

Statistical mechanics of a nonlinear relaxation process. II. Dynamical properties

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The dynamical properties of a semiclassical model of spin relaxation are carefully studied with the help of computer simulation. It is shown that, as an effect of the reaction field, the relaxation time as a function of temperature ranges from an Arrhenius-like behavior at weak temperatures to a linear power-law behavior at high temperatures. It is also shown that a fairly accurate theoretical prediction of the relaxation time can be obtained in both the low- and high-temperature limit. In the latter regime a simple factorization prescription, although leading to small deviations from the exact equilibrium value for the spin components, is shown to provide the most accurate predictions.

I. INTRODUCTION

The standard theories of spin relaxation lead to a dependence on temperature that is determined by the detailed nature of the system under study but, to the best of our knowledge, never with an Arrhenius-like law [1]. A relevant example of why a relaxation process might have, at least approximately [2], an Arrhenius-like behavior is provided by the dielectric relaxation of water [3,4]. Intuitively, this happens because the hydrogen bonds do not allow free rotations for the single molecule. Hence, to begin rotating, the water molecules must break one or more hydrogen bonds, thus leading to an Arrhenius-like behavior of the dielectric relaxation time. Usually a spin precessing around a magnetic field does not feel the action of space constraints and the relaxation process does not have any potential barrier to overcome. Thus the Arrhenius-like behavior for a spin-relaxation process comes as a surprise. Nevertheless, the Arrhenius-like character for the relaxation process of a spin is an important outcome of a property erroneously neglected for a long time: the reaction field, i.e., the action that the relaxing magnetic dipole has on its own bath.

The standard theories of spin relaxation have almost always disregarded the determinant role of this reaction field. The effects that the reaction field has on the equilibrium properties of the magnetic dipole system have been discussed in the companion paper of Ref. [5], hereafter referred to as I. The model considered in I concerns a magnetic dipole $\mu \equiv (\mu_x, \mu_y, \mu_z)$ precessing around the z axis with a constant Larmor frequency ω_0 under the influence of a fluctuating component of the magnetic field along the x direction. The mathematical structure of this model is

$$\begin{aligned} \dot{\mu}_x &= \omega_0 \mu_y, \\ \dot{\mu}_y &= -\omega_0 \mu_x - \alpha \mu_z x, \\ \dot{\mu}_z &= \alpha x \mu_y, \\ \dot{x} &= -\Gamma x + \Gamma \mu_x + F(t), \end{aligned} \quad (1.1)$$

where $F(t)$ and the damping Γ are the white Gaussian noise and the dissipation strength of the fluctuation-

dissipation process defined by

$$\langle F(0)F(t) \rangle = 2\Gamma \langle x^2 \rangle_{\text{eq}} \delta(t), \quad (1.2)$$

$$\langle x^2 \rangle_{\text{eq}} = \frac{2k_B T}{\alpha}. \quad (1.3)$$

According to the analysis of Ref. [6], this corresponds to the semiclassical approximation of the quantum-mechanical Hamiltonian of a spin- $\frac{1}{2}$ system interacting with a dissipative oscillator. The quantum-mechanical oscillator is replaced by a classical and stochastic oscillator in the overdamped regime. The reaction field is here expressed by the term $\Gamma \mu_x$ on the right-hand side of the last equation of the set of (1.1). The analysis of I was limited to the study of the equilibrium properties of the system.

The present paper deals with the dynamical properties of the model of Eq. (1.1). As in I, we will rely heavily on computer simulations, with no real experimental data being available. The conclusion will be that the spin-relaxation time ranges from a power-law behavior at high temperatures to an Arrhenius-like behavior at low temperatures. The theory used to account for these interesting results is essentially the same as that developed by Grigolini, Wu, and Kenkre [7] and Wu, Grigolini, and Kenkre [8] to study the effect of thermal fluctuations on the dynamics of the linear dimer [7,8].

As already stressed in I, the application of the theory of Refs. [7] and [8] to the problem of a spin relaxation shows that, due to the reaction field, the system exhibits two metastable equilibrium states for the magnetic dipole, very different from the usual (i.e., in the absence of the reaction field) equilibrium state. Then, as an effect of thermal fluctuations, the dipole jumps from one to the other metastable state leading to a relaxation time with an Arrhenius-like dependence on temperature.

The outline of the paper is as follows. Section II is devoted to the illustration of the phenomenology. In Sec. III we present a theoretical interpretation of the simulations in the small-temperature region. In Sec. IV we then turn to the large-temperature region. Finally, concluding remarks are found in Sec. V.

II. PHENOMENOLOGY

As it was done in I, we solve via computer simulation the set of stochastic differential equations of Eq. (1.1). The actual numerical algorithm has already been discussed in I and we should add here only that the number of averages for different parameter values has been changed to ensure a fairly small statistical uncertainty. The quantity that we have taken to characterize the relaxation process is $\mu_x(t)$.

A comprehensive theoretical approach is possible only when Γ is the largest frequency in the system, and we assume that this is always the case. This makes the energy of the system, defined according to the prescriptions of I (see also Sec. III), a very slowly fluctuating variable. We also always worked in a parameter region where the deterministic counterpart of Eq. (1.1) is characterized by the presence of two stable (localized) states (or attractors) for μ_x [see Refs. [7] and [8] and Eq. (3.6) below]. Before we start to analyze in detail the physics of the system, let us first give an idea of the phenomenology one observes in the computer simulations of the model given by Eq. (1.1). Basically, within our hypothesis, we have two different regimes, one for small and one for large temperatures (how small and how large will be quantified below). For small temperatures, $\mu_x(t)$ undergoes fast oscillations

around one of the two possible equilibrium states, and then jumps onto the other one, in a rather random fashion, reminiscent of a standard activation process over a potential barrier. In this physical condition we would be led to assume that the jump takes place only when the slowly changing energy of the system reaches a certain threshold value. In fact, this relatively simple-minded approach seems to be confirmed by the computer simulations: in Fig. 1(b) we have plotted the energy evolution corresponding to the evolution of μ_x in Fig. 1(a), and the time when $\mu_x(t)$ goes through zero roughly coincides with the time when the energy manages to reach values larger than the critical energy (dashed line) for an appreciable time (the dipole does have some inertia). For much larger temperatures, however, the relaxation process seems to be completely different, with the dipole erratically moving all over the available phase space and no significant correlation between the crossover and the actual value of the energy. This kind of motion is shown in Figs. 1(c) and 1(d), where we have plotted $\mu_x(t)$ and the corresponding energy for large temperatures. Clearly, the evolution of $\mu_x(t)$ is very different from the case of small temperatures. In this physical condition μ_x oscillates around $\mu_x=0$, without any significant sojourn time in each of the two "stable" states, and the energy of the system gets to values much larger than the critical one.

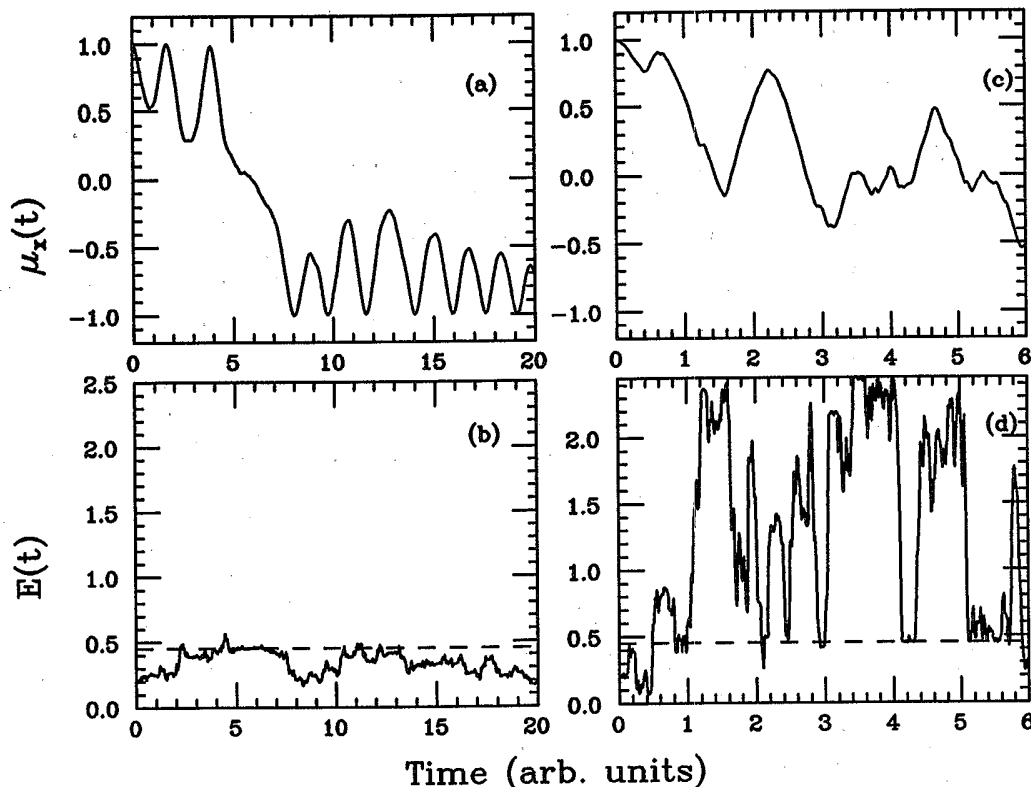


FIG. 1. Plots of $\langle \mu_x(t) \rangle$ [(a) and (c)] and $E(t)$ [(b) and (d)] vs time. Parameters are $\omega_0=2$, $\alpha=5$, $\Gamma=20$, and $T=E_T$ [(a) and (b)] and $T=100E_T$ [(c) and (d)]. In (b) and (d) the dashed horizontal line is the threshold energy E_T .

Taking several trajectories of $\mu_x(t)$ starting with the same initial condition [which we took to be $\mu_x(t=0)=1$], we can then compute the average $\langle \mu_x(t) \rangle$; a typical average is shown in Fig. 2 (solid line). We decided, following a standard procedure in nonequilibrium physics, to define as the nonlinear relaxation time of the system the area under a generic (see below) relaxation curve. For a general process we will define

$$\tau = \frac{\int_0^\infty [\langle c(t) \rangle - \langle c(\infty) \rangle] dt}{\langle c(0) \rangle - \langle c(\infty) \rangle}, \quad (2.1)$$

where τ is the nonlinear relaxation time and $\langle c(t) \rangle$ is a generic process. We took $\langle c(t) \rangle$ to be either the nonequilibrium $\langle \mu_x(t) \rangle$ or the equilibrium autocorrelation function $\langle \mu_x(0)\mu_x(t) \rangle$. It is easier to study, from a theoretical point of view, the nonequilibrium relaxation of $\langle \mu_x(t) \rangle$; however, it would be more fundamental to describe the relaxation process in terms of the autocorrelation function $\langle \mu_x(0)\mu_x(t) \rangle$. It is then very important to establish a relation between quantities computed using the two different prescriptions for $\langle c(t) \rangle$.

First, there is essentially just one relaxation process taking place in the system: hence, apart from short transients, the time over which the various quantities, say, energy or $\mu_x(t)$, evolve, must be related in some ways. For small temperatures, there is indeed a relation between relaxation of $\mu_x(t)$ and time for the energy to reach a certain threshold [see Figs. 1(a), 1(b), and Sec. III]. Other processes, like the nonequilibrium relaxation of $\mu_x(t)$ and the autocorrelation function $\langle \mu_x(0)\mu_x(t) \rangle$, are bound to be strongly coupled. From linear-response theory (LTR) [9] we know that the dynamics in the presence of an external perturbation must be related to some equilibrium correlation functions in the absence of perturbation. A particularly simple case is the free relaxation (i.e., without any external force) of a system where a constant

force has been applied for times $-\infty < t < 0$, force that is suddenly removed at $t=0$. Let us focus on Eq. (1.1), and add a constant term A to the right-hand side of the equation for the evolution of x [the fourth equation in the set of Eqs. (1.1)]. If A is fairly large, the deterministic (i.e., in the absence of noise) equilibrium solution of Eq. (1.1) for the dipole components is given by $\mu_y=0$, $\mu_z \sim 1/A$, $\mu_x \approx 1$, corresponding to our initial conditions. In the presence of A , the initial condition for x would be in general slightly different from the condition we used for our simulations. However, our choice of the other parameters (α and ω_0) helps in recovering initial conditions, for x closer to the ones actually used. Then we remove the perturbation A (the term ‘‘perturbation’’ does not necessarily mean that A is small [10,11]) and monitor the free relaxation of $\mu_x(t)$. From the LRT, however, we know straightaway that

$$\langle \mu_x(t) \rangle_{\text{relax}} \sim \langle x(t)\mu_x(0) \rangle_{\text{equil}}, \quad (2.2)$$

where ‘‘relax’’ and ‘‘equil’’ are meant to remind the reader that we are relating relaxation and equilibrium properties.

In the limit of very large Γ 's, $\mu_x(t) \approx x(t)$, and we arrive to

$$\langle \mu_x(t) \rangle_{\text{relax}} \approx \langle \mu_x(t)\mu_x(0) \rangle_{\text{equil}}, \quad (2.3)$$

which is exactly what we needed: in fact, apart from a factor which is completely irrelevant [it scales out of Eq. (2.1)], we have now that the relaxation of $\langle \mu_x(t) \rangle$ starting from the far-from-equilibrium value $\langle \mu_x(t=0) \rangle = 1$ is proportional to the equilibrium autocorrelation function $\langle \mu_x(0)\mu_x(t) \rangle$. It follows that, at least for large Γ 's, the nonlinear relaxation times computed using the two different prescriptions (equilibrium autocorrelation function and nonequilibrium relaxation from the initial condition) should roughly coincide.

Equation (2.3) is pretty well confirmed by the results of computer simulation (see Fig. 2). The autocorrelation function has been computed using standard fast-Fourier-transform techniques, applying the same baseline correction discussed in [12].

III. THE RELAXATION PROCESS FOR SMALL TEMPERATURES

The leading idea of the theoretical treatment of this section follows closely the arguments used in a recent investigation of the so-called nonlinear dimer [7,8]. As stressed in I, to take into account the influence of the reaction field within a simple second-order treatment of the bath-system interaction, it is necessary to replace the bare Hamiltonian with a renormalized one. At lowest order in the bath-system interaction, the spin precess around the z component of the magnetic field. Within a renormalized picture, on the other hand, the spin precess around a magnetic field that has also a component along the x axis. This x component of the magnetic field, however, depends on the x component of the spin and, according to the work of Refs. [7] and [8] and the earlier work of

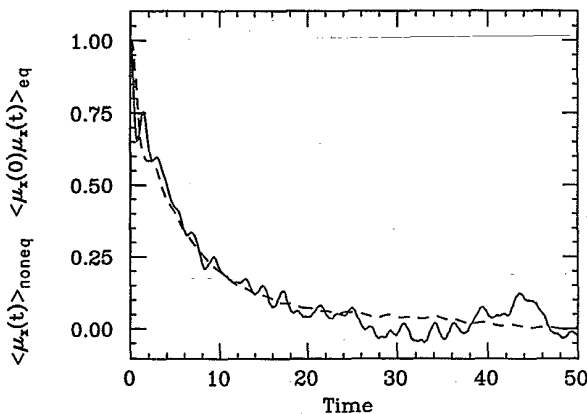


FIG. 2. Comparison between the equilibrium autocorrelation function of $\mu_x(t)$ (dashed line) and the nonequilibrium decay of $\mu_x(t)$ (solid line). The autocorrelation function has been scaled for an easier comparison (also see text). Parameters as in Fig. 1, with $T=E_T$.

Kenkre and co-workers [13], it acts as a potential barrier. Roughly speaking, there are two different magnetic-field directions around which the spin could precess both on the x - z plane, and both corresponding to stable situations in the absence of thermal fluctuations, the only difference being that the average value of the x component of the spin has opposite signs in the two states. The two possible quasiequilibrium directions of the magnetic dipole can be found by looking for the equilibrium solution of Eq. (1.1) in the absence of noise and in the adiabatic limit of infinitely large Γ 's [see also I, and below, Eq. (3.6)].

This zeroth-order approximation is found via the change of variables

$$\tilde{x} = x - \mu_x, \quad (3.1)$$

which yields for Eq. (1.1)

$$\begin{aligned} \dot{\mu}_x &= \omega_0 \mu_y, \\ \dot{\mu}_y &= -\omega_0 \mu_x - \alpha \mu_x \mu_z - \alpha \mu_x \tilde{x}, \\ \dot{\mu}_z &= \alpha \mu_x \mu_y + \alpha \tilde{x} \mu_y, \\ \dot{\tilde{x}} &= -\Gamma \tilde{x} + F(t) - \omega_0 \mu_y. \end{aligned} \quad (3.2)$$

The Fokker-Planck equation associated with Eq. (3.2) has the equilibrium distribution

$$\begin{aligned} \rho_{\text{eq}}(\mu_x, \mu_y, \mu_z, \tilde{x}) &\approx \exp(-H_{\text{ren}}/k_B T) \\ &\quad \times \exp(-\alpha \tilde{x}^2/4k_B T) \\ &\quad \times \delta(\mu_x^2 + \mu_y^2 + \mu_z^2 - 1), \end{aligned} \quad (3.3)$$

where

$$H_{\text{ren}} \equiv \frac{1}{2} \left[\omega_0 \mu_z - \frac{\alpha}{2} \mu_x^2 \right]. \quad (3.4)$$

To solve the problem of the spin-relaxation time in the case of very low temperatures, it is convenient to derive a Fokker-Planck equation for the slow variable E defined as

$$E(\mu_x, \mu_z) \equiv \mu_z - \frac{\alpha}{2\omega_0} \mu_x^2 + \frac{\omega_0}{2\alpha} + \frac{\alpha}{2\omega_0}, \quad (3.5)$$

which is $1/\omega_0$ times the energy corresponding to the renormalized Hamiltonian of Eq. (3.4), plus a constant so that the energy becomes zero when the magnetic dipole is in one of the two possible stable states, i.e.,

$$\begin{aligned} \langle \mu_x \rangle_{\text{eq}} &= \pm \left[1 - \frac{\omega_0^2}{\alpha^2} \right]^{1/2}, \\ \langle \mu_y \rangle_{\text{eq}} &= 0, \\ \langle \mu_z \rangle_{\text{eq}} &= -\frac{\omega_0}{\alpha}. \end{aligned} \quad (3.6)$$

These two stable states become metastable in the presence of thermal fluctuations. Then the spin-relaxation process takes place, more or less, as follows. At short times, the spin relaxes towards one of the two possible equilibrium states, then, due to thermal fluctuations, on a longer scale the spin jumps between the two stable states.

The overall relaxation process is dominated by the time intervals between these jumps.

To determine the sojourn time in one of the two states of Eq. (3.6), we can proceed as follows. Instead of following the trajectories of μ_x , we focus our attention on the energy E defined in Eq. (3.5). When the function $E(\mu_x, \mu_z)$ of Eq. (3.5) gets to the value E_T defined as

$$E_T = \frac{\alpha}{2\omega_0} + \frac{\omega_0}{2\alpha} - 1, \quad (3.7)$$

the system is free to jump into the other stable state. If we looked at Eq. (1.1) without noise and for very large Γ 's, it would be possible to derive an equation of motion for the variable μ_x . According to this equation of motion (basically an oscillator in a nonlinear potential), if $0 < E < E_T$, the evolution of the variable μ_x is confined within two disjoint intervals, $-p_+ < \mu_x < -p_-$ and $p_- < \mu_x < p_+$. In terms of E , one finds that the extrema of these intervals are

$$p_{\pm}(E) = \left[1 - \left[\frac{\omega_0}{\alpha} \right]^2 - \frac{2\omega_0}{\alpha} E \pm \left[\frac{2\omega_0}{\alpha} \right]^{3/2} \sqrt{E} \right]^{1/2}. \quad (3.8)$$

At $E = E_T$, however, we have that $p_- = 0$, which means that the upper limit of the lower interval coincides with the lower limit of the upper interval. Thus the system can cross from one stable state to the other. Note that the energy E can also get to values larger than E_T up to a maximum value given by $E_T + 2$.

Hence, the spin-relaxation time can be derived from the mean first passage time τ to go from $E = 0$ to $E = E_T$. To obtain the explicit expression for τ , we must first build up a Fokker-Planck equation for E . After an adiabatic elimination [7,8], one finds

$$\frac{\partial}{\partial t} P(E, t) = \frac{\partial}{\partial E} [\psi_1(E) P(E, t)] + \frac{\partial^2}{\partial E^2} [\psi_2(E) P(E, t)], \quad (3.9)$$

with

$$\psi_1(E) \equiv \frac{\omega_0 \alpha}{\Gamma} \langle \mu_y^2 \rangle_E + 2k_B T \frac{\alpha}{\Gamma} \langle \mu_z \rangle_E \quad (3.10)$$

and

$$\psi_2(E) \equiv 2k_B T \frac{\alpha}{\Gamma} \langle \mu_y^2 \rangle_E. \quad (3.11)$$

The subscript E on the angular brackets means that the averages are really integrals of the deterministic trajectory at fixed E with the weight $1/\mu_y(E)$ over the interval $\{p_-, p_+\}$.

For the mean value $\langle \mu_z \rangle_E$ we found that if $0 < E < E_T$,

$$\begin{aligned} \langle \mu_z \rangle_E &= E - E_T - 1 \\ &\quad + \frac{\alpha}{2\omega_0} p_+^2(E) \frac{\text{Ei}(1 - p_-^2(E)/p_+^2(E))}{\text{Ki}(1 - p_-^2(E)/p_+^2(E))}, \end{aligned} \quad (3.12)$$

and if $E_T \leq E \leq E_T + 2$,

$$\begin{aligned} \langle \mu_z \rangle_E &= E - E_T - 1 \\ &+ \frac{\alpha}{2\omega_0} \left[p_-^2(E) + [p_+^2(E) - p_-^2(E)] \right. \\ &\quad \times \left. \frac{\text{Ei}(p_+^2(E)/[p_+^2(E) - p_-^2(E)])}{\text{Ki}(p_+^2(E)/[p_+^2(E) - p_-^2(E)])} \right], \end{aligned} \quad (3.13)$$

where $\text{Ki}(m)$ and $\text{Ei}(m)$ are the complete elliptic integrals of the first and second kinds [14].

For the mean value $\langle \mu_y^2 \rangle_E$ we found that if $0 \leq E \leq E_T$,

$$\begin{aligned} \langle \mu_y^2 \rangle_E &= \frac{1}{3} \left[\frac{\alpha}{2\omega_0} \right]^2 p_+^2(E) \\ &\times \left[[p_+^2(E) + p_-^2(E)] \right. \\ &\quad \times \left. \frac{\text{Ei}(1 - p_-^2(E)/p_+^2(E))}{\text{Ki}(1 - p_-^2(E)/p_+^2(E))} - 2p_-^2(E) \right], \end{aligned} \quad (3.14)$$

and if $E_T \leq E \leq E_T + 2$,

$$\begin{aligned} \langle \mu_y^2 \rangle_E &= \frac{1}{3} \left[\frac{\alpha}{2\omega_0} \right]^2 [p_+^2(E) - p_-^2(E)] \\ &\times \left[[p_+^2(E) + p_-^2(E)] \right. \\ &\quad \times \left. \frac{\text{Ei}(p_-^2(E)/[p_+^2(E) - p_-^2(E)])}{\text{Ki}(p_-^2(E)/[p_+^2(E) - p_-^2(E)])} - p_-^2(E) \right]. \end{aligned} \quad (3.15)$$

It can be verified that the stationary solution of the Fokker-Planck equation of Eq. (3.9) coincides with the one derived from Eq. (3.3) using the obvious prescription:

$$\begin{aligned} P_{\text{eq}}(E) &= \int d\mu_x d\mu_y d\mu_z d\tilde{x} \rho_{\text{eq}}(\mu_x, \mu_y, \mu_z, \tilde{x}) \\ &\quad \times \delta \left[E - \left[\mu_z - \frac{\alpha}{2\omega_0} \mu_x^2 + \frac{\omega_0}{2\alpha} + \frac{\alpha}{2\omega_0} \right] \right]. \end{aligned} \quad (3.16)$$

Calculating the integral in Eq. (3.16), we get that [4], if $0 \leq E \leq E_T$,

$$\begin{aligned} \tau &= \frac{3}{2k_B T} \left[\frac{\Gamma}{\alpha} \right] \left[\frac{2\omega_0}{\alpha} \right]^2 \int_0^{E_T} dE \frac{\exp[(\omega_0/2k_B T)E]}{p_+(E) \{ [p_+^2(E) + p_-^2(E)] \text{Ei}(1 - p_-^2(E)/p_+^2(E)) - 2p_-^2(E) \text{Ki}(1 - p_-^2(E)/p_+^2(E)) \}} \\ &\quad \times \int_0^E dE' \frac{\exp[-(\omega_0/2k_B T)E']}{p_+(E')} \text{Ki}(1 - p_-^2(E')/p_+^2(E')). \end{aligned} \quad (3.20)$$

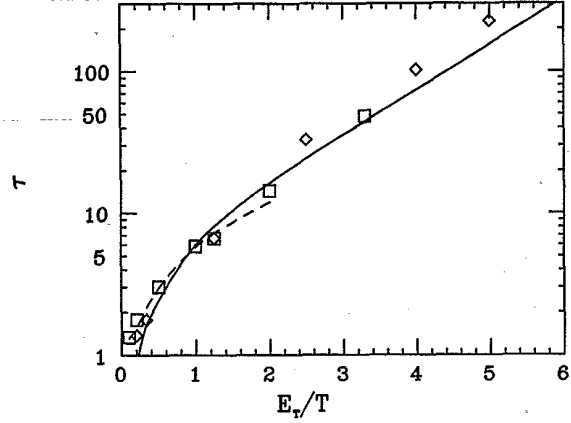


FIG. 3. The nonlinear relaxation time τ vs $1/T$ in units of critical energy E_T . The solid line is the theory from Eq. (3.20), the dashed line is the theory from Eq. (4.2) with $\langle \mu_x^{\text{eq}} \rangle = 0$, and the symbols are the result of numerical simulations [squares, relaxation of the nonequilibrium decay of $\mu_x(t)$; diamonds, relaxation of the autocorrelation function of $\mu_x(t)$]. Parameters as in Fig. 1.

$$P_{\text{eq}}(E) \propto \exp \left[-\frac{\omega_0}{2k_B T} E \right] \frac{\text{Ki}(1 - p_-^2(E)/p_+^2(E))}{p_+(E)}, \quad (3.17)$$

and if $E_T \leq E \leq E_T + 2$,

$$\begin{aligned} P_{\text{eq}}(E) &\propto \exp \left[-\frac{\omega_0}{2k_B T} E \right] \\ &\times \frac{\text{Ki}(p_-^2(E)/[p_+^2(E) - p_-^2(E)])}{\sqrt{p_+^2(E) - p_-^2(E)}}. \end{aligned} \quad (3.18)$$

We are now ready to calculate the relaxation time τ . The mean first passage time for the energy E to reach the value E_T starting from the reflecting point $E = 0$ is [15]

$$\tau = \int_0^{E_T} \frac{dE}{\psi_2(E) P_{\text{eq}}(E)} \int_0^E dE' P_{\text{eq}}(E'). \quad (3.19)$$

Using the analytical expressions for $P_{\text{eq}}(E)$ and $\psi_2(E)$ of Eqs. (3.17) and (3.11), respectively, we finally obtain from Eq. (3.19) the following expression for the spin-relaxation time:

The comparison between Eq. (3.20) and the numerical simulations is presented in Fig. 3: the different symbols refer to the nonlinear relaxation time computed using either the area under the nonlinear relaxation of $\mu_x(t)$ or the area under the autocorrelation function. The solid line is the numerical solution of Eq. (3.20) and the dashed line is what one would obtain factorizing the equations of motion of the dipole moments (see the next section), an approach valid in the large-temperature limit. The agreement between numerical simulations and theory based on the mean first passage time of the energy can be regarded as very satisfactory. We would like to add here that the good agreement up to temperatures of the order of E_T is surprising; in fact, we expected our approach to give reliable results only for temperatures much smaller than E_T .

IV. THE RELAXATION PROCESS FOR LARGE TEMPERATURES

In the region of large temperatures the idea used in the previous section cannot be applied, and a completely different approach must be followed. Working in the limit of large Γ 's, one adiabatically eliminates the motion of the fast variable x , and obtains a reduced Fokker-Planck equation for the evolution of the dipole components. Using this Fokker-Planck operator, it is then possible to show (see [6,7], and I) that the first moments of the components of the spin obey the following set of equations:

$$\begin{aligned} \langle \dot{\mu}_x(t) \rangle &= \omega_0 \langle \mu_y(t) \rangle, \\ \langle \dot{\mu}_y(t) \rangle &= -\omega_0 \langle \mu_x(t) \rangle - \alpha \langle \mu_x(t) \mu_z(t) \rangle \\ &\quad + \alpha \frac{\omega_0}{\Gamma} \langle \mu_y(t) \mu_z(t) \rangle - \frac{2k_B T \alpha}{\Gamma} \langle \mu_y(t) \rangle, \\ \langle \dot{\mu}_z(t) \rangle &= \alpha \langle \mu_x(t) \mu_y(t) \rangle - \alpha \frac{\omega_0}{\Gamma} \langle \mu_y^2(t) \rangle \\ &\quad - \frac{2k_B T \alpha}{\Gamma} \langle \mu_z(t) \rangle. \end{aligned} \quad (4.1)$$

A thorough discussion of the various terms appearing in Eq. (4.1) has already been carried out in I.

These equations (see I) are really the first ones of an infinite hierarchy. To truncate this hierarchy, Kenkre and Grigolini [16] proposed the following approximated set of equations:

$$\begin{aligned} \langle \dot{\mu}_x(t) \rangle &= \omega_0 \langle \mu_y(t) \rangle, \\ \langle \dot{\mu}_y(t) \rangle &= -\omega_0 \langle \mu_x(t) \rangle - \alpha \langle \mu_x(t) \rangle \langle \mu_z(t) \rangle \\ &\quad + \alpha \frac{\omega_0}{\Gamma} \langle \mu_y(t) \rangle \langle \mu_z(t) \rangle - \frac{2k_B T \alpha}{\Gamma} \langle \mu_y(t) \rangle, \\ \langle \dot{\mu}_z(t) \rangle &= \alpha \langle \mu_x(t) \rangle \langle \mu_y(t) \rangle - \alpha \frac{\omega_0}{\Gamma} \langle \mu_y(t) \rangle^2 \\ &\quad - \frac{2k_B T \alpha}{\Gamma} (\langle \mu_z(t) \rangle - \langle \mu_z^{\text{eq}} \rangle), \end{aligned} \quad (4.2)$$

where the equilibrium value of $\langle \mu_z \rangle$ should be calculated from the equilibrium distribution.

We expected these equations to be reliable only in the

large-temperature regime, when the transition between the two attractors takes place without essentially involving a diffusion: a diffusive process would, in fact, undermine the factorization assumption. Operatively, one has simply to compute the equilibrium value of $\langle \mu_z \rangle$ and integrate Eqs. (4.2) starting from the appropriate initial conditions to get the area under $\langle \mu_x(t) \rangle$. In practice, we have found that this procedure does not work very well: the nonlinear relaxation times one derives from the theory agree with the numerical ones only over a very narrow range, and certainly not in the limit of large temperatures; in fact, the disagreement between theory and simulations becomes worse as the temperature is increased.

Looking at the structure of Eqs. (4.1) and (4.2), it is clear why the latter may have problems: remembering that the simulation starts from the initial conditions $\mu = (1, 0, 0)$, we have that the time derivative of $\langle \mu_z \rangle$ in Eqs. (4.1) at time equals zero is zero, whereas, due to the term $\langle \mu_z^{\text{eq}} \rangle$, the same is not true for Eqs. (4.2). The reason for this goes back to the way Eqs. (4.2) were derived: the moments appearing in Eqs. (4.1) were decoupled using their equilibrium values. Consequently, though Eqs. (4.2) may work reasonably well for small fluctuations around equilibrium, they fail in reproducing a far-from-equilibrium process like the one we are studying here; in particular, the short-to-mid time dynamics is fairly slower than in reality, with the result that the relaxation time is grossly overestimated. The situation does not improve when T becomes larger, in fact, in Eqs. (4.2), the quantity $\langle \mu_z^{\text{eq}} \rangle$ decreases only like $1/T$ for large T 's, hence the total contribution to the relaxation of $\langle \mu_z(t) \rangle$ is, to a good approximation, constant at large T .

We have failed to find an exact way to cure this problem. However, if we imposed the condition $\langle \mu_z^{\text{eq}} \rangle = 0$, the consequence would be that the value of $\langle \mu_z^{\text{eq}} \rangle$ one derives from Eqs. (4.2) would be wrong, but, given that for large

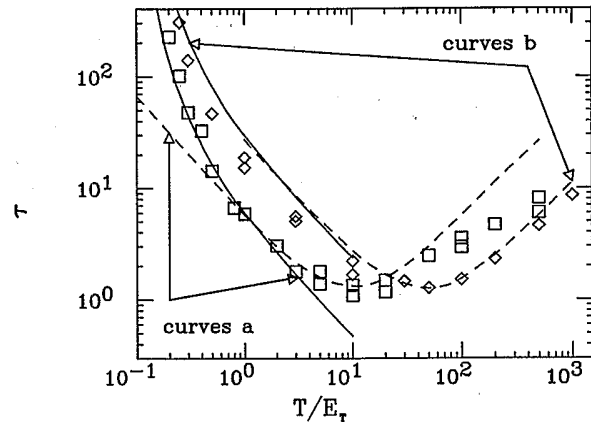


FIG. 4. The nonlinear relaxation time τ vs T in units of E_T . Solid lines are the theory from Eq. (3.20), dashed lines are the theory from Eq. (4.2) with $\langle \mu_z^{\text{eq}} \rangle = 0$, symbols are the result of numerical simulations (squares, $\Gamma = 20$; diamonds, $\Gamma = 100$). Parameter values are $\omega_0 = 2$, $\alpha = 5$, and $\Gamma = 20$ (curves labeled a) and $\Gamma = 100$ (curves labeled b).

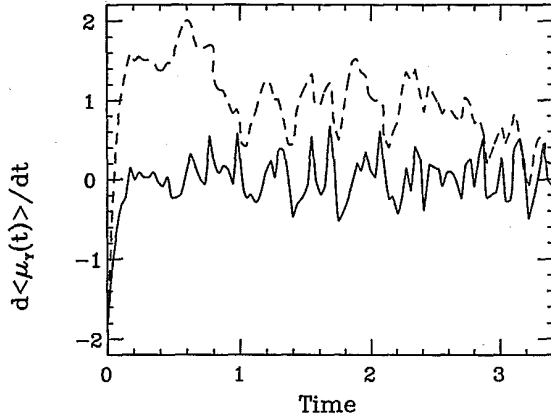


FIG. 5. Comparison between $d\mu_z/dt$ from simulations (solid line) and the right-hand side (dashed line) of the second equation in Eqs. (4.1) vs time for the parameter values of Fig. 1 and for $T=100E_T$.

$T\langle\mu_z^{\text{eq}}\rangle$ goes like $1/T$, clearly the mistake we are making will be minute in the region where we expect Eqs. (4.2) to be reliable. In Fig. 4 we present the comparison between theory [solid lines, Eq. (3.8); dashed lines, Eq. (4.2)] and simulations (symbols): the data are obtained for two different values of Γ , and we used the same symbol for the simulation results carried out using the nonequilibrium relaxation function or the equilibrium autocorrelation function. The results obtained using Eqs. (4.2) with the "right" $\langle\mu_z^{\text{eq}}\rangle$ are not plotted: they would lay parallel to the dashed lines in the large- T region, giving much larger values for the relaxation times. The agreement between theory and simulations is satisfactory at small temperatures, for both values of Γ considered. At large temperatures, however, although the theory is very close to the simulation results for large Γ 's, for the case of $\Gamma=20$ the agreement between theory and simulations is very poor (the actual power law is, in fact, different): the explanation could be that Eqs. (4.1) break down for this value of Γ , or, in other words, that Γ is still too small to carry out an adiabatic elimination. As a check, we have plotted in Fig. 5 the two sides of the evolution equation of $\langle\mu_z\rangle$ in Eqs. (4.1). If the adiabatic elimination were legitimate, we would expect coincidence between the two sides of the $\langle\mu_z\rangle$ equation when we substitute the actual quantities obtained from the simulation to the various cross moments and time derivatives. Despite the statistical noise, it emerges that this is not the case, and hence Eqs. (4.1) cannot be applied to this case, i.e., for this value of Γ , the adiabatic elimination is not appropriate.

The particular structures of Eqs. (4.2), when applicable, do suggest that in the limit of very large temperatures the relaxation time should have a power-law dependence on T . We find that for large T 's,

$$\tau \simeq 2k_B T \alpha / (\Gamma \omega_0^2). \quad (4.3)$$

It is clear from Fig. 4 that there is a linear dependence of τ on T for large T 's, at least for larger Γ 's. We have plotted Eq. (4.3) in Fig. 6 together with the result of our simulations and the agreement is again very good, at least for large Γ 's. Strangely enough, we do observe a power-

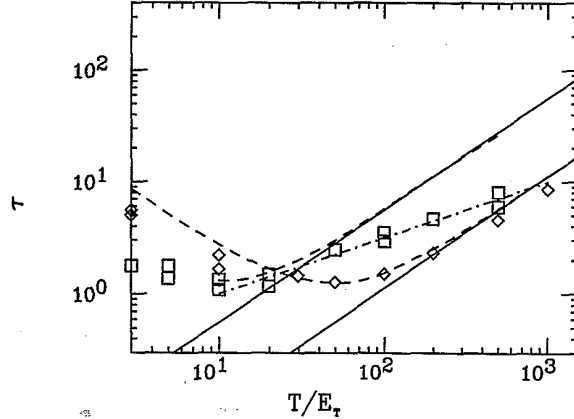


FIG. 6. Relaxation time τ vs T , in units of E_T , for the parameter values of Fig. 4. Dashed lines are the theory from Eq. (4.2) with $\langle\mu_z^{\text{eq}}\rangle=0$; symbols are the result of computer simulations (squares, $\Gamma=20$; diamonds, $\Gamma=100$). The solid lines are the linear power-law behavior predicted by Eq. (4.3), the dotted-dashed line is the best fit to the simulations (squares) with a square-root power law.

law behavior of τ versus T/Γ at large T 's even when Γ is not large enough to allow for an adiabatic elimination, only the power seems to be around $\frac{1}{2}$ rather than 1 (see Fig. 6). We have been unable to explain why this should be so.

V. CONCLUDING REMARKS

We have studied the relaxation properties of a system describing a semiclassical dipole in a fluctuating magnetic field. We have shown that renormalizing the interaction between the dipole and its thermal bath allows us to introduce a Hamiltonian description for the process, thus naturally leading to an interpretation of the relaxation process borrowed from standard canonical systems. In the limit of fast relaxation of the bath we have been able to conclude that the system will exhibit an Arrhenius-like behavior at small temperatures and a power-law behavior at large temperatures. Numerical simulations of the associate stochastic process support very strongly our theoretical conclusions. Finally, though our theoretical approach has largely benefited from the work on the effects of temperature fluctuations on the so-called nonlinear dimer [7], significant advances have been made on high-temperature dynamics, leading to the conclusion that the mere factorization prescription, which is a natural outcome of the microscopic derivation of Ref. [17], provides a satisfactory prediction of the dynamical problem, in spite of the inaccuracy on the equilibrium properties.

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