Stochastic heating of a molecular nanomagnet

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We study the excitation dynamics of a single molecular nanomagnet by static and pulsed magnetic fields. Based on a stability analysis of the classical magnetization dynamics we identify analytically the fields parameters for which the energy is stochastically pumped into the system in which case the magnetization undergoes diffusively and irreversibly a large angle deflection. An approximate analytical expression for the diffusion constant in terms of the fields parameters is given and assessed by full numerical calculations.

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I. INTRODUCTION

In molecular nanomagnets (MNMs) (Ref. 1) such as in Mn_{12} acetates the magnetic core of the molecule is surrounded by organic nonmagnetic ligands that extinguish the inter-MNM exchange interactions. Hence, much of the on-site physical properties are deducible by studying a single MNM, albeit the dipolar interaction is present and is essential for ordering phenomena.[2] Characteristic for MNMs is the relatively large effective spin (e.g., $S=16$ for Mn_{12}) and the magnetic anisotropy.[1] MNMs exhibit a series of phenomena[1] that are relevant for applications in spintronics and quantum information;[3] most notably is the bistability behavior, the resonance tunneling of magnetization[4] and the large spin-relaxation time. The dynamical control and switching of the magnetization via external fields is a key ingredient on the way to utilizing MNM for technological applications. In this context, the role of thermal and environmental effects have been considered.[5,6] For a single MNM (Refs. 4 and 7–13) at very low temperatures the main switching mechanism is the quantum tunneling of magnetization.[8] This is due to the large anisotropy barrier.[14] For an initially excited MNM and driven by external magnetic fields we have shown recently[15] that the phase space of the magnetization has a rich structure containing a separatrix of topologically different domains. Switching occurs at the separatrix as a consequence of a transition between these domains. However, the issue how to inject energy in this nonlinear system, i.e., how to realize the initially excited state near the separatrix, has not been addressed. Starting from the ground state, this question is not answered by resonant fields as the system is nonlinear, i.e., it changes its eigenfrequency as the oscillation amplitude varies.[15] Formally the equations of motion (EOM) for a single molecular magnet resemble the Landau-Lifshitz-(Gilbert) equation without the Gilbert damping. In the present case the negligible damping is an inherent system property and not a shortcoming of theory. This difference is insofar important as without dissipation a precessional switching, e.g., as proposed in Ref. 16, is not achievable. Hence, new switching schemes are needed for MNM that are different from those known for magnetic materials. One scheme proposed recently[15] relies on a stochastic, diffusion-type switching. For this to work, however, the system has to be excited to a desired state (near the separatrix). The question of how to achieve that is still open. In this paper we show that using appropriate polychromatic magnetic pulses we can achieve a stochastic heating of a molecular magnet as appropriate for the stochastic switching. We derive approximate analytical expressions for the field parameters that allow for the stochastic heating and test for our analytical predictions with full numerical calculations.

II. THEORETICAL MODEL

We study a single molecular magnet (MM), e.g., Fe_8 or Mn_{12} acetates and choose the z axis to be along the uniaxial anisotropy direction (easy axis). The MM is subjected to a constant magnetic field with an amplitude H_0, applied along the x axis (hard axis) as well as to a series of magnetic pulses $\mathcal{F}(t)$ that are linearly polarized in the x direction. The Hamiltonian we write as[15]

$$\hat{H} = \hat{H}_0 + \hat{H}_I,$$

$$\hat{H}_0 = -D\hat{S}_z^2 + g\mu_B H_0 \hat{S}_x, \quad \hat{H}_I = g\mu_B \mathcal{F}(t)\hat{S}_x. \tag{1}$$

Here D is the longitudinal anisotropy constant, S_x, S_y, and S_z are the spin operators projections along x, y, and z directions, respectively. g is the Landé factor and μ_B is the Bohr magneton. Since the spin of the molecular nanomagnet is quite large a classical approximation is appropriate. Hence, it is advantageous to introduce the variables (S_z, φ) via the transformation

$$S_x = \sqrt{1-S_z^2}\cos\varphi, \quad S_y = \sqrt{1-S_z^2}\sin\varphi$$

and rewrite Eq. (1) in the compact form[15]

$$H(t) = -\frac{\lambda}{2}S_z^2 + \sqrt{1-S_z^2}\cos\varphi - \sqrt{1-S_z^2}\cos\varphi F(t)$$

$$\lambda = \frac{2DS}{g\mu_B H_0}, \quad F(t) = \frac{\mathcal{F}(t)}{g\mu_B H_0}. \tag{2}$$

Suppose that the applied constant field is weak $\lambda > 1$ and at the initial moment of time the system resides near to the ground state $S_z(t=0) \approx \pm 1$. How to pump efficiently energy into the system such that we reach the excited states near the separatrix, where then a dynamically induced switching is realizable? To answer this question we make a transition to the action-angle variables (I, φ) in which Hamiltonian (2) reads

$$H = H_0(I) + V(I,\varphi)F(t),$$

$$H_0 = \omega(I)I, \quad \omega(I) = \left[\frac{dI(\Sigma)}{d\Sigma}\right]^{-1},$$

$$I(\Sigma) = \frac{1}{\pi}\int S_z(\Sigma,\varphi)d\varphi, \quad \Sigma = -H. \tag{3}$$

The EOM are

$$\dot{I} = -\frac{\partial H}{\partial \varphi} = -\frac{\partial V(I,\varphi)}{\partial \varphi}F(t),$$

$$\dot{\varphi} = \frac{\partial H}{\partial I} = \omega(I) + \frac{\partial V(I,\varphi)}{\partial I}F(t). \tag{4}$$

In absence of the time-dependent perturbation, I is an integral of motion [$\varphi(t)$ is fast variable, however]. For an applied monochromatic field it is not possible to keep in resonance with ω, for ω depends on I and hence it changes in time. A polychromatic field offers a wider range of frequencies that may match the dynamical frequency of the system. To be more concrete let us assume the applied field to be of the form

$$F = \varepsilon_0 T \sum_{n=-\infty}^{\infty} \delta_\tau(t - nT) = \varepsilon_0 \frac{\tau}{T} \sum_{n=-1/\tau}^{1/\tau} \cos\left(\frac{2\pi}{T} nt\right), \tag{5}$$

where τ is the pulse duration, $T > \tau$ is the interval between the pulses, and ε_0 is the pulse strength.

III. STOCHASTIC HEATING

Of particular interest for us is the situation of overlapping resonances which is realized when[17]

$$K' = \varepsilon_0 T I(\Sigma)\frac{d\omega(I)}{dI(\Sigma)} > 1, \tag{6}$$

in which case the dynamics turns chaotic,[17] i.e., the system jumps from one resonance to other in a random way. A key point here is the irreversibility of the dynamics that emerges due to nonlinearity and without any thermal effects nor external random forces. Hence, we expect a "stochastic heating" of a MM subjected to the pulses [Eq. (5)] when the criterion [Eq. (6)] is fulfilled.

Assuming that $S_z(t=0) \approx \mp 1$ and $\lambda > 1$ we find

$$I(\Sigma) = \int S_z(\Sigma,\varphi)d\varphi = \frac{2}{\sqrt{\lambda}}\sqrt{\Sigma - 1}E\left(\frac{2}{\Sigma - 1}\right),$$

$$\omega(I) = \left[\frac{dI(\Sigma)}{d\Sigma}\right]^{-1} = \frac{\sqrt{\lambda}\sqrt{\Sigma - 1}}{K\left(\frac{2}{\Sigma - 1}\right)},$$

$$\frac{d\omega(I)}{dI} = \frac{d\omega(I)}{d\Sigma}\frac{d\Sigma}{dI} = \omega(I)\frac{d\omega(I)}{d\Sigma},$$

$$\frac{d\omega(I)}{d\Sigma} = \frac{\sqrt{\lambda}\sqrt{\Sigma - 1}E\left(\frac{2}{\Sigma - 1}\right)}{2(\Sigma - 3)K^2\left(\frac{2}{\Sigma - 1}\right)}. \tag{7}$$

Thus we infer

$$K' = \varepsilon_0 T \frac{\sqrt{\lambda}(\Sigma - 1)^{3/2}E^2\left(\frac{2}{\Sigma - 1}\right)}{(\Sigma - 3)K^3\left(\frac{2}{\Sigma - 1}\right)} > 1. \tag{8}$$

$E(\frac{2}{\Sigma-1})$, $K(\frac{2}{\Sigma-1})$ are the complete elliptic integrals in the notation of Ref. 18. In the regime of chaotic motion, when Eq. (6) holds, a dynamical description becomes inappropriate. The adequate language for the study of the magnetization dynamics in this case is an approach based, for example, on the Fokker-Planck equation. A Fokker-Planck equation for the distribution function of the action $f(I,t)$ can be setup in a similar way as done in Ref. 19

$$\frac{\partial f(I,t)}{\partial t} = \frac{1}{2}\frac{\partial}{\partial I}D(I)\frac{\partial f(I,t)}{\partial I}, \quad D(I) = \frac{\varepsilon^2 T}{2}\left[1 - \frac{2}{\lambda}I\omega(I)\right].$$

The relevant quantity is the averaged value of the action $\langle I(t,\Sigma)\rangle_f = \overline{I(t)}$. Using the relations

$$\int I\frac{\partial f(I,t)}{\partial t}dI = \frac{1}{2}\int I\frac{\partial}{\partial I}D(I)\frac{\partial f(I,t)}{\partial I}dI, \tag{9}$$

$$\dot{\overline{I}}(t) = -\frac{1}{2}\int D(I)\frac{\partial f(I,t)}{\partial I}dI = \frac{1}{2}\int \frac{\partial D(I)}{\partial I}f(I,t)dI, \tag{10}$$

$$\frac{\partial D(I)}{\partial I} = -\frac{\varepsilon_0^2 T}{2\lambda}\omega(I)\left(1 + I\frac{d\omega(I)}{d\Sigma}\right), \tag{11}$$

we infer that

$$\dot{\overline{I}} = -\frac{\varepsilon_0^2 T}{2\lambda}\omega(\overline{I})\left(1 + \overline{I}\frac{d\omega(\overline{I})}{d\Sigma}\right). \tag{12}$$

We note that I and $\omega(I)$ are functions of Σ and make the approximation that

$$\overline{I}(\Sigma) = I(\overline{\Sigma}), \quad \overline{\omega}(I) = \overline{\omega}[I(\overline{\Sigma})], \tag{13}$$

meaning that correlations between the random variables are neglected. Formally, we can systematically improve on this

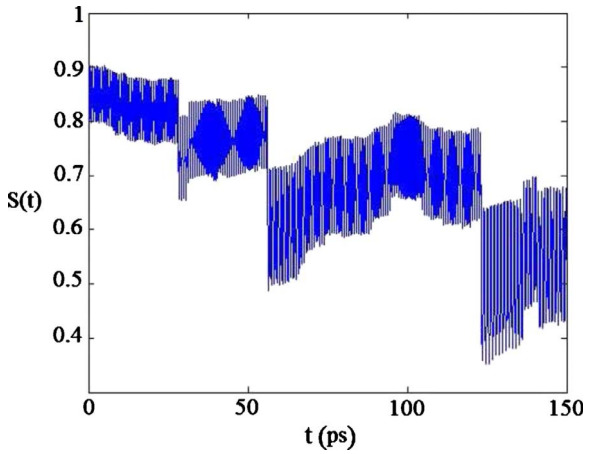

FIG. 1. (Color online) A demonstration of the stochastic deflection of the magnetization vector from the initial state $S_z(0)=0.9$ upon applying a series of rectangular pulses. The results are obtained by solving numerically for the Hamiltonian equations. The time scale is set by values of the constant magnetic field $t \to t/g\mu_B H_0$ (Ref. 15). The parameters are $\varepsilon_0=0.4$, $T=1$ ps, $\tau=0.01$ ps, and $\lambda=10$. Note the criterion of the stochasticity, given by Eq. (8) is realized since $K'=1.75>1$.

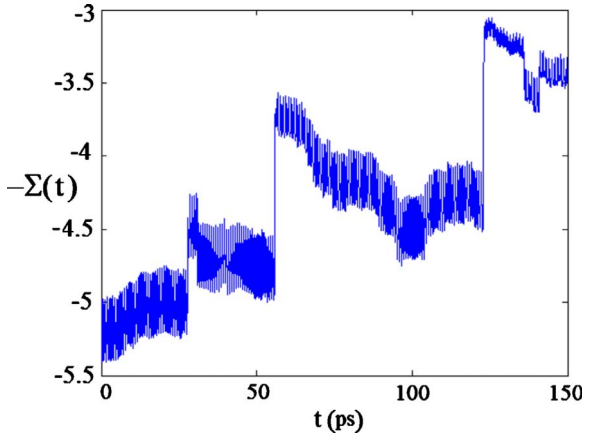

FIG. 2. (Color online) Illustration of the stochastic heating by external driving fields after having started from the initial state $S_z(0)=0.9$. The results are obtained as in Fig. 1 with the parameters $\varepsilon_0=0.4$, $T=1$ ps, $\tau=0.01$ ps, and $\lambda=10$. The criterion of stochasticity in Eq. (8) is fulfilled ($K'=1.75>1$) and a diffusive increase in the system energy is observed. This numerical result is qualitatively consistent with the analytical prediction, given by Eq. (17). For a better numerical agreement one has to go beyond the approximation in Eq. (13) and consider higher moments for the correlation functions of the random variables.

approximation by accounting for higher moments for the correlation functions of the random variables. Neglecting these correlations we find for $\dot{\bar{I}}$

$$\dot{\bar{I}} = \overline{\frac{dI}{d\Sigma}\frac{d\Sigma}{dt}} = -\frac{\varepsilon_0^2 T}{2\lambda}\omega(\bar{I})\left(1+\bar{I}\frac{d\omega(\bar{I})}{d\Sigma}\right), \tag{14}$$

$$\overline{\frac{d\Sigma}{dt}} = -\frac{\varepsilon_0^2 T}{\lambda}\omega^2(\bar{I})\left(1+\bar{I}\frac{d\omega(\bar{I})}{d\Sigma}\right), \tag{15}$$

and we conclude that

$$\frac{d\bar{\Sigma}}{dt} = -\varepsilon_0^2 T\frac{\bar{\Sigma}-1}{K^2\left(\frac{2}{\bar{\Sigma}-1}\right)}\left(1+\frac{\bar{\Sigma}-1}{\bar{\Sigma}-3}\frac{E^2\left(\frac{2}{\bar{\Sigma}-1}\right)}{K^2\left(\frac{2}{\bar{\Sigma}-1}\right)}\right). \tag{16}$$

From the asymptotical solution of $\bar{\Sigma}>1$

$$\bar{\Sigma}(t) = \frac{\lambda}{2}\exp\left[-\frac{4}{\pi}\varepsilon_0^2 T t\right] \tag{17}$$

we uncover a diffusive decay of $\bar{\Sigma}(t)$, meaning that the energy is increased diffusively due to the relation $H=-\bar{\Sigma}(t)$, albeit Eq. (8) must be obeyed.

IV. NUMERICAL RESULTS

The exact numerical results shown in Figs. 1–3 evidence that the mechanism of stochastic heating is indeed present and is quite efficient.

Other scenario for the magnetization control is to employ only the periodic series of rectangular pulses applied along the hard axis, i.e., to switch off the static field. Measuring the energy in units of D we write for the scaled Hamiltonian

$$\bar{H}(t) = H(t)/D = -\frac{S_z^2}{2} + V(s_z,\varphi),$$

where

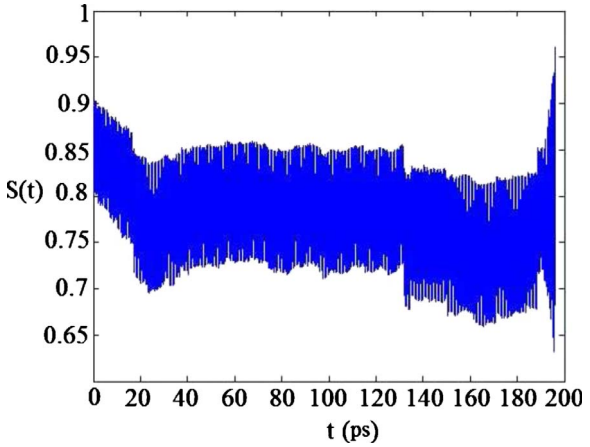

FIG. 3. (Color online) The time evolution of the magnetization vector when starting from the initial state $S_z(0)=0.9$. The results are obtained by solving numerically for the Hamiltonian equations with a series of rectangular pulses. The parameters are $\varepsilon_0=0.1$, $T=1$ ps, $\tau=0.01$ ps, $\lambda=10$, and $K'=0.43<1$. The criterion of stochasticity is not fulfilled. The orientation of the magnetic vector is not changed significantly, and only fluctuations around the ground state is observed.

$$V(s_z,\varphi) = V_0(s_z,\varphi)T\sum_{k=-\infty}^{+\infty}\delta(t-kT)$$

and

$$V_0(s_z,\varphi) = \varepsilon\sqrt{1-s_z^2}\cos\varphi$$

with

$$\varepsilon = \frac{\varepsilon_0}{D}.$$

The equations of motion

$$\dot{\varphi} = \omega(s_z) + \varepsilon\frac{\partial V_0(s_z,\varphi)}{\partial s_z}T\sum_{k=-\infty}^{+\infty}\delta(t-kT),$$

$$\dot{s}_z = -\varepsilon\frac{\partial V_0(s_z,\varphi)}{\partial\varphi}T\sum_{k=-\infty}^{+\infty}\delta(t-kT), \quad \omega(s_z) = -s_z \tag{18}$$

can be integrated exactly in this case by formulating them as recurrence relations[17] using the evolution operator $\hat{T}$ that propagates the system from the time t_0 to T, i.e.,

$$(\overline{s_z},\overline{\varphi}) = \hat{T}(s_z,\varphi), \quad \overline{s_z} = s_z(t_0+T-0),$$

$$\overline{\varphi} = \varphi(t_0+T-0), \quad s_z = s_z(t_0-0), \quad \varphi = \varphi(t_0-0).$$

Note, that $\hat{T}=\hat{T}_R\hat{T}_\delta$ consists of two parts, one describing the free rotations $\hat{T}_R$ and the other, $\hat{T}_\delta$, the action of the applied pulses, i.e.,

$$\hat{T}_R(s_z;\varphi) = [s_z;\varphi+\omega(s_z)T].$$

For $\hat{T}_\delta$ we find upon integrating EOM

$$\hat{T}_\delta(s_z,\varphi) = \left(s_z - \varepsilon\frac{\partial V_0(s_z,\varphi)}{\partial\varphi}, \varphi + \varepsilon T\frac{\partial V_0(s_z,\varphi)}{\partial\varphi}\right).$$

Thus the recurrence relation applies

$$\overline{s_z} = s_z - \varepsilon T\sqrt{1-s_z^2}\sin\varphi,$$

$$\overline{\varphi}_n = \varphi_n - s_z T + \varepsilon T^2\sqrt{1-s_z^2}\sin\varphi + \frac{\varepsilon T s_z}{\sqrt{1-s_z^2}}\cos\varphi. \tag{19}$$

Depending on the chosen parameters, these relations in Eq. (19) may be stable or unstable as signified by the corresponding Lyapunov exponents. Here, we inspect the Jacobean matrix

$$M = \frac{\partial(\overline{s_z},\overline{\varphi})}{\partial(s_z,\varphi)} = \begin{pmatrix} \frac{\partial\overline{s_z}}{\partial s_z} & \frac{\partial\overline{s_z}}{\partial\varphi} \\ \frac{\partial\overline{\varphi}}{\partial s_z} & \frac{\partial\overline{\varphi}}{\partial\varphi} \end{pmatrix}$$

and find for the eigenvalues

$$\lambda_{1,2} = 1 + \frac{1}{2}K \pm \sqrt{\left(1+\frac{1}{2}K\right)^2 - 1},$$

$$K = \varepsilon T^2\frac{\partial^2 V_0}{\partial\varphi^2} = \varepsilon T^2\sqrt{1-s_z^2}\cos\varphi = K_0\sqrt{1-s_z^2}\cos\varphi,$$

where $K_0=\max K=\varepsilon T^2$. Chaos is expected if $\lambda_1>1$, i.e., for $K>0$, meaning even weak pulses $K_0=\varepsilon T^2>0$ may lead to a diffusion. That is, $S_n^z(t)=S^z(t=nT)$ and the magnetization can be deflected diffusively and irreversibly if K_0 exceeds a critical value K_0', as demonstrated by the numerical calculations in Fig. 3. Essential for this phenomena is the existence of two time scales, the slow variables $S_n^z(t)$ and the fast random phase $\varphi(t)$. Using the random-phase approximation for the fast phases[19] one infers the Fokker-Planck equation

$$\frac{\partial f(s_z,t)}{\partial t} = \frac{1}{2}\frac{\partial}{\partial s_z}D(s_z)\frac{\partial f(s_z,t)}{\partial s_z},$$

where

$$D(s_z) = \frac{\pi\varepsilon^2}{\Omega}\sum_{m=-\infty}^{+\infty}m^2|V_m(s_z)|^2\Omega = \frac{2\pi}{T}$$

and $V_m(s_z)$ are the Fourier coefficients as deduced from the expansion

$$V_0(s_z,\varphi) = \sum_{n=-\infty}^{+\infty}V_m(s_z)\exp(in\varphi).$$

Explicitly we have

$$V_0(s_z,\varphi) = -\sqrt{1-s_z^2}\cos\varphi = \frac{\sqrt{1-s_z^2}}{2}(e^{i\varphi}+e^{-i\varphi}).$$

Therefore,

$$V_1 = V_{-1} = -\frac{\sqrt{1-s_z^2}}{2},$$

and the diffusion coefficient is

$$D(s_z) = \frac{\pi\varepsilon^2}{\Omega}(|V_{-1}|^2+|V_1|^2) = \frac{\pi\varepsilon^2}{2\Omega}(1-s_z^2) = \frac{\varepsilon^2 T}{4}(1-s_z^2).$$

Consequently, we write

$$\frac{\partial f(s_z,t)}{\partial t} = D_{diff}\frac{\partial}{\partial s_z}(1-s_z^2)\frac{\partial f(s_z,t)}{\partial s_z},$$

where

$$D_{diff} = \frac{\varepsilon^2 T}{4}$$

is the coefficient of diffusion which is completely defined by the pulse parameters

$$\frac{d}{dt}\langle S_z\rangle = -2D\langle S_z\rangle, \quad \langle S_z\rangle = \int_{-1}^{+1}S_z f(S_z,t)dS_z. \tag{20}$$

Thus, the mean value of the spin projection behaves as

$$\langle S_z(t)\rangle = \langle S_z(0)\rangle e^{-2D_{diff}t}.$$

Since the diffusion coefficient is given by

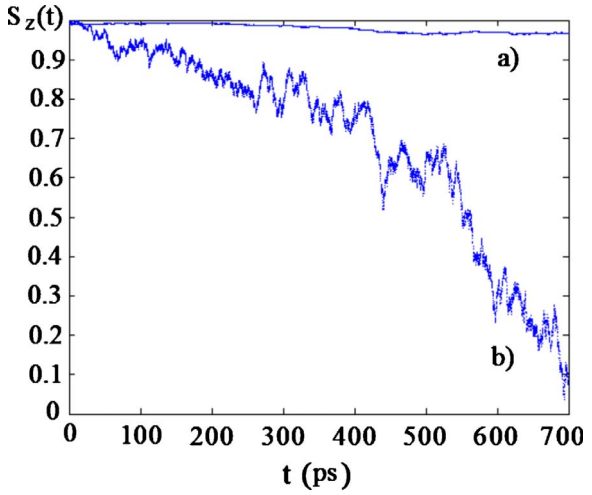

FIG. 4. (Color online) $S_n^z(t)$ as a function of the number of applied kicks $t=nT$ obtained from a full numerical integration of the recurrence relations in Eq. (19). The basic control parameter is the time interval between the pulses T which allows a tuning of the values of stochasticity coefficient [because $D_{diff}=\frac{1}{4}(\frac{\varepsilon_0}{D})^2T$] and hence allows a realization of different types of dynamics. We consider the following initial conditions for Eq. (19): (a) $\varepsilon=0.1$, $S_z(0)=0.99$, $\varphi(0)=0$, $T=0.01$ ps, and $K_0=10^{-5}$. (b) $\varepsilon=0.1$, $S_z(0)=0.99$, $\varphi(0)=0$, $T=0.1$ ps, and $K_0=10^{-3}$.

$$D_{diff}=\frac{\varepsilon^2 T}{4}=\frac{1}{4}\left(\frac{\varepsilon_0}{D}\right)^2 T$$

(D is the magnetic anisotropy constant) D_{diff} can be tuned by changing appropriately the external field parameters, e.g., by varying the amplitude of the pulses ε_0 and/or the interval T between them. Depending on these parameters different types of the dynamics is realized. To test for this analytical prediction we performed full numerical calculations that are in good accord with the analytical results (see Fig. 4). This statement is based on the fact that for $\varepsilon=0.1$, $T=0.1$, $D_{diff}=\varepsilon^2 T=0.001$ the analytically estimated decay rate $1/D_{diff}$ coincides with the numerically deduced one (cf. Fig. 4).

V. SUMMARY

The aim of this work is to point out the possibility of a stochastic energy pumping and magnetization deflection in a single molecular magnet subjected to a static and a time-variable, polychromatic magnetic fields. The key point is that the parameters of the applied static and pulsed magnetic fields can be tuned such that the system is driven nearby a separatrix where the magnetization dynamics turns diffusive allowing thus for a magnetization switching even in the absence of damping (that conventionally originates from coupling to other degrees of freedom).

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