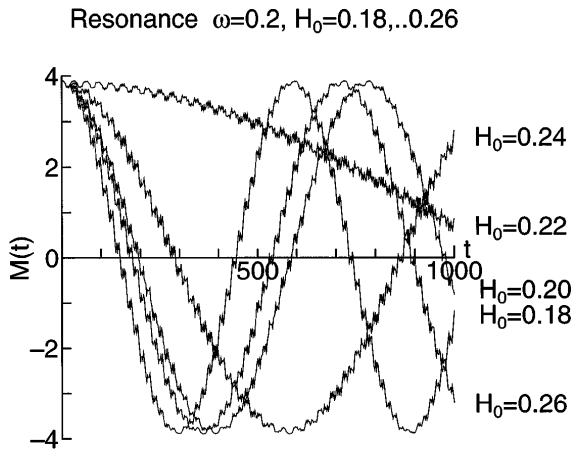

FIG. 4. Time dependences of magnetization for various amplitudes, H_0.

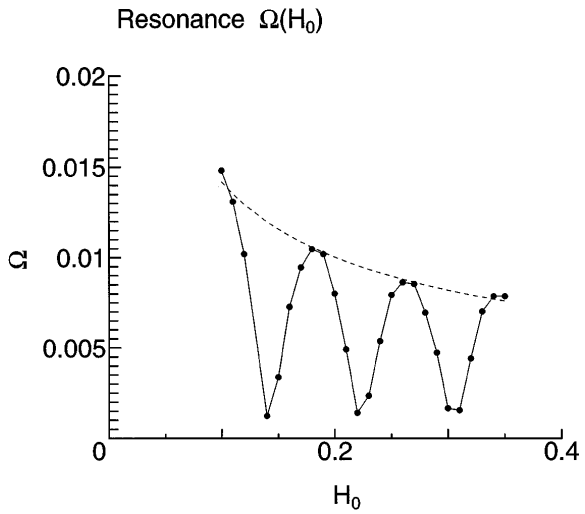

FIG. 5. H_0 dependence of Ω. $\Omega_{\max}$ is shown by a dotted line.

consider the state at $t = 2\pi m/\omega$, $m = 0, 1, 2, \ldots$, where $H(t) = H_0$. First, let us consider the time evolution for the half period, namely, during the field changes from H_0 to $-H_0$: $X = \exp[-i \int_0^{\pi/\omega} \mathcal{H}(s)\, ds]$. As far as H_0 is small and only the lowest two states play an important role, X is expressed by a 2×2 unitary matrix. Here we take the ground state $|G\rangle$ and the first excited state $|1\rangle$ at $t = 0$ as the basis. After a half period the time evolution is expressed as

$$\begin{aligned} X|G\rangle &= t_{11}|G'\rangle + t_{21}|1'\rangle, \\ X|1\rangle &= t_{12}|G'\rangle + t_{22}|1'\rangle, \end{aligned} \tag{7}$$

where $|G'\rangle$ and $|1'\rangle$ denote the ground state and the first excited state at $t = \pi/\omega$, respectively. They can be expressed as a linear combination of $|G\rangle$ and $|1\rangle$: $|G'\rangle = Q_{11}|G\rangle + Q_{21}|1\rangle$ and $|1'\rangle = Q_{12}|G\rangle + Q_{22}|1\rangle$. Let the transformation matrix be Q. The most general form of the unitary matrix for the time evolution which yields the transition probability Eq. (3) must be of the form

$$T = \begin{pmatrix} t_{11}, & t_{12} \\ t_{21}, & t_{22} \end{pmatrix} = \begin{pmatrix} e^{i\theta}\sqrt{p}, & e^{i\phi}\sqrt{1-p} \\ \sqrt{1-p}, & -e^{i(-\theta+\phi)}\sqrt{p} \end{pmatrix}, \tag{8}$$

where θ and ϕ are unknown phases which depend on H_0 and ω. Thus X is expressed by $Q^{-1}T$. To obtain the time evolution for $\pi/\omega \le t \le 2\pi/\omega$ we change the sign of the z component of the spins [for this change we can use the unitary transformation $(\sigma_x, \sigma_y, \sigma_z) \rightarrow (\sigma_x, -\sigma_y, -\sigma_z)$ for all sites], and observe that the time evolution during which the field changes from $-H_0$ to H_0 is identical to X (i.e., the Hamiltonian is identical). Therefore, in the second half of the period $2\pi/\omega$, the time evolution X' can be written as $X' = \exp[-i \int_{\pi/\omega}^{2\pi/\omega} \mathcal{H}(s)\, ds], = Q^{-1}XQ$. The time-evolution operator for one period reads

$$\mathcal{L} = Q^{-1}XQX. \tag{9}$$

When the scattering region of $H(t)$ is very narrow, which is the present case as is clear from Fig. 1, we may take

$$Q = \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}. \tag{10}$$

Combining Eqs. (7), (8), and (10), $\mathcal{L}$ is given by the matrix

$$\begin{pmatrix} e^{2i\theta}p + (1-p)e^{i\phi}, & (e^{i\theta+i\phi} - e^{-i\theta+2i\phi})\sqrt{p(1-p)} \\ (e^{i\theta} - e^{-i\theta+i\phi})\sqrt{p(1-p)}, & e^{-2i(\theta-\phi)}p + (1-p)e^{i\phi} \end{pmatrix}. \tag{11}$$

The eigenvalues, $\lambda_\pm$, of $\mathcal{L}$ are given by

$$\lambda_\pm = (q \pm i\sqrt{1-q^2})e^{i\phi}, \tag{12}$$

where $q = 1 - p + p\cos(\alpha), \alpha = 2\theta - \phi$. In terms of these eigenvalues the frequency Ω reads $\lambda_\pm \equiv \exp[\pm i(\frac{2\pi}{\omega})(\frac{\Omega}{2}) + i\phi]$. Thus the frequency Ω is given by

$$\begin{aligned} \tan\left(\frac{\pi\Omega}{\omega}\right) &= \frac{\sqrt{1-q^2}}{q} \\ &= \frac{\sqrt{2p(1-\cos\alpha) - p^2(1-\cos\alpha)^2}}{1 - p(1-\cos\alpha)}, \end{aligned} \tag{13}$$

and for $p \ll 1$ we have

$$\frac{\pi\Omega}{\omega} \simeq \sqrt{2p(1-\cos\alpha)}. \tag{14}$$

The probability of remaining in the ground state after n periods is given by $x(2\pi n/\omega) = a + b\cos(2\pi\Omega n/\omega + \gamma)$, where a, b, and γ are constants depending on the initial state. When the initial state is the ground state and $p \ll 1$, $a \simeq b \simeq 1/2$ and $\gamma \ll 1$, in concert with the data shown in Fig. 4.

The unknown phase factor $1 - \cos(\alpha)$ can be estimated from an observation within a single period. From

Eqs. (8) and (11), we have $x(\frac{\pi}{\omega}) = p$ and $x(\frac{2\pi}{\omega}) = 1 - 2p(1 - \cos\alpha) + O(p^2)$. Hence the phase factor is given by the ratio $R = \frac{1-x(2\pi/\omega)}{x(\pi/\omega)} = 2(1 - \cos\alpha)$. We found that R estimated from numerical calculations well coincides with $\Omega/\Omega_{\max}$ where $\Omega_{\max} = 2\omega\sqrt{p}/\pi$ which gives the envelope of $\Omega(H_0)$.

In general, we know neither α nor p but even in such a situation, we can estimate $\Omega_{\max}$ by studying Ω as a function of H_0 and ω. Alternatively, knowing $\Omega_{\max}$, we can estimate p from $\Omega_{\max}$ and therefore also ΔE by making use of the relation given by Eq. (3). Here we would like to emphasize that $\sqrt{p}$ is much larger than p since p is assumed to be very small. Therefore even in cases where p is very small Eq. (14) can be used to estimate p.

Effects of an oscillating field on the LZS transition have been studied in different contexts [25–31]. Grossman *et al.* have discovered the coherent destruction of tunneling in a periodically driven two-level system [26–30]. Their results are essentially the same as Eq. (13) for $p \ll 1$. The complete destruction point corresponds to the case $R = 0$. The analysis presented above can be used for any periodic function of $H(t)$, not necessarily $H(t) = H_0 \cos\omega t$. For instance, a piecewise linear, periodic (i.e., a zigzaglike dependence) $H(t)$ yields results (not shown) which are similar to those presented in this paper. The nontrivial oscillation of $M(t)$ is due to nonadiabatic transitions and is a peculiar property of quantum dynamics with a time dependent field. As Kayanuma has pointed out, an oscillation of the transverse field also yields interesting phenomena [31]. It would be of interest to study the response of the magnetization to periodic fields in a more general framework, going beyond the usual perturbative treatment. Evidently a study of the effect on this resonance of a nonzero temperature is a challenging problem for future research.

Nonadiabatic transitions may occur whenever systems become metastable. The nontrivial resonance discussed in this paper is so generic that it should appear if some kind of metastable state is present. We suggest such nontrivial oscillation might be searched for in experiments on nanoscale magnetic systems.

The authors thank Professor Y. Kayanuma for valuable discussions and also appreciate Professor K. M. Slevin for his encouraging discussion and kind critical reading of the manuscript. The present study is partially supported by the Grant-in-Aid for Scientific Research from the Ministry of Education, Science and Culture.

[1] P. A. Rikvold, H. Tomita, S. Miyashita, and S. W. Sides, Phys. Rev. E **49**, 5080 (1994), and references therein.
[2] W. Wernsdorfer, E. Bonet Orozco, K. Hasselbach, A. Benoit, B. Barbara, N. Demony, A. Loiseau, H. Pascard, and D. Mailly, Phys. Rev. Lett. **78**, 1791 (1997).
[3] E. M. Chudnovsky and L. Gunter, Phys. Rev. Lett. **60**, 661 (1988); Phys. Rev. B **37**, 9455 (1988).
[4] D. D. Awshalom, J. F. Smyth, G. Grinstein, D. P. DiVincenzo, and D. Loss, Phys. Rev. Lett. **68**, 3092 (1992); **70**, 2198 (1993); **71**, 4249 (1993).
[5] S. Gider, D. D. Awschalom, T. Douglas, S. Mann, and M. Chaparala, Science **268**, 77 (1995).
[6] L. Landau, Phys. Z. Sowjetunion **2**, 46 (1932).
[7] C. Zener, Proc. R. Soc. London A **137**, 696 (1932).
[8] E. C. G. Stückelberg, Helv. Phys. Acta **5**, 369 (1932).
[9] S. Miyashita, J. Phys. Soc. Jpn. **64**, 3207 (1995).
[10] S. Miyashita, J. Phys. Soc. Jpn. **65**, 2734 (1996).
[11] J. R. Friedman, M. P. Sarachik, T. Tejada, and R. Ziolo, Phys. Rev. Lett. **76**, 3830 (1996).
[12] L. Thomas, F. Lionti, R. Ballou, D. Gatteschi, R. Sessoli, and B. Barbara, Nature (London) **383**, 145 (1996).
[13] J. M. Hernandez, X. X. Zhang, F. Luis, T. Tejada, J. R. Friedman, M. P. Sarachik, and R. Ziolo, Phys. Rev. B **55**, 5858 (1997).
[14] L. Thomas *et al.*, Nature (London) **383**, 145 (1996).
[15] F. Lionti, L. Thomas, R. Ballou, B. Barbara, A. Sulpice, R. Sessoli, and D. Gatteschi, J. Appl. Phys. **81**, 4608 (1997).
[16] D. García-Pablos, N. García, P. Serena, and H. De Raedt, Phys. Rev. B **53**, 741 (1996).
[17] D. García-Pablos, N. García, and H. De Raedt, Phys. Rev. B **55**, 931 (1997).
[18] D. García-Pablos, N. García, and H. De Raedt, Phys. Rev. B **55**, 937 (1997).
[19] H. De Raedt, S. Miyashita, K. Saito, D. García-Pablos, and N. García, Phys. Rev. B. (to be published).
[20] E. M. Chudnovsky, Science **274**, **938** (1996).
[21] P. C. E. Stamp, Nature (London) **383**, 125 (1996).
[22] N. V. Prokof'ev and P. C. E. Stamp, J. Low Temp. Phys. **104**, 143 (1996).
[23] M. Suzuki, J. Phys. Soc. Jpn. **61**, 3015 (1992).
[24] M. Suzuki, Proc. Jpn. Acad. Ser. B **69**, 161 (1993).
[25] J.-M. Lopez-Castillo, A. Filali-Mouhim, and J.-P. Jay-Gerin, J. Chem. Phys. **97**, 1905 (1992).
[26] F. Grossmann, T. Dittrich, P. Jung, and P. Hänggi, Phys. Rev. Lett. **67**, 516 (1991).
[27] F. Grossmann, T. Dittrich, P. Jung, and P. Hänggi, Z. Phys. B **84**, 315 (1991).
[28] J. M. Gomez Llorente and J. Plata, Phys. Rev. A **45**, R6958 (1992).
[29] Y. Kayanuma, Phys. Rev. A **50**, 843 (1994).
[30] Y. Kayanuma, Phys. Rev. B **47**, 9940 (1993).
[31] Y. Kayanuma, Phys. Rev. A **55**, R2495 (1997).