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Dynamic switching of the magnetization in a driven molecular nanomagnet

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Abstract
We study the magnetization dynamics of a single molecular nanomagnet driven by static and variable magnetic fields within a classical treatment. The underlying analysis is valid for a regime where the energy is definitely lower than the anisotropy barrier, but still a substantial number of states are excited. We find the phase space to contain a separatrix line. Solutions far from it are oscillatory whereas the separatrix solution is of a soliton type. States near the separatrix are extremely sensitive to small perturbations, a fact that we utilize in obtaining dynamically induced magnetization switching. A new type of magnetization switching is proposed.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Single molecular nanomagnets (MNM) are molecular structures with a large effective spin (S), e.g. for the prototypical MNM Mn_{12} acetates [1] $S = 10$. MNM show a number of interesting phenomena that have been the focus of theoretical and experimental research [1–11]. To name but a few, as a result of the strong uniaxial anisotropy, MNM show a bistable behaviour [1]; they also exhibit a resonant tunnelling of magnetization [2] that shows up as steps in the magnetic hysteresis loops [3–5]. Of special relevance for applications in information storage is the large relaxation time of MNM [12].

The present theoretical work focuses on the dynamics of the magnetization. The established picture of macroscopic quantum tunnelling of the magnetization is as follows: the MNM effective spin Hamiltonian $\hat{H} = -DS_z^2$ possesses degenerate energy levels $\pm M_S, -S < M_s < S$ separated by the finite barrier $E_B = DS^2$. At low temperatures only the lowest levels $M_S = \pm S$ are populated. Those two states are orthogonal to each other and no tunnelling is possible. An anisotropic perturbation $E(S_x^2 - S_y^2)$ does not commute with the Hamiltonian $\hat{H} = -DS_z^2$ and mixes therefore the states at both sides of the anisotropy barrier leading thus to tunnelling [8]. Reversal of the magnetization due to macroscopic quantum tunnelling has a maximum for the states close to the top of the barrier. This case corresponds to the high temperature limit.

In this work we consider the magnetization dynamics induced by constant and harmonic external magnetic fields: the influence of a variable magnetic field on MNM at low temperatures, i.e. when only 2–3 levels are excited was considered in [10, 11]. It was shown that in this case the problem is reduced to a three level Jaynes–Cummings model, the so called Lambda configuration. Therefore, it is analytically solvable in principle. The low temperature assumption is, however, quite restrictive [12]: if only the levels E^0, E^1, E^2 are involved the low temperature approximation is applicable for temperatures obeying [11] $k_B T < E^1 - E^0$, where k_B is the Boltzmann constant. For Mn_{12} this leads to the estimate $T < 0.6$ K [13]. Obviously, if the temperature exceeds T, an approximation with a large number of levels participating in the process is more appropriate. In this case the quasi-classical approximation for the spin dynamics becomes applicable [14, 15]. It is our aim here to conduct such a study. MNM will be modelled as in previous studies, e.g. in [10, 11]. We consider the dynamical reversal of the magnetization, caused not by an anisotropic perturbation but by a constant and varying magnetic field. The energy is such that a large number of levels are excited, but still low enough such that tunnelling induced by an anisotropic perturbation is weak. We shall show that under different conditions (depending on the field's parameters), different types of magnetization dynamics are realized. For the time evolution of the magnetization vector, under certain conditions we obtain a solution of the soliton type. The various types of the dynamics will be linked to the structure of the phase space of the system. In particular, the existence of the separatrix in the phase space has a profound influence on the system's behaviour. We will show that in this case a new type of field-assisted magnetization dynamics emerges, namely a dynamically induced switching. This occurs when the energy of the system (in the presence of the field) is still lower than the re-scaled anisotropy barrier [16] and coincides with the separatrix values of the energy. Therefore, the domain close to the separatrix is identified as the phase space area where the dynamically induced switching takes place.

We note that classical dynamic of magnetic nanoparticles is well studied in a number of special cases. Problems concerning nanoparticle magnetization switching were addressed in several works [17–21]. In the case when the steady state precession is around equilibrium, the problem can be linearized. A switching effect was then predicted for the case of a zero static field [17, 18]. In contrast, as will be shown in the present study, a constant field applied along the hard axis (x-axis) leads to a complex phase space of the system, a fact that will be utilized to identify a new switching mechanism. We also note that due to the high anisotropy barrier of the single molecular magnets, dissipative processes are less relevant for MNP dynamics. Dissipative processes become important at higher energies. However at the energies near barrier hight transverse anisotropy terms become important [6] and lead to a markedly changed structure of the phase space. These effects therefore are beyond the scope of the present paper.

2. Model

We consider a molecular magnet, e.g. Fe_8 or Mn_{12} acetate. The uniaxial anisotropy axis (easy axis) sets the z-direction. The MNM is subjected to a constant magnetic field directed along the x-axis and a radio frequency (rf) magnetic field polarized in the x–y-plain. The Hamiltonian of the single molecular magnet reads [11]

$$\begin{aligned}\hat{H} &= \hat{H}_0 + \hat{H}_I,\\ \hat{H}_0 &= -D\hat{S}_z^2 + g\mu_B H_0 \hat{S}_x,\\ \hat{H}_I &= -\tfrac{1}{2} g\mu_B H_1 e^{i\omega_0 t}(\hat{S}_y + \hat{S}_x) + \text{h.c.}\end{aligned} \tag{1}$$

Here D is the longitudinal anisotropy constant, $\hat{S}_x$, $\hat{S}_y$, $\hat{S}_z$ are the projections of the spin operators along the x, y, z-axis, g is the Landé factor, and μ_B is the Bohr magneton. H_0 stands for the constant magnetic field amplitude whereas H_1, and ω_0 are the amplitude and the frequency of the rf field and h.c. means hermitian conjugate. The problem when both fields H_0, H_1 are time dependent was studied in [22]. Using quantum-mechanical perturbation theory, the probability of quantum tunnelling of magnetization has been estimated. However, here we are interested in the exact solution of the classical equations of motion. Typical values of the parameter D are 90 GHz for Mn_{12} and $D = 30$ GHz for Fe_8 [13, 15]. Since we are interested in the case when a large number of levels are excited, the spin of the magnetic molecule can be treated as a classical vector on the Bloch sphere. Taking into account that $S^2 = S_x^2 + S_y^2 + S_z^2$ is an integral of motion, it is appropriate to switch to the new variables (S_z, φ) via the transformation [14]: $S_x = \sqrt{1 - S_z^2}\cos\varphi$, $S_y = \sqrt{1 - S_z^2}\sin\varphi$ and rewrite (1) in the compact form:

$$\begin{aligned}H &= -\frac{\lambda}{2}S_z^2 + \sqrt{1 - S_z^2}\cos\varphi\\ &\quad - \varepsilon\sqrt{1 - S_z^2}(\sin\varphi + \cos\varphi)\cos(\omega_0 t).\end{aligned} \tag{2}$$

Hereafter, if not otherwise stated the energy and the timescales are set by the constant magnetic field $H \mapsto H/g\mu_B H_0 S$, $t \mapsto \frac{2DS}{\lambda}t$, $\omega_0 \mapsto \frac{\lambda}{2DS}\omega_0$. We introduced two dimensionless parameters $\lambda = \frac{2DS}{g\mu_B H_0}$, $\varepsilon = \frac{H_1}{H_0} < 1$. The corresponding Hamilton equations are

$$\begin{aligned}\dot{S}_z &= -\frac{\partial H}{\partial\varphi} = \sqrt{1 - S_z^2}\sin\varphi\\ &\quad + \varepsilon\sqrt{1 - S_z^2}(\cos\varphi - \sin\varphi)\cos(\omega_0 t),\\ \dot{\varphi} &= \frac{\partial H}{\partial S_z} = -\left(\lambda + \frac{\cos\varphi}{\sqrt{1 - S_z^2}}\right)S_z\\ &\quad + \varepsilon\frac{S_z}{\sqrt{1 - S_z^2}}(\sin\varphi + \cos\varphi)\cos(\omega_0 t).\end{aligned} \tag{3}$$

These equations are nonlinear. Therefore, the solutions to (3) can be regular or chaotic, depending on the values of the magnetic fields (parameters λ, ε). From the intuitive point of view it is obvious, that for the low energy case, i.e. close to the ground states $S_z \approx \pm 1$, the system equation (2) should become linear. However in the language of variables action angle (S_z, φ) that is not so trivial. Therefore, we will discuss this question in more detail when studying solutions of the autonomous system.

3. Autonomous system: an exact solution

We inspect at first the autonomous system, i.e. when $\varepsilon = 0$. In this case the system can be integrated exactly: taking into energy conservation $H = \text{const} = -\Sigma$

$$\frac{\lambda}{2}S_z^2 - \sqrt{1 - S_z^2}\cos\varphi = \Sigma \tag{4}$$

and

$$\dot{S}_z = \sqrt{1 - S_z^2}\sin\varphi, \tag{5}$$

we find

$$\dot{S}_z^2 + \left[\frac{\lambda S_z^2}{2} - \Sigma\right]^2 = 1 - S_z^2. \tag{6}$$

Consequently from equation (6) we infer

$$\frac{\lambda t}{2} = \int_{S_z(t)}^{S_z(0)} \frac{dS_z}{\sqrt{(\frac{2}{\lambda})^2(1 - S_z^2) - [S_z^2 - \frac{2\Sigma}{\lambda}]^2}}. \tag{7}$$

This relation can be rewritten in the form

$$\frac{\lambda t}{2} = \int_{S_z(t)}^{S_z(0)} \frac{dS_z}{\sqrt{(a^2 + S_z^2)(b^2 - S_z^2)}}, \tag{8}$$

where $a^2 = \frac{2}{\lambda^2}[\theta^2/2 - (\Sigma\lambda - 1)]$, $b^2 = \frac{2}{\lambda^2}[\theta^2/2 + (\Sigma\lambda - 1)]$, $\theta^2(\lambda) = 2\sqrt{\lambda^2 - 2\Sigma\lambda + 1}$. Performing the integration (8) and inverting the result we obtain

$$S_z(t) = \begin{cases} b\,\mathrm{cn}[(b\lambda/k)(t-\alpha), k], & 0 < k < 1, \\ b\,\mathrm{dn}[(b\lambda/k)(t-\alpha), 1/k], & k > 1. \end{cases} \tag{9}$$

Here cn$(\cdots)$ and dn$(\cdots)$ are the Jacobi periodic functions. The coefficients that enter equation (9) read

$$k^2 = \frac{1}{2}\left(\frac{b\lambda}{\theta(\lambda)}\right)^2 = \frac{1}{2}\left[1 + \frac{(\Sigma\lambda - 1)}{\sqrt{\lambda^2 + 1 - 2\Sigma\lambda}}\right], \\ \alpha = 2[\lambda\sqrt{a^2 + b^2}F(\arccos[S_z(0)/b], k)]^{-1}. \tag{10}$$

With $F(\varphi, k) = \int_0^{\varphi} \mathrm{d}q(1 - k^2 \sin^2 1)^{-1/2}$ being the incomplete elliptical integral of the first kind. From equation (9) we conclude that, depending on the values of the parameter k (10), the dynamics of the magnetization is described by different solutions. They are separated by the special value $k = 1$ of the bifurcation parameter k indicating thus the presence of topologically distinct solutions. In equation (9) the Jacobian elliptic functions cn(φ, k) and dn(φ, k) are periodic in the argument φ with the period $4K(k)$ and $2K(k)$ respectively, where $K(k) = F(\pi/2, k)$ is the complete elliptic integral of the first kind [23]. The time period of the oscillation of the magnetization $S_z(t)$ is given by

$$T = \begin{cases} \dfrac{4kK(k)}{b\lambda} & \text{for } 0 < k < 1, \\ \dfrac{2kK(1/k)}{b\lambda} & \text{for } k > 1. \end{cases} \tag{11}$$

If $k \longrightarrow 1$, the period becomes infinite because $K(k) \longrightarrow \ln(4/\sqrt{1-k^2})$. The evolution in this special case is given by the non-oscillatory soliton solution

$$S_z(t) = b/\cosh[b\lambda(t - \alpha)]. \tag{12}$$

Considering equation (10), we infer that the bifurcation value of the parameter $k = 1$ is connected with an initial energy of the system via the ratio

$$\Sigma_S = -H_S/g\mu_B H_0 = 1, \\ H(S_z(t=0); \varphi(t=0)) = -g\mu_B H_0 = H_S. \tag{13}$$

If this condition (13) is not fulfilled the dynamics of the magnetization is described by the solutions (9). Finally, to conclude this section we consider linear limit of solutions equation (9):

$$\mathrm{cn}(u, k) \approx \cos(u) + k^2 \sin(u)(u - \tfrac{1}{2}\sin(2u)), \qquad k^2 \ll 1,$$

and

$$\mathrm{d}n(u, k) \approx 1 - \sin(u)^2/k^2, \qquad k^2 \gg 1.$$

The interpretation of those asymptotic solutions is clear. First one corresponds to the case when in the effective magnetic field $H_{\text{eff}} = (g\mu_B H_0, 0, -DS_z)$, the x-component is dominant. Therefore the magnetization vector performs small oscillations $|S_z(t)| < 1$ trying to be aligned along effective magnetic field. While in the second case, corresponding to the ground state solution (system is near to the bottom of double potential well) the effective magnetic field is directed along the z-axis.

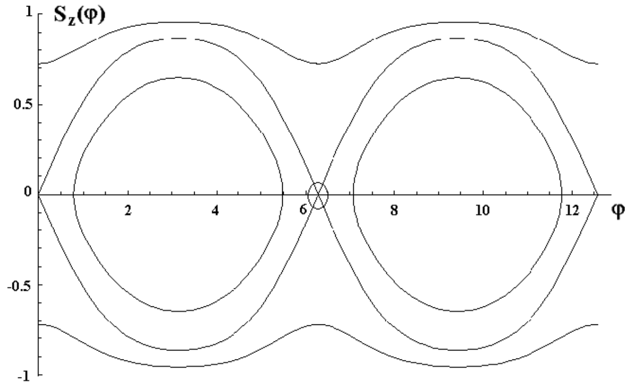

Figure 1. Two types of phase trajectories of the system separated by the separatrix $k = 1$, $\Sigma = \Sigma_S$. The open trajectory (solution $S_z(t) = \mathrm{dn}(t, 1/k)$, $k = 1.52$, $\Sigma > \Sigma_S$) corresponds to the rotational regime of motion. The closed trajectory (solution $S_z(t) = \mathrm{cn}(t, k)$, $k = 0.89$, $\Sigma < \Sigma_S$) to the oscillatory regime. The separatrix crossing point $\otimes$ is of special interest: around this point any perturbation leads to the formation of homoclinic structure.

4. Topological properties of solutions

As established [24, 25], the existence of a bifurcation parameter indicates that the solutions separated by it, have different topological properties. Therefore, it is instructive to consider the properties of the solutions (9) in the phase plane. The existence of the integral of motion (4) in the autonomous case makes it possible to express S_z as a function of φ: $S_z(\varphi, \Sigma)$. The phase portrait of the system is shown in figure 1: the different phase trajectories correspond to the solutions (9). The phase trajectories corresponding to the solution $S_z(t) = \mathrm{dn}(\varphi, k)$, $k > 1$ are open and they describe a rotational motion of the magnetization. Trajectories corresponding to $S_z(t) = \mathrm{cn}(\varphi, k)$, $k < 1$ are closed and they describe the oscillatory motion of the magnetization. Closed and open phase trajectories are separated from each other by the special line called separatrix. The existence of a separatrix is insofar important as the states in the phase space area near the separatrix are very sensitive [25] to external perturbations, which signals the onset of chaotic behaviour. The role of perturbations in our particular case is played by the applied periodic magnetic field. We recall that the stochastic layer has finite size and it occupies a small part of the phase space.

5. Formation of a stochastic layer

To determine the width of the stochastic layer we follow [25]. For details of the formation of the stochastic layer and for the general formalism we refer to the monograph [25]. Here we only present the main findings. We introduce the canonical variable of action $I = \frac{1}{\pi}\oint S_z(\Sigma, \varphi)\,\mathrm{d}\varphi$ and rewrite the driven nonlinear system (2) in the following form:

$$H = H_0 + \varepsilon V(I, \varphi)\cos(\omega_0 t). \tag{14}$$

Here $H_0 = \omega(I)I$, $\omega(I) = [\frac{\mathrm{d}I(\Sigma)}{\mathrm{d}\Sigma}]^{-1}$. The trajectories laying far from separatrix of the unperturbed Hamiltonian H_0 are not influenced by perturbation. The motion near the homoclinic points of the separatrix is very slow [25]. Because the period of motion described by (11) is logarithmically divergent, even small perturbations end up with a finite influence due to the large period of motion. Thus, the equations of motion for the canonical variables (I, φ)

$$\dot{I} = \frac{\partial I}{\partial H_0}\dot{H} = -\frac{\varepsilon}{\omega(I)}\frac{\partial V}{\partial S_z}\dot{S}_z\cos(\omega_0 t), \tag{15}$$

$$\dot{\varphi} = \frac{\partial H}{\partial I} = \omega(I) + \varepsilon\frac{\partial V}{\partial S_z}\dot{S}_z\cos(\omega_0 t), \tag{16}$$

may be integrated taking into account the features of the motion near to the separatrix. Namely, the acceleration $\dot{S}_z$ gives a nonzero contribution in the integral $\int \mathrm{d}t \frac{\partial V}{\partial I}\dot{S}_z cos(\omega_0 t)$ only near to the homoclinic points [25] (the particle moves along the phase trajectory very fast and spends most of the time near the homoclinic points). Therefore, the differential equations (15), (16) can be reduced to the following recurrence relations:

$$\bar{I} = I - \frac{\varepsilon}{\omega(I)}\int_{\Delta t}\mathrm{d}t\frac{\partial V}{\partial S_z}\dot{S}_z\cos(\omega_0 t), \tag{17}$$

$$\bar{\varphi} = \varphi + \frac{\pi\omega_0}{\omega(\bar{I})}. \tag{18}$$

Here $\bar{I}, \bar{\varphi}$, and I, φ are the values of the canonical variables just after and before passing the homoclinic point, Δt is the interval of the time where $\dot{S}_z$ is different from zero. One can deduce the coefficient of stochasticity by evaluating the maximal Lyapunov exponent for the Jacobian matrix

$$\begin{pmatrix} \frac{\partial\bar{I}}{\partial I} & \frac{\partial\bar{I}}{\partial\varphi} \\ \frac{\partial\bar{\varphi}}{\partial I} & \frac{\partial\bar{\varphi}}{\partial\varphi} \end{pmatrix}, \tag{19}$$

of the recurrence relations (17), (18). All of this subsume to the following expression for the width of the stochastic layer

$$K_0 = \frac{\pi\varepsilon\omega_0}{\omega^2}\left|\frac{\mathrm{d}\omega}{\mathrm{d}H}\right|. \tag{20}$$

Here ε, ω_0 are the amplitude and the frequency of the perturbation. Note that the expression (20) is general [25] and the only thing one has to do is to calculate the nonlinear frequency $\omega(I)$ and its derivative with respect to the energy for the particular system. Thus, even for small perturbation (in our case it is the magnetic field with the frequency ω_0 and the amplitude ε, see equation (2)) the dynamics near the separatrix $k = 1$, $H_c = -g\mu_b H_0$ is chaotic and unpredictable. Consequently, the solutions (9) have no meaning near the separatrix. At the same time far from the separatrix $H \neq H_S$, $\Sigma \neq 1$, $k \neq 1$ they are valid. We note that the expression (20) is valid for a low frequency perturbation $\omega_0 \ll D$ and for a high frequency perturbation $\omega_0 \geqslant D$ as well. For estimation of the width of the stochastic layer K_0 the variable of action should be determined. Taking into account (4) we find

$$I^{\pm}(\Sigma) = \oint\left[\frac{1}{2\lambda^2}\left(2\lambda\Sigma - \cos^2\varphi \pm 2\lambda\cos\varphi\sqrt{1 + \frac{1}{4\lambda^2}\cos^2\varphi}\right)\right]^{1/2}\mathrm{d}\varphi. \tag{21}$$

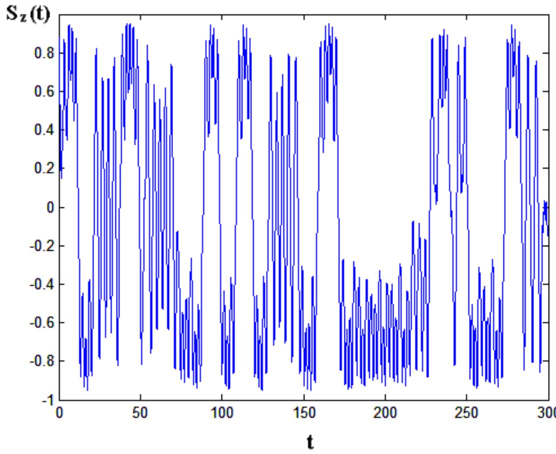

Figure 2. Chaotic motion near the separatrix ($k = 1$, $\Sigma = \Sigma_S = 1$), $D = 90$ GHz. Time independent field H_0, is chosen such that $\lambda = \frac{2DS}{g\mu_B H_0} = 4$, and the ratio between the time independent and variable fields is $\varepsilon = H_1/H_0 = 0.3$. The initial energy $H = -4.5 \times 10^3$ GHz is 8/9 of the re-scaled barrier height $E_B' = DS^2(1 - \frac{1}{\lambda})^2$. Frequency of the variable field is $\omega_0 = 5$. One observes that the orientation of the magnetization is changing in time chaotically.

If the static magnetic field is weak then $\lambda = \frac{2DS}{g\mu_B H_0} \gg 1$ is a large parameter. Therefore, terms of the order λ^{-2} can be neglected. Retaining $1/\lambda$ terms we find from (21) the expression

$$I(\Sigma) = I^+(\Sigma > 1) = I^-(\Sigma > 1) = 2\sqrt{\frac{\Sigma+1}{\lambda}}E\left(\frac{2}{\Sigma+1}\right), \tag{22}$$

where $E(k)$ is the complete elliptic integral of the second kind. Taking into account (22) the expression for the width of the stochastic layer acquires the following form:

$$K_0 \approx \frac{\pi\varepsilon\omega_0}{\sqrt{\lambda(\Sigma+1)}|\Sigma-1|}K\left(\frac{2}{\Sigma+1}\right)E\left(\frac{2}{\Sigma+1}\right). \tag{23}$$

Condition $K_0 > 1$ of the emergence of stochasticity imposes certain restrictions on the parameters of the magnetic field ε, ω, H_0 and on the initial energy Σ of the system. When the energy approaches the separatrix value $\Sigma \longrightarrow 1$ the condition $K_0 > 1$ becomes valid even for a very small $\varepsilon \ll 1$ perturbation. This testifies the fact that the system near the separatrix is sensitive to small perturbations. The emergence of chaos is proved by numerical calculations as well, see figure 2. As one can see from this plot, the dynamics is not regular. The projection $S_z(t)$ of magnetization changes orientation in a chaotic manner.

However, a chaotic change of orientation is not a reversal to a stationary target state. Under dynamical switching we understand here the transition between the oscillatory and the rotational types of motion. To be more specific let us discuss the geometrical aspects of the motion for the trajectories near the separatrix. Upon applying a static magnetic field, the magnetization precessional motion in our case is markedly different from that in the standard NMR set up: the key issue is that the effective magnetic field $H_{\text{eff}} = (g\mu_B H_0, 0, -DS_z)$, due to the nonlinearity of the system, depends on the values

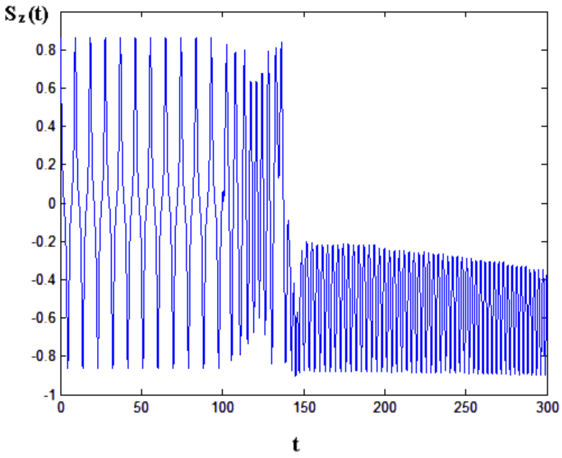

Figure 3. Motion near the separatrix ($k = 1$, $\Sigma = \Sigma_S = 1$), $D = 90$ GHz, $H = -4.5 \times 10^3$ GHz, $\varepsilon = 0.3$, $\lambda = 4$, $\omega_0 = 5$. The variable field is applied during the finite time interval between $\tau_1 = 100$ and $\tau_2 = 150$. Before applying the variable filed, the motion is regular and is of an oscillatory nature. The variable field produces a transition into the rotary regime and then is switched off. During the transition the motion is chaotic.

of S_z. The magnetization vector tends to align as dictated by the effective field. However, the orientation of effective field changes in as much as S_z does. Only in the special case $S_z = 0, \varphi = 0, 2\pi$ which corresponds to the homoclinic points the magnetization vector tends parallel to the effective field $\overrightarrow{M} || \overrightarrow{H_{\text{eff}}}$. On the other hand, the homoclinic point is an unstable equilibrium point. Therefore, the influence of the variable field leads to a switching between the two types of the solutions (9). Hence the following scenario emerges: suppose at the initial time the system is prepared in the degenerated ground state $M_s = S$. We apply a constant magnetic along the x-axis and tune its amplitude to realize the separatrix condition $\Sigma_s = g\mu_b H_0$. A small perturbation can then lead to the transitions. In particular, switching off the perturbation we end up with the transformed state (cf figure 3).

6. Dynamics far from the separatrix: the mean Hamiltonian method

To conclude our study, finally we consider dynamics far from the separatrix. The key point is the fact that stochasticity emerges in the small phase space domain located near the separatrix. Far from the separatrix the dynamics is regular, even in the presence of small perturbations. In this regime, if the frequency of the variable field is high, analytical solutions are found with the help of the mean Hamiltonian method. The basic idea of the mean Hamiltonian method is the following: for a system having different timescales, one averages over the fast variables and obtains thus an explicit expression for the time independent averaged Hamiltonian [26]. In our case, the following condition should then hold:

$$g\mu_B H_0 < D < \omega_0 \left(\frac{2DS}{\lambda} \right), \qquad \varepsilon = H_1/H_0 < 1. \quad (24)$$

This condition implies that the amplitude of the magnetic fields should be small and the frequency should be high. Provided those conditions hold it is possible to average the dynamic over the fast frequency ω_0. The averaged Hamiltonian is determined by the following expression:

$$H_{\text{av}} = \bar{H} + \tfrac{1}{2}\overline{\{\langle \delta H \rangle, H\}} + \tfrac{1}{3}\overline{\{\langle \delta H \rangle, \{\langle \delta H \rangle, H + \tfrac{1}{2}\bar{H}\}\}} + \cdots \quad (25)$$

where $\{A, B\}$ is the Poisson bracket, $\delta H = H - \bar{H}$, $\langle \delta H \rangle = \int \delta H \, \mathrm{d}t$, $\overline{(\ldots)}$ means averaging over the time. Applying the procedure (25) to the Hamiltonian (1) and after straightforward but laborious calculations with the accuracy up to the second order terms $(1/\omega_0)^2$ we find

$$H_{\text{av}} = DS_z^2 + g\mu_b H_0 \sqrt{1 - S_z^2} \cos\varphi + \frac{1}{2} \left[\frac{(g\mu_B H_1)^2}{\omega_0^2} \times \left(-S_z^2 (\cos(\varphi) + \sin(\varphi))^2 + (1 - 2S_z^2) \times (\cos(\varphi) - \sin(\varphi))^2 \right) \right]. \quad (26)$$

The Hamiltonian (26) allows for further simplification: considering that the variable φ is fast in comparison with S_z^2, rotating wave approximation can be used. The Hamiltonian obtained in this way is completely identical to (4). This means, that the solutions (9) are still valid. The difference is that, the constant λ has a different form and depends on the parameters of the variable field

$$\lambda = \left(1 - \left(\frac{H_1 g\mu_B}{2\omega_0} \right)^2 \right) \frac{2DS}{g\mu_b H_0}. \quad (27)$$

By comparing the analytical solutions with the results of the numerical integration of the system of equations (3) far from the separatrix we verify the validity of our approximations.

Figure 4 is for the parameters of the perturbations that are analogous to figure 2. However, unlike figure 2, where the system is near the separatrix $k \approx 1$, in the case of figure 4 $k = 1.6$ which means that the system is far from the separatrix. That is why the dynamics of magnetization is periodic in time. The difference, between figure 4 and the analytical solution (9) is that the amplitude of the oscillations is modulated in time. This observation can be explained with the aid of the average Hamiltonian. The point is that the solutions (9) do not account for the existence of multiple angles in the average Hamiltonian that were ignored by us. They may lead to the appearance of breathing and amplitude modulations.

From the experimental point of view, the systems studied in [13] are suitable to realize the effects predicted here; previous experiments however, were done at too low temperatures where our scheme becomes less reliable and the tunnelling was induced by sweeping a magnetic field. As discussed above our switching scheme is realized differently.

7. Conclusions

We have considered the spin dynamics of a molecular magnet, when the number of the involved levels is large. The dynamics of MNM driven by a variable field has been studied before [22]. However, in contrast to [22], the applied fields in our case are quite strong, i.e. we are in the strongly nonlinear, non-perturbative regime. The underlying dynamics is then treated

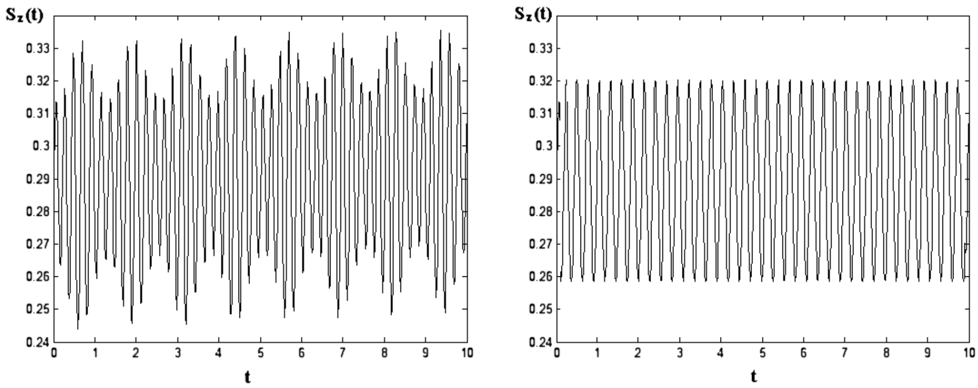

Figure 4. The dynamics far from the separatrix ($k = 1.6$, $\Sigma = 4\Sigma_S$) is regular; $D = 90$ GHz, $H = -0.57 \times 10^3$ GHz, $\varepsilon = 0.3$, $\lambda = 100$, $\omega_0 = 10$. The orientation of the magnetization oscillates with time, however without a change of sign. Dynamically induced switching is not possible far from the separatrix. The left plot corresponds to the numerical solution of equation (3). The right side corresponds to the solution (9) $S_z(t) = b\mathrm{dn}[b\lambda/k(t-\alpha), 1/k]$, with re-scaled λ constant (27). The solutions are in a good agreement with each other. The only difference is the absence of amplitude modulation in the analytical approximation.

semi-classically. We showed that the phase space of the system contains two domains separated by a separatrix line. The solutions far from the separatrix correspond to the rotating and the oscillatory regime, while the separatrix solution is non-oscillating and is of a soliton type. The existence of the separatrix is important as the states in the domain near to it are extremely sensitive to small perturbations. Therefore, if a variable field is applied, instead of a soliton type solutions, the spin dynamics turns chaotic and unpredictable. The control parameter is the initial energy of the system. By a proper choice of it each type of the dynamic can be realized. The structure of the system's phase space is directly related to the possible mechanisms of the magnetization reversal. Namely, if the energy is equal to $H_S = \frac{8}{9}E_B'$ of the re-scaled anisotropy barrier $E_B' = DS^2(1-\frac{1}{\lambda})^2$ [16] (the separatrix condition) an external variable field leads to a chaotic change of the magnetization orientation. The switching process is random and with the equal probability 1/2, the system may appear in the new state as well as stay in the old one. The information about initial state is lost. This result is different from the case of weak applied fields [22], where the dynamics shows a long-term memory of the initial state.

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