Feedback effect on Landau-Zener-Stückelberg transitions in magnetic systems

Anthony Hams and Hans De Raedt
Institute for Theoretical Physics and Materials Science Centre, University of Groningen, Nijenborgh 4, NL-9747 AG Groningen, The Netherlands

Seiji Miyashita and Keiji Saito
Department of Applied Physics, School of Engineering, University of Tokyo, Bunkyo-ku, Tokyo 113, Japan
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We examine the effect of the dynamics of the internal magnetic field on the staircase magnetization curves observed in large-spin molecular magnets. We show that the size of the magnetization steps depends sensitively on the intermolecular interactions, even if these are very small compared to the intramolecular couplings.

The magnetization dynamics of nanoscale magnets, i.e., systems like Mn_{12}-acetate and Fe_8, have been studied experimentally and theoretically lately.[1–7] At sufficiently low temperatures quantum effects are observed, due to the discreteness of the energy levels involved. When the magnetization of a crystal of such molecules is measured during a sweep of the external magnetic field, a staircase hysteresis loop is obtained. The steep parts of the staircase correspond to the values of the external magnetic field where there is a crossing of adiabatic energy levels. Several aspects of this quantum effect were studied in Refs. 8–13. In a zero-temperature calculation, one finds that the magnetization can only change in steps, very similar to the steps observed in recent experiments on high-spin molecules Mn_{12}-acetate and Fe_8. At every crossing, only two levels play a role and the transition probability can be calculated using the Landau-Zener-Stückelberg (LZS) mechanism.[14–16] Two parameters determine the LZS transition: the energy splitting at the crossing and the sweep rate of the magnetic field.

The size of the energy splitting which leads to a LZS transition probability is determined by the off-diagonal terms in the Hamiltonian describing the system. A straightforward perturbative calculation shows that this splitting is roughly scaled like $\Gamma^{2|\Delta m|}$ where Γ determines the magnitude of the off-diagonal terms and Δm denotes the difference in magnetization of the two relevant levels. In the absence of a transverse applied field the energy-level splittings in the high-spin molecules mentioned above are so small that the probability for a single LZS transition is effectively zero, unless the applied longitudinal field is rather large (see, for example, Ref. 7).

In the crystal the magnetic field felt by a particular molecule is the sum of the external field and the internal field due to the presence of other magnetic molecules. As the intermolecular magnetic couplings in these materials are weak compared to the intramolecular interaction between the spins, it seems reasonable to consider the former as a perturbation. The purpose of this paper is to demonstrate that this argument fails in the case of LZS transitions. The point is that the LZS transition probability depends on the rate of change of the effective magnetic field *at the crossing*, which can be changed significantly by the presence of the internal magnetic field. The magnetization steps are found to be strongly affected by the type of interactions among molecules. We call this mechanism the feedback effect on magnetization steps (FEMS).

We first illustrate the effect for the case of the Mn_{12}-acetate molecules. As a model Hamiltonian for this $S=10$ system we take[7]

$$\mathcal{H} = -D_1 S_z^2 - D_4(S_x^4 + S_y^4 + S_z^4) - ct \sin\theta S_x - (ct\cos\theta + \lambda\langle S_z\rangle)S_z . \quad (1)$$

Compared to the model of Ref. 7 the extra feature in Hamiltonian (1) is the presence of a mean-field term, the strength of which we parametrize by λ. It is clear that in this mean-field approach any new effect appears as a result of global changes of the internal field generated by all the molecules and is not due to local fluctuations which should be treated separately.[19] It is important to note that the FEMS is due to the internal spin dynamics and is also present in the absence of interactions with other degrees of freedom.[20–26]

Quantitative results for the zero-temperature nonequilibrium dynamics of model (1) can only be obtained through a numerical integration of the Schrödinger equation. Using standard techniques[10] we compute the magnetization steps for several values of λ. The results for $D_1=0.64$, $D_4=0.004$, tilt angle $\theta=1°$, and sweep rate $c=0.001$ (see Ref. 7; we use dimensionless units throughout this paper) are shown in Fig. 1.

It is clear that the dynamics of the internal field can change the size of the magnetization steps considerably. The FEMS is observed for all $\lambda\neq 0$. Note that the values of $|\lambda|$ we used are not unrealistic ($|\lambda|\approx D_4 \ll D_1$), but rather small if we relate λ to the dipole-dipole interaction which would yield a λ which is 10–100 times larger.[18]

At very low temperatures experiments[7] show steps at lower values of H than the ones at which we observe steps in our calculation. In fact, for the set of model parameters given in Eq. (1) a much slower sweep rate c, much too slow for numerical calculations, is required if we want to study the effect of the internal field at all level crossings.

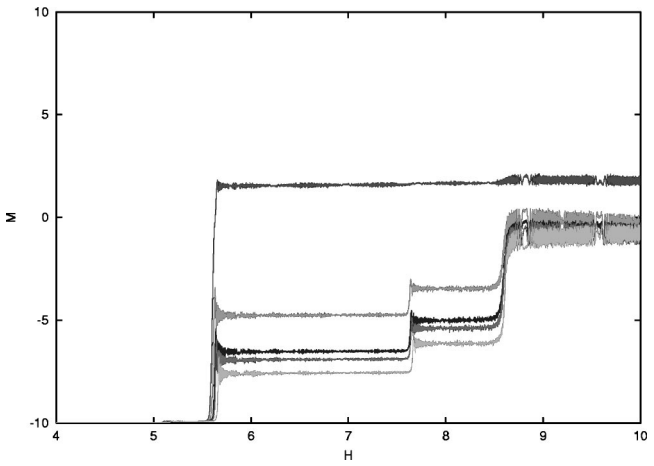

FIG. 1. Magnetization dynamics of the Mn_{12}-acetate model (1), for several values for the intermolecular coupling, $\lambda=-0.005$ (top curve), -0.003, -0.001, 0, 0.003 (bottom curve).

Therefore it is expedient to turn to a toy model inspired by the one used to describe Fe_8.[6] We take a $S=2$ model with the following Hamiltonian:[6]

$$H=-DS_z^2+E(S_x^2-S_y^2)+\Gamma S_x-(ct+\lambda\langle S_z\rangle)S_z, \qquad (2)$$

where we take $D=1$, $E=0.08$, and $\Gamma=0.08$. These parameters are chosen such that we get two steps with a probability of about one-half.

In Fig. 2 we show the magnetization during a sweep of the magnetic field, with a sweep rate $c=0.01$, for several values of λ. We see that the FEMS effect is large.

The transition probabilities are given in Table I. We clearly see a large change in the transition probabilities due to the presence of the internal field.

A deeper understanding of the origin of the FEMS effect can be obtained by considering the system of N $S=1/2$ molecules described by the Hamiltonian

$$\mathcal{H}=\sum_{i=1}^{N}\left[-\Gamma\sigma_i^x-J\sum_{j>i}^{N}\sigma_i^z\sigma_j^z+ct\sigma_i^z\right], \qquad (3)$$

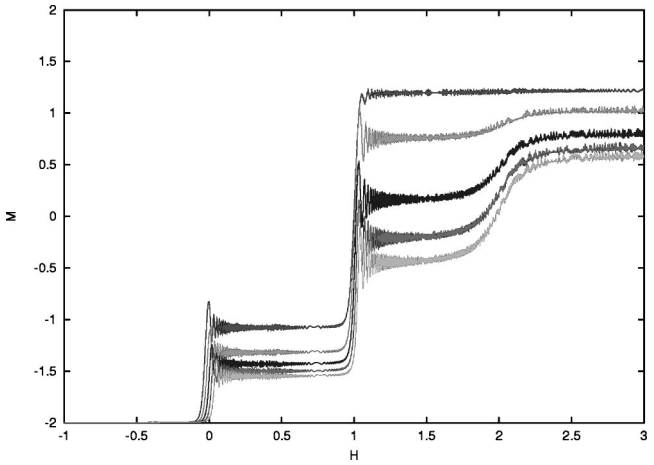

FIG. 2. Magnetization dynamics of the $S=2$ model (2), for several values for the intermolecular coupling, $\lambda=-0.03$ (top curve), -0.02, -0.01, 0, 0.01 (bottom curve).

TABLE I. Transition probabilities corresponding to the steps in Fig. 2.

λ	-0.03	-0.02	-0.01	0	0.01
Step 1	0.23	0.17	0.15	0.13	0.12
Step 2	0.90	0.78	0.59	0.48	0.40

where c is the sweep rate, Γ is the transverse field, and J determines the interaction strength between the molecules ($|J|\ll\Gamma$). For simplicity we consider couplings between z components only and assume the coupling between the molecules to be the same. Since $|J|$ is small, we assume that we can make a mean-field-like approximation. The occurrence of the FEMS does not depend on these simplifications (see below). This yields a Hamiltonian of a single molecule in a background field:

$$\mathcal{H}=-\Gamma\sigma_x-(ct+\lambda\langle\sigma_z\rangle)\sigma_z, \qquad (4)$$

where $\lambda\propto J$ is an effective interaction. The system is prepared in the ground state, corresponding to a large negative time t, and the magnetic field is swept with constant velocity, until a large positive time is reached. Then, in the LZS case with $\lambda=0$, the transition probability is given by the well-known LZS formula $p=1-\exp(-\pi\Gamma^2/c)$. For $\lambda\neq 0$ we write the Schrödinger equation corresponding to Eq. (4) in component form:

$$iu'=[-ct-\lambda(2|u|^2-1)]u-\Gamma d, \qquad (5)$$

$$id'=[ct+\lambda(2|u|^2-1)]d-\Gamma u, \qquad (6)$$

where we also have the normalization condition $|u|^2+|d|^2=1$. From numerical simulations we (see below) find that the tunneling is suppressed (enhanced) by the presence of a feedback term with positive (negative) λ. This can be understood in terms of a changed effective sweep rate at the point of the transition. Because the effective magnetic field at the position of the molecule is given by $ct+\lambda(2|u|^2-1)$, the effective sweep rate would be $c+\lambda d\langle\sigma_z\rangle/dt$.

If λ is small but nonzero, the mean-field term only contributes at the point of the crossing. So we look at a Taylor expansion around the point of the transition t_c (to be determined later),

$$u(t)=u_0+u_1(t-t_c)+\mathcal{O}((t-t_c)^2), \qquad (7)$$

and a similar expression for $d(t)$. We insert this expansion in Eqs. (5) and (6) and obtain

$$i\tilde{u}'(\tilde{t})=-\tilde{c}\tilde{t}\tilde{u}(\tilde{t})-\Gamma\tilde{d}(\tilde{t}), \qquad (8)$$

$$i\tilde{d}'(\tilde{t})=\tilde{c}\tilde{t}\tilde{d}(\tilde{t})-\Gamma\tilde{u}(\tilde{t}), \qquad (9)$$

with $\tilde{t}=t-t_c$, $\tilde{c}=c+4\lambda\,\mathrm{Re}(u_0u_1^*)$ is the renormalized sweep rate, and were $\mathrm{Re}(u)$ denotes the real part of u. We define t_c as the point at which $ct+\lambda\langle\sigma_z\rangle$ changes sign, so

$$t_c=\frac{\lambda}{c}(1-2|u_0|^2). \qquad (10)$$

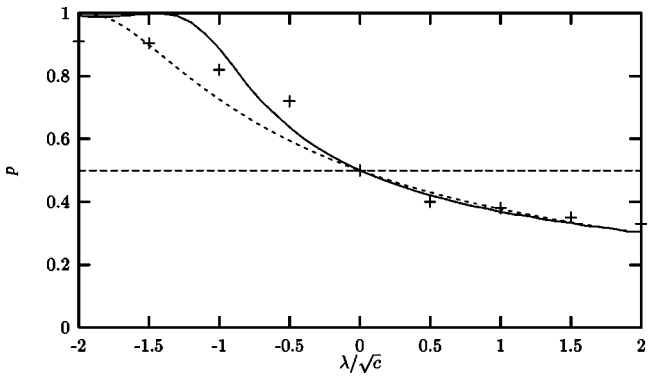

FIG. 3. Transition probability as a function of $\lambda/\sqrt{c}$, with Γ^2/c such that in the LZS case, $p=1/2$. The solid line is based on a numerical integration of Eq. (4), the crosses are taken from a simulation of four interacting $S=1/2$ spins, and the dashed line is based on Eq. (16).

This enables us to write $\tilde{u}_0=u_0$ and $\tilde{u}_1=u_1$. To determine these constants we use Zener's solution[15,17] and the properties of Weber functions. We find

$$|u_0|^2=\frac{\pi}{4}\delta e^{-\pi\delta/4}\left|\frac{1}{\Gamma\left(1+\frac{i\delta}{4}\right)}\right|^2=\frac{1}{2}[1-(1-p)^2], \tag{11}$$

where p is the new probability for crossing, i.e., $p=1-\exp(-\pi\Gamma^2/\tilde{c})$ and $\delta=\Gamma^2/\tilde{c}$, and $t_c=\lambda(1-p)^2/c$. The shift of the field at which the transition occurs can be written as $\Delta H=\lambda(1-p)^2$. To determine $\tilde{c}$ we calculate

$$\mathrm{Re}(u_0\,u_1^*)=\sqrt{\tilde{c}}\,\frac{\pi\delta}{2}e^{-\delta\pi/4}\,\mathrm{Re}\,\frac{e^{-i\pi/4}}{\Gamma(1+i\delta/4)\Gamma\left(\frac{1}{2}-i\delta/4\right)}. \tag{12}$$

We find that Eq. (12) can be approximated by

$$\mathrm{Re}(u_0\,u_1^*)\approx\sqrt{\frac{\pi\tilde{c}}{8}}\delta e^{-\delta\pi/4}, \tag{13}$$

with an error of maximally 10% (see Fig. 3). Within this approximation, $\tilde{c}$ is given by the implicit equation

$$\tilde{c}\approx c+\lambda\Gamma^2\sqrt{2\pi/\tilde{c}}e^{-\Gamma^2\pi/4\tilde{c}}. \tag{14}$$

A simple relation can be obtained by replacing $\tilde{c}$ by c on the right hand side. Then

$$\tilde{c}\approx c+\lambda\Gamma^2\sqrt{2\pi/c}e^{-\pi\Gamma^2/4c} \tag{15}$$

and

$$p\approx 1-\exp\left(-\frac{\pi\Gamma^2}{c}\,\frac{1}{1+\lambda\Gamma^2\sqrt{2\pi/c^3}e^{-\Gamma^2\pi/4c}}\right). \tag{16}$$

The resulting probabilities are shown in Fig. 3. The resulting probabilities based on a numerical solution of Eq. (13) or (14) for $\tilde{c}$ show similar behavior. Also shown are the results obtained from the exact numerical solution of the Schrödinger equation (4). As a test of the validity of the mean-field approximation we also show the result of four interacting $S=1/2$ spins, where we assumed $\lambda=(N-1)J$. Clearly the exact results confirm the validity of the mean-field approximation and the simple analytic expression (16).

For values of λ below approximately -2.0 the description in terms of a renormalized sweep rate breaks down, which can also be seen from the singularity in the argument of the exponent in Eq. (16). This is because the picture of a simple, single crossing breaks down and the effective magnetic field at the position of the spin will no longer be a strictly increasing function of time. We conclude that the expression (16) captures the main features of the FEMS at a single crossing.

The relevant parameters, controlling the size of the FEMS, are $\Gamma/\sqrt{c}$ and $\lambda/\sqrt{c}$. Only for $S=1/2$ is the energy-level splitting directly proportional to Γ^2. For the high-spin molecules this is not the case (see above), in particular for the levels with large $|m|$. Although in these cases the effective energy level splitting that enters the approximate two-level description can be small, a rather small value of λ can nevertheless change the transition probability significantly.

We have shown that the magnetization steps in the hysteresis loops of clusters of high-spin molecules may depend sensitively on the change of the internal magnetic field at these steps. This implies that the dynamics of this internal field has to be incorporated in a description of the magnetization dynamics, even if its magnitude appears to be small compared to the other model parameters (for large spin). At finite temperatures the effect described in this paper will be enhanced further due to the thermalization to states with lower energy and larger magnetization.[27]

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