Dynamic switching of magnetization in driven magnetic molecules

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Abstract

We study the magnetization dynamics of a molecular magnet driven by static and variable magnetic fields within a semiclassical treatment. The underlying analyzes is valid in a regime, when 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 which we utilize for dynamically induced magnetization switching.

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I. INTRODUCTION

Molecular magnets (MM) are molecular structures with a large effective spin ($S$), e.g. for the prototypical MM $Mn_{12}$ acetates \[ S = 10 \]. MM show a number of interesting phenomena that have been in the focus of theoretical and experimental research \[ 1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11 \]. To name but few, as a result of the strong uniaxial anisotropy, MM show a bistable behavior \[ 1 \]; they also exhibit a resonant tunnelling of magnetization \[ 2 \] that shows up as steps in the magnetic hysteresis loops \[ 3, 4, 5 \]. Of special relevance for applications in quantum computing is the large relaxation time of MM \[ 12 \].

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 MM 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 MM 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 \[ 12, 13 \]: 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 $T < 0.6K$ \[ 13 \]. 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. MM will be modelled as in previous studies, e.g. in Refs. \[ 10, 11 \]. 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 fields 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 behavior. 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.

II. 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 MM 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 \[11\]

\[\begin{align*}
\hat{H} &= \hat{H}_0 + \hat{H}_I, \\
\hat{H}_0 &= -D\hat{S}_z^2 + g\mu_B\hat{H}_0\hat{S}_x, \\
\hat{H}_I &= -\frac{1}{2}g\mu_B\hat{H}_1 e^{i\omega_0 t}(\hat{S}_y + \hat{S}_x) + H.c. .
\end{align*}\]

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 \(\mu_B\) is the Bohr magneton. \(\hat{H}_0\) stands for the constant magnetic field amplitude whereas \(\hat{H}_1\), and \(\omega_0\) are the amplitude and the frequency of the rf field. The problem when both fields \(\hat{H}_0\), \(\hat{H}_1\) are time dependent was studied in \[17\]. 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 semi-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 large number of levels are exited, 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 \((S_z, \varphi)\) via the transformation \[ S = \sqrt{1 - S_z^2} \cos \varphi, \quad S_y = \sqrt{1 - S_z^2} \sin \varphi \] and rewrite (1) in the compact form: \[ H = -\frac{\lambda}{2} S_z^2 + \sqrt{1 - S_z^2} \cos \varphi - \varepsilon \sqrt{1 - S_z^2} (\sin \varphi + \cos \varphi) \cos(\omega_0 t). \] \hspace{1cm} (2)

Hereafter, if not otherwise stated the energy and the time scales are set by the constant magnetic field \( H \mapsto \frac{H}{g \mu_B H_0} \), \( t \mapsto \frac{2DS}{\lambda} t \), \( \omega_0 \mapsto \frac{\lambda}{2DS} \omega_0 \). 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

\[
\begin{align*}
\dot{S}_z &= -\frac{\partial H}{\partial \varphi} = \sqrt{1 - S_z^2} \sin \varphi + \varepsilon \sqrt{1 - S_z^2} (\cos \varphi - \sin \varphi) \cos(\omega_0 t), \\
\dot{\varphi} &= \frac{\partial H}{\partial S_z} = -(\lambda + \cos \varphi)S_z + \varepsilon \frac{S_z}{\sqrt{1 - S_z^2}} (\sin \varphi + \cos \varphi) \cos(\omega_0 t).
\end{align*}
\] \hspace{1cm} (3)

These equations are nonlinear. Therefore, the solutions to (3) can be regular or chaotic, depending on the values of the magnetic fields (parameters \( \lambda, \varepsilon \)). 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 Eq.(2) should become linear. However in the language of variables action angle \((S_z, \varphi)\) that is not so trivial. Therefore, we will discuss this question in more details when studying solutions of the autonomous system.

III. 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 \( H = \text{const} = -\Sigma \)

\[ \frac{\lambda}{2} S_z^2 - \sqrt{1 - S_z^2} \cos \varphi = \Sigma \] \hspace{1cm} (4)

and

\[ \dot{S}_z = \sqrt{1 - S_z^2} \sin \varphi, \] \hspace{1cm} (5)

we find

\[ \dot{S}_z^2 + \left[ \frac{\lambda S_z^2}{2} - \Sigma \right]^2 = 1 - S_z^2. \] \hspace{1cm} (6)

Consequently from eq.(6) we infer

\[ \frac{\lambda t}{2} = \int_{S_z(0)}^{S_z(t)} \frac{dS_z}{\sqrt{\left( \frac{2}{\lambda} \right)^2 (1 - S_z^2) - \left[ S_z^2 - \frac{2\Sigma}{\lambda} \right]^2}}. \] \hspace{1cm} (7)
This relation can be rewritten in the form

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

(8)

where $a^2 = \frac{2}{\lambda^2} \left[ \theta^2/2 - (\Sigma \lambda - 1) \right]$, $b^2 = \frac{2}{\lambda^2} \left[ \theta^2/2 + (\Sigma \lambda - 1) \right]$, $\theta^2(\lambda) = 2\sqrt{\lambda^2 - 2\Sigma \lambda + 1}$.

Performing the integration (8) and inverting the result we obtain

$$S_z(t) = \begin{cases} b \, \text{cn}(b\lambda/k)(t - \alpha), & k, \quad 0 < k < 1, \\ b \, \text{dn}(b\lambda/k)(t - \alpha), & 1/k, \quad k > 1. \end{cases}$$

(9)

Here $\text{cn}(\ldots)$ and $\text{dn}(\ldots)$ are the Jacobi periodic functions. The coefficients that enter eq. (9) read

$$k^2 = \frac{1}{2} \left( \frac{b\lambda}{\theta(\lambda)} \right)^2 = \frac{1}{2} \left[ 1 + \frac{(\Sigma \lambda - 1)}{\sqrt{\lambda^2 + 1 - 2\Sigma \lambda}} \right],$$

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

(10)

With $F(\varphi, k) = \int_0^\varphi dq (1 - k^2 \sin^2 1)^{-1/2}$ being the incomplete elliptical integral of the first kind. From eq.(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 eq.(9) the Jacobian elliptic functions $\text{cn}(\varphi, k)$ and $\text{dn}(\varphi, k)$ are periodic in the argument $\varphi$ 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 [18]. The time period of the oscillation of the magnetization $S_z(t)$ is given by

$$T = \begin{cases} \frac{4K(k)}{b\lambda} & \text{for} \quad 0 < k < 1, \\ \frac{2K(1/k)}{b\lambda} & \text{for} \quad k > 1. \end{cases}$$

(11)

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

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

(12)

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

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

(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 eq.(9):

\[ \text{cn}(u, k) \approx \cos(u) + k^2 \sin(u)(u - \frac{1}{2}\sin(2u)), \quad k^2 \ll 1, \]

and

\[ \text{dn}(u, k) \approx 1 - \sin(u)^2/k^2, \quad k^2 \gg 1. \]

The interpretation of those asymptotic solutions is clear. First one corresponds to the case when in the effective magnetic field \( H_{\text{eff}} = (g\mu_BH_0, 0, -DS_z) \), 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.

IV. TOPOLOGICAL PROPERTIES OF SOLUTIONS

As established [19, 20], 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 \( \varphi: S_z(\varphi, \Sigma) \). The phase portrait of the system is shown in Fig. (1): The different phase trajectories correspond to the solutions (9). The phase trajectories corresponding to the solution \( S_z(t) = \text{dn}(\varphi, k), k > 1 \) are open and they describe a rotational motion of the magnetization. Trajectories corresponding to \( S_z(t) = \text{cn}(\varphi, k), k < 1 \) 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 [20] to external perturbations, which signals the onset of chaotic behavior. 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 phase space.
FIG. 1: Two types of phase trajectories of the system separated by the separatrix $k = 1, \Sigma = \Sigma_S$. The open trajectory (solution $S_z(t) = \text{dn}(t, 1/k)$, $k = 1.52$, $\Sigma > \Sigma_S$) corresponds to the rotational regime of motion. The closed trajectory (solution $S_z(t) = \text{cn}(t, k)$, $k = 0.89$, $\Sigma < \Sigma_S$) 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.

V. FORMATION OF A STOCHASTIC LAYER

To determine the width of the stochastic layer we follow Ref. [20]. For details of the formation of the stochastic layer and for the general formalism we refer to the monograph [20]. Here we only present the main findings. We introduce the canonical variable of action $I = \frac{1}{\pi} \oint S_z(\Sigma, \varphi) d\varphi$ and rewrite the driven nonlinear system (2) in the following form:

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

Here $H_0 = \omega(I)I$, $\omega(I) = \left[ \frac{dI(\Sigma)}{d\Sigma} \right]^{-1}$. The trajectories laying far from separatrix of the unperturbed Hamiltonian $H_0$ are not influenced by perturbation. The motion near the homo-clinic points of the separatrix is very slow [20]. 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 $(I, \varphi)$
\[ I = \frac{\partial I}{\partial H_0} \frac{dI}{dt} = -\varepsilon \frac{\partial V}{\omega(I) \partial S_z} \dot{S}_z \cos(\omega_0 t), \quad (15) \]

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

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 \( \int dt \frac{\partial V}{\partial S_z} \dot{S}_z \cos(\omega_0 t) \) only near to the homoclinic points [20] (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:

\[ I = I - \varepsilon \omega(I) \int_{\Delta t} dt \frac{\partial V}{\partial S_z} \dot{S}_z \cos(\omega_0 t), \quad (17) \]

\[ \dot{\varphi} = \varphi + \pi \frac{\omega_0}{\omega(I)}. \quad (18) \]

Here \( \bar{T}, \bar{\varphi}, \) and \( I, \varphi \) are the values of the canonical variables just after and before passing the homoclinic point, \( \Delta t \) 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

\[ \begin{pmatrix} \frac{\partial \bar{T}}{\partial \bar{I}} & \frac{\partial \bar{T}}{\partial \bar{\varphi}} \\ \frac{\partial \bar{\varphi}}{\partial \bar{I}} & \frac{\partial \bar{\varphi}}{\partial \bar{\varphi}} \end{pmatrix}, \quad (19) \]

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

\[ K_0 = \frac{\pi \varepsilon \omega_0}{\omega^2} \left| \frac{d\omega}{dH} \right|, \quad (20) \]

Here \( \varepsilon, \omega_0 \) are the amplitude and the frequency of the perturbation. Note that the expression (20) is general [20] 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 \( \omega_0 \) and the amplitude \( \varepsilon, \) see Eq.(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

\[ D \]
frequency perturbation $\omega_0 \geq D$ 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

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

If the static magnetic field is weak then $\lambda = \frac{2DS}{g\mu_B H_0} \gg 1$ is a large parameter. In this limit, we can simplify expression (21) to obtain

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

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

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

Condition $K_0 > 1$ of the emergence of stochasticity imposes certain restrictions on the parameters of the magnetic field $\varepsilon$, $\omega$, $H_0$ and on the initial energy $\Sigma$ of the system. When the energy approaches the separatrix value $\Sigma \rightarrow 1$ 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 Fig.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 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 $\vec{M}||\vec{H}_{\text{eff}}$. 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
FIG. 2: Chaotic motion near the separatrix \((k = 1, \Sigma = \Sigma_s = 1), D = 90\text{GHz}\). 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 \(H = -4.5 \cdot 10^3\text{GHz}\) is 8/9 of the re-scaled barrier height \(E_B' = DS^2\left(1 - \frac{1}{\lambda}\right)^2\). Frequency of the variable field is \(\omega_0 = 5\). One observes that the orientation of the magnetization is changing in time chaotically.

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. Fig. (3)).

VI. 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 time scales, one averages over the fast variables and obtains thus an explicit expression for the time independent averaged Hamiltonian [21]. In our case, the following condition should then
FIG. 3: Motion near the separatrix \((k = 1, \Sigma = \Sigma_S = 1)\), \(D = 90 GH z, H = -4.5 \cdot 10^3 GH z, \varepsilon = 0.3, \lambda = 4, \omega_0 = 5\). 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.

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

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 \(\omega_0\). The averaged Hamiltonian is determined by the following expression:

\[
H_{av} = \bar{H} + \frac{1}{2} \{\langle \delta H \rangle, H\} + \frac{1}{3} \{\langle \delta H \rangle, \{\langle \delta H \rangle, H + \frac{1}{2} \bar{H}\}\} + \ldots \quad (25)
\]

where \(\{A, B\}\) is the Poisson bracket, \(\delta H = H - \bar{H}, \langle \delta H \rangle = \int \delta H dt, (...)\) 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

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

The Hamiltonian (26) allows for further simplification: Considering that the variable \(\varphi\) is fast in comparison with \(S_z^2\), rotating wave approximation can be used. The Hamiltonian
FIG. 4: The dynamics far from the separatrix \((k = 1.6, \Sigma = 4\Sigma_S)\) is regular; \(D = 90\text{GHz}, H = -0.57 \cdot 10^3\text{GHz}, \varepsilon = 0.3, \lambda = 100, \omega_0 = 10\). 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 eq.(3). The right side corresponds to the solution \(S_z(t) = b\lambda/k(t - \alpha), 1/k\), with re-scaled \(\lambda\) 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.

obtained in this way is completely identical to (4). This means, that the solutions (9) are still valid. The difference is that, the constant \(\lambda\) has a different form and depends on the parameters of the variable field

\[
\lambda = \left(1 - \frac{H_1 g \mu_B}{2\omega_0}\right)^2 \frac{2DS}{g \mu_B H_0}. \tag{27}
\]

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.

Fig.(4) is for the parameters of the perturbations that are analogous to Fig.(2). However, unlike Fig.(2), where the system is near the separatrix \(k \approx 1\), in the case of Fig.(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 Fig.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.

VII. CONCLUSIONS

We have considered the spin dynamics of a molecular magnet, when the number of the involved levels is large. The dynamics of MM driven by variable filed has been studied before [17]. However, in contrast to [17], 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 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

$$E_B' = DS^2(1 - \frac{1}{X})^2$$

[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 [17], where the dynamics shows a long-term memory of the initial state.

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