Statistical mechanics of a nonlinear relaxation process: Equilibrium properties

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We have studied a spin-$\frac{1}{2}$ system interacting with a classical stochastic oscillator in the overdamped regime, simulating a thermal bath. Our aim is to understand the influence of the reaction field, i.e., the effect of the dipole on its bath, which is usually neglected in the current treatments. We have faced the problem of the role of this reaction field with the help of computer simulation. We found that the trajectories of the dipole x component exhibit behaviors suggesting that, in some range of the parameters of the system, the model should have a bistable character. Moreover, the average equilibrium value of the dipole along the z axis shows a significant deviation from the "weak-coupling predictions" of either classical or quantum statistical mechanics. We have explained all these properties via a renormalization procedure which, in the strong-coupling regime, makes the Hamiltonian of interest very different from the bare Hamiltonian. Among other things, this renormalized Hamiltonian leads to a bimodal distribution for the energy, in excellent agreement with the numerical simulations.

I. INTRODUCTION

The role of the reaction field is the missing key property of current theories of spin relaxation.[1,2] In the past few years, there have been some proposals[3–6] to amend the stochastic Liouville equation (SLE) of Kubo[1,2] by removing its main fault, the fact that the influence of the reaction field is disregarded. The quoted papers have focused on the nonlinear character of the fluctuation-dissipation process necessary to recover the proper equilibrium distribution within a spin relaxation process. The proper equilibrium distribution of the magnetic dipole μ in an external magnetic field along the z direction, is the canonical equilibrium distribution prescribed by the basic principles of statistical mechanics (ω_0 is the Larmor frequency)

$$\rho_{\rm eq}(\mu) \propto \exp\left[-\frac{\mu_z \omega_0}{2k_B T}\right] . \tag{1.1}$$

This equilibrium distribution yields the Langevin function[7]

$$\langle \mu_z(T) \rangle = \mu_0 \left[\frac{1}{\theta} - \coth\theta\right] , \tag{1.2}$$

where μ_0 is the value of the magnetic dipole and θ is defined by

$$\theta \equiv \frac{\mu_0 \omega_0}{2k_B T} . \tag{1.3}$$

Equation (1.2) predicts a monotonic increase of $\langle \mu_z(T) \rangle$ as the absolute temperature T is changed from $\langle \mu_z(T) \rangle = -\mu_0$ at $T=0$ to $\langle \mu_z(T) \rangle = 0$ at $T=\infty$. This is a basic property of paramagnetic substances and a proper theory of dipole relaxation should recover it.

Unfortunately, these theoretical investigations ignored another important effect of the coupling between a system and a bath. This has to do with the renormalization of the Hamiltonian of the system of interest. It has been shown[8] that to properly take into account the reaction field, it is necessary to use a suitable basis set, within which the Hamiltonian of the system of interest appears to be changed into an effective one modified by the coupling with the bath. Grigolini, Kenkre, and Vitali[9] have shown the importance of this aspect within the context of the spin-relaxation theory. A much earlier and notable example of renormalization of the Hamiltonian of interest, to explicitly take into account a strong coupling between system and bath, is Ref. 10, dealing with electrons moving in polar crystals. Incidentally, to the best of our knowledge, the term polaronic transformation, widely used afterwards, has been borrowed from Ref. 10.

In this paper, we plan to look at the experimental effects of the reaction field rather than limit ourselves to a theoretical discussion of it. Unfortunately, it is not easy to infer the important role of the renormalization from real experiments. We expect that in real physical systems, the role of the reaction field might be hidden by many other effects, though in some physical regions it could be fairly evident. However, it is not yet clearly understood which parameter (temperature, medium viscosity, transport coefficients, to name some) would enhance the role of the reaction field itself, making it the dominant relaxation mechanism.

An approach that should help, however, is the semiclassical approximation,[9] within which the problem of spin relaxation can be described in terms of a set of a few nonlinear stochastic differential equations. Nevertheless, even in this approximation, the solution remains a challenging problem (see Ref. 11 for a description of possible theoretical tools). On the other hand, it is relatively simple and straightforward to simulate the relevant stochastic differential equations[12] on a digital computer. Let us say that it would not be the first case in which the use of digital simulation has played a crucial role in settling

problems that have been controversial among different theoretical groups.[11] With respect to a hypothetical experiment, there is lack of reality, but on the other hand, all parameters are under control and can be varied at will. Furthermore, the quality of the data themselves is only limited by statistics. We can also monitor single trajectories of the magnetic dipole, whereas a real experiment can only afford data on the statistical superposition of the trajectories of the whole magnetic spin of the sample under study.

The result of our investigation is very interesting. The relaxation process is different from the predictions of the standard linear theories, and strong deviations from the canonical prediction of the Langevin function are found. The spin relaxation does not take place with the usual damped oscillations, but exhibits a much more complicated behavior. We show that these results can be satisfactorily interpreted using the spin-relaxation theory developed in Ref. 9. The key ingredient of this theory is the Hamiltonian renormalization induced by the reaction field.

The outline of the paper is as follows. In Sec. II we present our model of spin relaxation together with the most relevant data of the digital simulations of its dynamics. In Sec. III, following Ref. 9, we show that a nonlinear fluctuation-dissipation process[3–6] alone cannot explain the experimental data, which are, on the contrary, satisfactorily accounted for by the renormalization of the Hamiltonian of interest. Section IV is devoted to concluding remarks.

II. THE STOCHASTIC NONLINEAR MODEL UNDER STUDY AND THE RESULTS OF DIGITAL SIMULATION

The model we discuss in this paper is that of a dipole $\mu \equiv (\mu_x, \mu_y, \mu_z)$ in a magnetic field whose z component is constant (with an associated Larmor frequency ω_0) and whose x component fluctuates. The mathematical structure of this model is

$$\begin{aligned} \dot{\mu}_x &= \omega_0 \mu_y \ , \\ \dot{\mu}_y &= -\omega_0 \mu_x - \alpha \mu_z x \ , \\ \dot{\mu}_z &= \alpha x \mu_y \ , \\ \dot{x} &= -\Gamma x + \Gamma \mu_x + F(t) \ , \end{aligned} \tag{2.1}$$

where $F(t)$ is a white Gaussian noise and where the damping Γ and the strength of the stochastic force are related via a fluctuation-dissipation process defined by

$$\langle F(0)F(t) \rangle = 2\Gamma \langle x^2 \rangle_{\text{eq}} \delta(t) \ , \tag{2.2}$$

$$\langle x^2 \rangle_{\text{eq}} = \frac{2k_B T}{\alpha} \ . \tag{2.3}$$

According to the analysis of Ref. 9, this corresponds to the semiclassical approximation of a 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. It can be proved that, with a few mild assumptions, this in turn is equivalent to a spin coupled to a linear oscillator interacting with an infinite number of degrees of freedom, which may be taken to simulate a canonical thermal bath under a suitable hypothesis.

The new and relevant aspect is the presence of the reaction field (the term $\Gamma \mu_x$) on the fourth equation of Eq. (2.1). If we disregard this term, the dynamics implied by Eqs. (2.1) is fairly well understood. In the Markovian case (fast relaxation of the coupled oscillator, i.e., large Γ's), the system is qualitatively well described by the Bloch equations [Eqs. (2.1) indeed yield the "classical" Bloch equations in this limit]. The use of the SLE would allow us to extend the investigation to the case when the relaxation of the overdamped oscillator is not very fast. In this case, the relaxation of $\langle \mu_x(t) \rangle$ is thought to be characterized by damped oscillations around the mean (vanishing) value. These damped oscillations have a frequency ω_0 near the Markovian case, whereas in the non-Markovian and highly non-Markovian cases, their frequency would in general change (but such frequency could still be derived, for instance, via the recursion method of Ref. 2).

The digital simulation of Eqs. (2.1) corresponds to a treatment of this problem with no approximations, thereby fully including the reaction field. The resulting relaxation process can be studied evaluating the trajectories starting from a given initial condition. We have assumed that the initial condition is given by $\mu_x(0)=1$, $\mu_y(0)=0$, $\mu_z(0)=0$. In Fig. 1, we have plotted one of these trajectories. The result looks quite different than expected on the basis of standard linear theories. It is possible to see that in the initial part of the motion, the x component of the spin executes fast oscillations of relatively small amplitude around a nonvanishing mean value. Then it jumps into the symmetric state with just the opposite mean value and fluctuates for a while around the new equilibrium position. This behavior is closely reminiscent of a Kramers-like process.[13,14] It is well known that the coordinate of a particle moving in a double-well potential executes stochastic oscillations around the bottom of the potential wells and then, from time to time, makes random jumps into the other well. The resulting trajectory would be quite similar to the initial part of the trajectory illustrated in Fig. 1(a).

This seems to suggest that the spin-relaxation process takes place via a thermally activated process and supports a theoretical interpretation based on the renormalization of the Hamiltonian of interest.[9] However, the experimental trajectories show that the dynamics cannot be simply described by a standard Kramers-like process. From Fig. 1(a), it is clear that the motion soon evolves into something fairly unexpected, with the x component of the dipole executing large amplitude oscillations around the zero mean value. The digital simulation shows that these large amplitude oscillations are quite persistent and, once brought into existence, they continue for virtually unlimited periods of time. Note that these oscillations are real, not a product of the limited precision of our simulations or due to problems with the pseudo-random-number generator. As far as the pre-

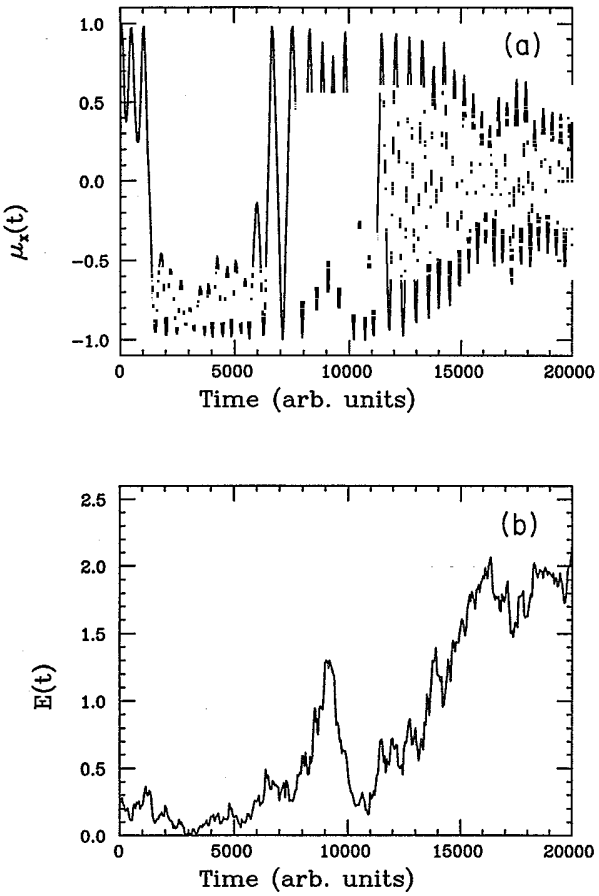


FIG. 1. Time evolution of a single trajectory: (a) $\mu_x(t)$ vs time, (b) E [energy, see Eq. (3.9)] vs time for the same trajectory. The values of the parameters are $\omega_0=0.01$, $\alpha=0.023$, $\Gamma=10$, and $T=0.1E_T=0.0369$ [see Eq. (2.5)].

cision is concerned, we always checked that the norm of the dipole, which is an integral of the motion, would stay constant, which was indeed the case. Changing the initial seed of the random number generator yielded very similar trajectories, though the exact time at which the oscillations started did depend on the seed itself. However, if we started the simulation from an initial condition corresponding to the dipole components at a time after the oscillations had set in, for any chosen seed of the random number generator, an oscillatory regime immediately set in and the different trajectories (corresponding to different realizations of the noise) were very similar. The persistence of these oscillations seems to contradict a Kramers-like picture based on the energy of the dipole. Indeed, if we accepted a Kramers-like picture, the observation that the jumping time between one stable state and the other is much shorter than the waiting time at the bottom of the potential minima would lead to the conclusion that higher-energy states are more probable than lower-energy states. This would be unusual, because one would expect that the energy is canonically distributed.

In Sec. III, we will show that this behavior, seemingly in conflict with the predictions of statistical mechanics, can be easily explained using the theory of Ref. 9. In addition, the seemingly noncanonical behavior arises from the proper zeroth-order Hamiltonian of the system being the renormalized Hamiltonian of Ref. 9:

$$\mathcal{H}\equiv\frac{1}{2}\left[\omega_0\mu_z-\frac{\alpha}{2}\mu_x^2\right] \tag{2.4}$$

rather than the conventional "bare" Hamiltonian

$$\mathcal{H}\equiv\tfrac{1}{2}\omega_0\mu_z\ . \tag{2.4'}$$

Simulations are used to determine the statistical distribution of the energy according to the definition of (2.4). The result is shown in Figs. 2. We see that this deviates from the exponential-like behavior assumed by Refs. 3-6. The peak exhibited by the energy distribution confirms that the trajectories corresponding to higher values of E, as given by Eq. (2.4), can have a statistical weight higher than that of trajectories corresponding to a lower energy. It is interesting to note that the peak appears at

$$E=E_T\equiv\frac{\omega_0}{2\alpha}+\frac{\alpha}{2\omega_0}-1 \tag{2.5}$$

and E_T has a special physical meaning. If we take into account the fact that the three components of the dipole must satisfy the constraint

$$\mu_x^2+\mu_y^2+\mu_z^2=1\ , \tag{2.6}$$

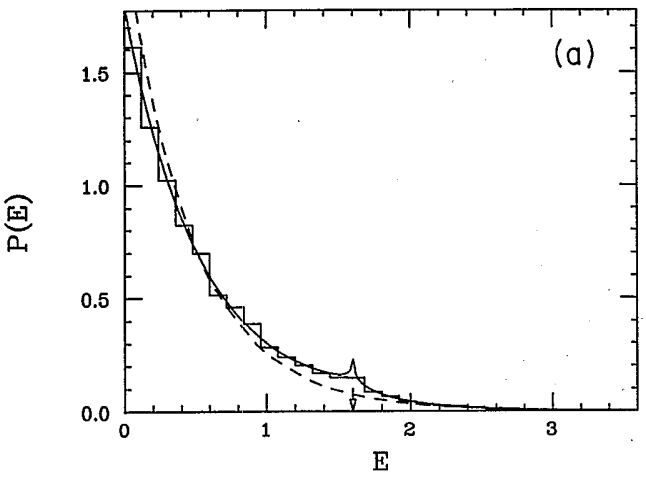
it is easy to show that for $E<E_T$, the range of variation of μ_x is given by two disjoint intervals. This accounts for the bistable nature of this stochastic variable and is reflected in the initial behavior of the trajectory of Fig. 1(a). At $E=E_T$, the two intervals start overlapping and μ_x loses its bistable nature, which explains the appearance of the trajectory oscillations around a null mean value. The persistent nature of these orbits is explained by the presence of the peak itself, which implies that higher-energy states are more probable than lower-energy states.

The bimodal energy distribution of Fig. 2 somehow is reflected on the dynamics of E. In Fig. 3 we show a single time trajectory illustrating the time behavior of E, which seems to suggest that the variable E has a sort of bistable behavior and executes random jumps between one small and one large value. In other words, we find that not only is the stochastic coordinate $\mu_x(t)$ bistable, as suggested by the theory of Ref. 9, but also the renormalized energy E has, to a certain extent, a bistable character.

We have also checked the prediction of the Langevin theory. The result is a significant deviation from the monotonic behavior predicted by the Langevin curve for paramagnetic substances (see Fig. 4).

A true spin-relaxation process implies that an average is done over a very large number of dipoles. Thus to simulate the result of a real experiment, we have also determined the mean trajectory of μ_x starting with the initial conditions characterized by the dipole polarized along the x axis. The result of the simulations is shown in Fig. 5.

We are taught, by the theory of thermally activated



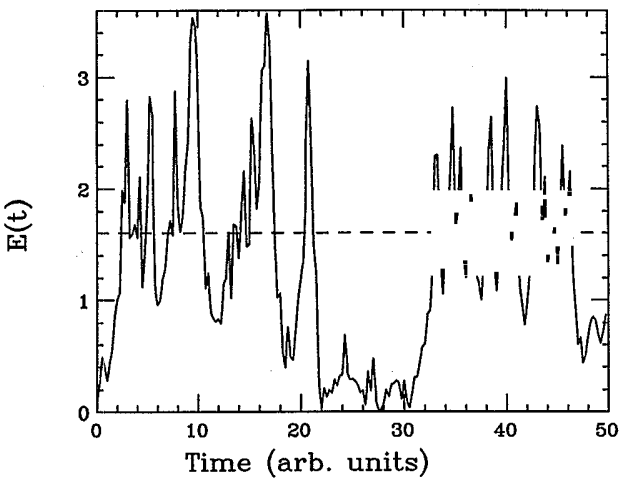
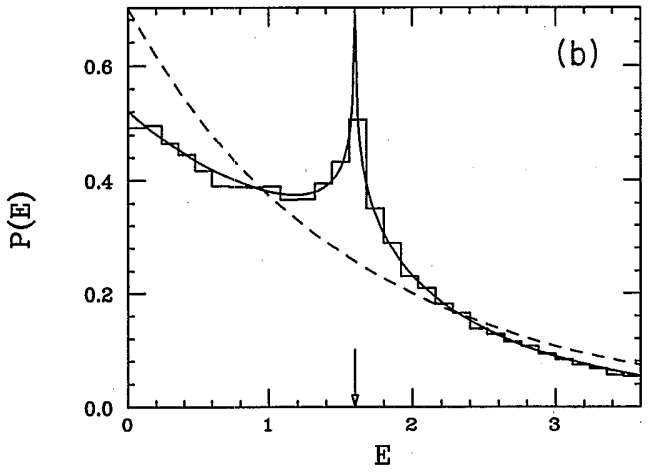


FIG. 3. Time evolution of the energy E vs time for the same parameter values of Fig. 2(b).



processes, that if the relaxation process takes place through an escape over a potential barrier, then the relaxation process consists of two subsequent steps. At the beginning, over a very short time scale, a sort of quasistationary state is reached within the potential minima. Then the true activation process sets in over a much more extended time scale. This is confirmed by the results of digital simulation. Figure 5 (note that we have here changed the parameters so that the two steps are well distinct) shows that at an early stage, a fast relaxation process takes place, corresponding to the attainment of a nonvanishing mean value of the x component of the dipole. This fact by itself is quite unusual in the field of spin relaxation. Then, with a much slower relaxation process, the final and true equilibrium state is reached.

It is thus confirmed that the relaxation process of the

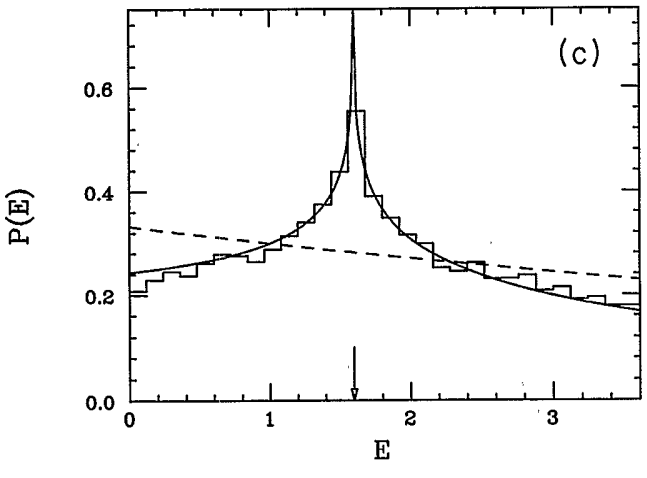


FIG. 2. Probability distribution of the energy E for different values of the temperature T: (a) $T=0.3E_T$, (b) $T=E_T$, and (c) $T=6E_T$. The parameters value are $\omega_0=2$, $\alpha=10$, and $\Gamma=10$, and E_T is defined in Eq. (2.5). The histogram is the result of numerical simulations, the solid line is the theory of Eqs. (3.13) and (3.14), and the dashed line is the result one would obtain using the bare Hamiltonian of Eq. (3.8).

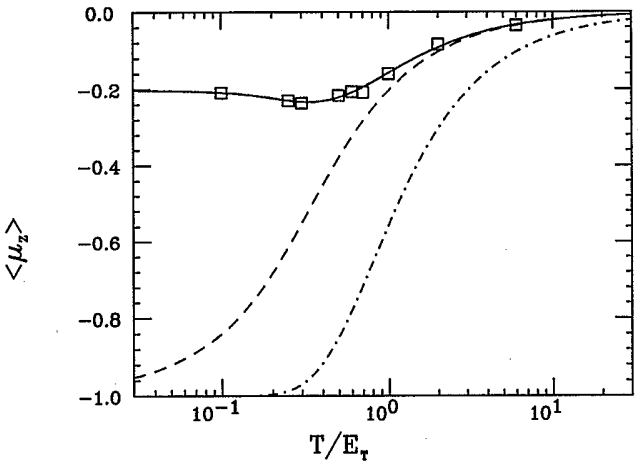


FIG. 4. $\langle \mu_z(T) \rangle$ vs temperature (T). Symbols are digital simulations and the solid line is the theoretical prediction. For comparison, the standard predictions of classical [Langevin function, Eq. (1.2), dashed line] and quantum mechanical (hyperbolic tangent, dash-dotted line) statistical mechanics are also plotted. Parameter values are the same as in Fig. 2.

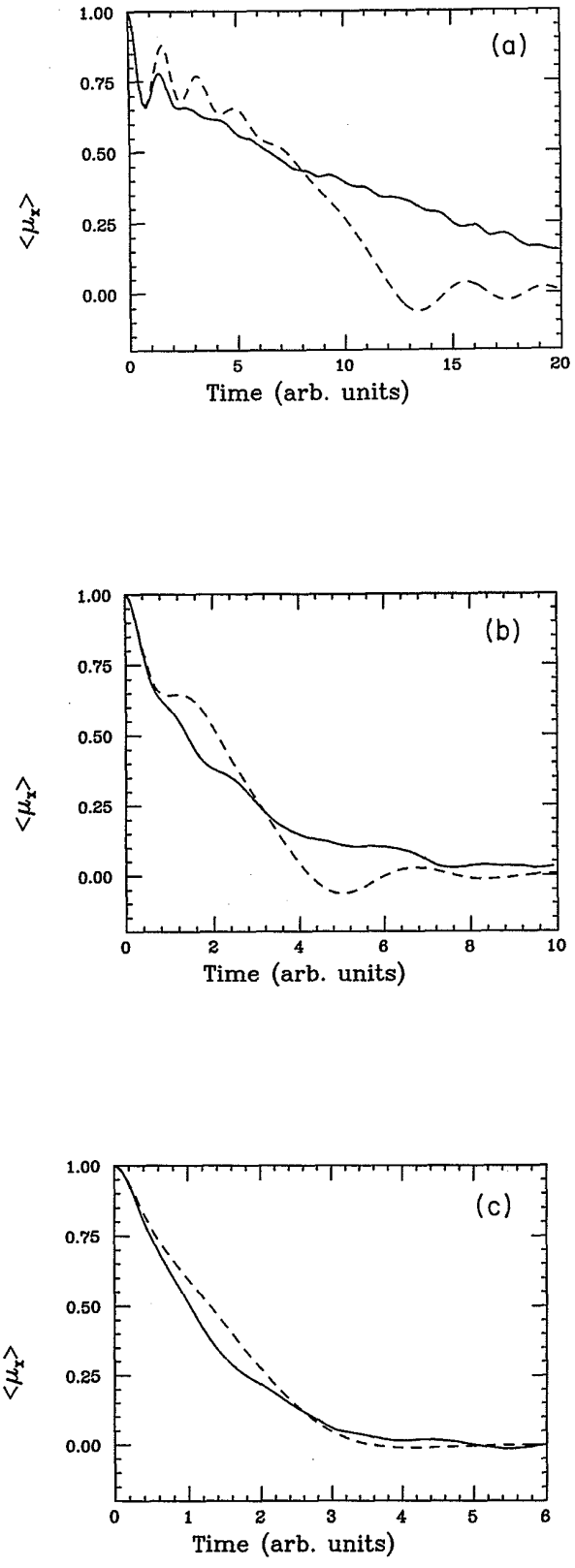


FIG. 5. Mean value of $\mu_x(t)$ vs time (solid line) and corresponding theoretical prediction from the factorized equations [Eq. (3.16), dashed line]: (a) $T=E_T/2$, (b) $T=2E_T$, and (c) $T=6E_T$. The other parameters are the same as in Fig. 2, apart from $\alpha=5$.

system of Eq. (2.1) has new features. These new features cannot be accounted for by the standard theories of relaxation[1,2] and not even by the more refined theories[3–6] more recently developed to take into account the role of the reaction field. It has been shown by several authors[3–6] that this leads to a nonlinear relaxation process and we are going to show that indeed some of the features revealed by the digital simulation can be explained taking this nonlinear relaxation process into account. However, it will be clear that this nonlinear process alone is insufficient to interpret and explain all the observed properties. We should say that to a very large extent, there is a fairly close analogy between our results and the nonlinear dimer studied by Kenkre and coworkers.[15–19] Without this close theoretical connection, the results of our digital simulations, concerned with the problem of spin relaxation rather than that of dimer dynamics, would not have received an easy explanation.

III. THEORETICAL INTERPRETATION

The key ingredients for the theoretical interpretation of the spin-relaxation process can be found in Ref. 9. We limit ourselves to the basic aspects of that theory, in particular, those that will allow us to understand the most interesting results of the digital simulations described in the preceding section. First of all, it is convenient to adopt the following change of variables:

$$\tilde{x}=x-\mu_x \ , \tag{3.1}$$

which transforms the set of Eq. (2.1) as

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

The Fokker-Planck equation for the probability distribution $\pi(\mu_x,\mu_y,\mu_z,\tilde{x};t)$ corresponding to Eqs. (3.2) reads

$$\frac{\partial}{\partial t}\pi(\mu_x,\mu_y,\mu_z\tilde{x};t)=\mathcal{L}\pi(\mu_x,\mu_y,\mu_z,\tilde{x};t) \ , \tag{3.3}$$

where

$$\begin{aligned}
\mathcal{L}\equiv&-\omega_0\mu_y\frac{\partial}{\partial\mu_x}+\omega_0\mu_x\frac{\partial}{\partial\mu_y}+\alpha\mu_x\mu_z\frac{\partial}{\partial\mu_y}-\alpha\mu_x\mu_y\frac{\partial}{\partial\mu_z} \\
&+\alpha\tilde{x}\mu_z\frac{\partial}{\partial\mu_y}-\alpha\tilde{x}\mu_y\frac{\partial}{\partial\mu_z}+\omega_0\mu_y\frac{\partial}{\partial\tilde{x}} \\
&+\Gamma\left[\frac{\partial}{\partial\tilde{x}}\tilde{x}+\langle x^2\rangle_{\rm eq}\frac{\partial^2}{\partial\tilde{x}^2}\right] .
\end{aligned} \tag{3.4}$$

We have considered (μ_x,μ_y,μ_z) as three independent variables, ranging over the whole real axis, and the constraint of Eq. (2.6) as a consequence of the dynamics originated by Eqs. (2.1). Note that Eqs. (3.3) and (3.4) yield the exact evolution for the whole system. This is due to the fact that we have used a standard fluctuation-dissipation process in Eq. (2.2), which makes the corre-

sponding multidimensional Fokker-Planck equation exact. It is straightforward to see that the stationary solution of this Fokker-Planck equation is[9]

$$\pi_{\rm eq}(\mu_x,\mu_y,\mu_z,\tilde{x}) \propto \exp\left[-\frac{1}{2k_BT}\left[\omega_0\mu_z-\frac{\alpha}{2}\mu_x^2+\frac{\alpha}{2}\tilde{x}^2\right]\right] \times\delta(\mu_x^2+\mu_y^2+\mu_z^2-1)\ , \tag{3.5}$$

so that, after integration over the bath variable $\tilde{x}$, we get the correct equilibrium distribution for the dipole components[9]

$$\rho_{\rm eq} \propto \exp\left[-\frac{1}{2k_BT}\left[\omega_0\mu_z-\frac{\alpha}{2}\mu_x^2\right]\right] \times\delta(\mu_x^2+\mu_y^2+\mu_z^2-1)\ . \tag{3.6}$$

This means that the system of interest has a canonical equilibrium distribution with respect to the following renormalized Hamiltonian:

$$\mathcal{H}_{\rm ren}\equiv\frac{1}{2}\left[\omega_0\mu_z-\frac{\alpha}{2}\mu_x^2\right]\ . \tag{3.7}$$

Put in another way, the canonicity of the bath, represented by the standard fluctuation-dissipation process driving the overdamped oscillator in contact with the spin variables, is transmitted to the system of interest. It was also shown in Ref. 9 that the renormalization of the Hamiltonian follows directly from the introduction of the reaction field in Eqs. (2.1), (3.2), and (3.4).

On the basis of Eq. (3.6), we can explain some of the most interesting results of the digital simulation. It is straightforward to derive from Eq. (3.6) the behavior of $\langle\mu_z\rangle_{\rm eq}$ as a function of the absolute temperature T. The result is shown in Fig. 4, where we have compared the theoretical prediction after averaging over Eq. (3.6) with the Langevin function and the prediction of standard quantum-statistical mechanics, in these last two cases using the bare Hamiltonian

$$\mathcal{H}_{\rm unren}\equiv\tfrac{1}{2}\omega_0\mu_z\ . \tag{3.8}$$

The agreement between theory and simulations is very good. The deviation from the monotonic behavior of the Langevin function is due to the nonlinear effects produced by the reaction field. It is interesting to notice that with the parameters used in Fig. 4 the deviation from the classical prediction is much stronger than the discrepancy between the classical and the quantum-mechanical statistical predictions. This might have remarkable consequences for *real* experiments on paramagnetic materials. If the deviation of the Langevin function from the hyperbolic tangent is an indication of the error associated with the semiclassical approximation, then we expect that in real quantum-mechanical paramagnetic systems, an effect of the type illustrated in Fig. 4 might be discovered. In the field of quantum-mechanical dissipation, recently developed theories show that the equilibrium distribution of the system could be forced to deviate significantly from a canonical prediction.[20] According to Kassner,[21] as an effect of the strong coupling between system and bath, the standard rule according to which at zero temperature only the ground state is populated can be violated. Even the excited state of a two-level system can be still populated at $T=0$. We feel that there is some evidence that the spin-relaxation theory of Ref. 9 also incorporates the effect pointed out by Kassner.

From Eq. (3.6) we can also derive the equilibrium distribution of the energy, defined for convenience as

$$E\equiv\mu_z-\frac{\alpha}{2\omega_0}\mu_x^2+\frac{\omega_0}{2\alpha}+\frac{\alpha}{2\omega_0}\ , \tag{3.9}$$

so that its minimum value is zero. We begin expressing the distribution

$$\pi_{\rm eq}(\mu_x,\mu_y,\mu_z)d\mu_x d\mu_y d\mu_z \propto \exp\left[-\frac{H_{\rm ren}}{k_BT}\right]\delta(\mu_x^2+\mu_y^2+\mu_z^2-1)d\mu_x d\mu_y d\mu_z \tag{3.10}$$

in terms of the variables E and μ_x. The transformation from the three components of the spin to E and μ_x, plus the constraint given by Eq. (2.6), leads to a Jacobian weighting factor inversely proportional to μ_y. Finally, we have to integrate over μ_x within the appropriate limits given by the condition

$$\mu_y(E,\mu_x)=0\ . \tag{3.11}$$

The integration must therefore be performed over the range $P_-<\mu_x<P_+$ for values of the energy E less than E_T and over the range $0<\mu_x<P_+$ when the energy E is greater than E_T. P_+ and P_- are given by the following expression:

$$P_\pm=\left[1-\left[\frac{\omega_0}{\alpha}\right]^2-\frac{2\omega_0}{\alpha}E\pm\frac{2\omega_0}{\alpha}\left[\frac{2\omega_0}{\alpha}E\right]^{1/2}\right]^{1/2}\ . \tag{3.12}$$

The resulting equilibrium distribution can be written in terms of the complete elliptic function of the first kind $K(m)$[22] as

$$\rho_{\rm eq}(E) \propto \exp\left[-\frac{\omega_0 E}{2k_BT}\right]I(E)\ , \tag{3.13}$$

where

$$I(E)=\begin{cases}\dfrac{1}{P_+(E)}K\left[1-\left[\dfrac{P_-(E)}{P_+(E)}\right]^2\right], & E<E_T\\[2ex] \dfrac{1}{[P_+^2(E)-P_-^2(E)]^{1/2}}K\left[\dfrac{P_+^2(E)}{P_+^2(E)-P_-^2(E)}\right], & E>E_T\end{cases} \tag{3.14}$$

Note that, for $E>E_T$, P_- becomes imaginary. In this

case, in the corresponding Eq. (3.14), the quantity $-P^2_-$ should be understood as positive. In Fig. 2, the equilibrium distribution of Eq. (3.13) is compared with the results of the numerical simulation and the agreement is remarkable.

We conclude by saying that this result does not conflict with the basic principles of statistical mechanics. We have seen that because the equilibrium distribution is canonically distributed with respect to the renormalized Hamiltonian of Eq. (3.7) rather than the bare Hamiltonian of Eq. (3.8), a conflict with the basic principles of statistical mechanics would be raised by a theory based on the bare Hamiltonian. An observer who is only aware of the bare Hamiltonian would be quite surprised by the simulation results of this paper, which would imply an inexplicable deviation from the canonical behavior. If, on the contrary, we trust the renormalized Hamiltonian of Eq. (3.7) and the corresponding equilibrium distribution of Eq. (3.6), then we conclude that the appearance of a peak at $E=E_T$ is only a consequence of the higher degeneracy of that state.

A detailed study of the dynamical properties of this system will be dealt with elsewhere.[23] Here, let us just give a glimpse of how the relaxation takes place, mainly to roughly explain the result of averaging over trajectories such as the one shown on Fig. 1. Using the Fokker-Planck approach, it is possible to show[9] that the first moments of the components of the spin obey the following set of equations:

$$\begin{aligned}
&\langle\dot{\mu}_x(t)\rangle=\omega_0\langle\mu_y(t)\rangle\ ,\\
&\langle\dot{\mu}_y(t)\rangle=-\omega_0\langle\mu_x(t)\rangle-\alpha\langle\mu_x(t)\mu_z(t)\rangle\\
&\qquad+\alpha\frac{\omega_0}{\Gamma}\langle\mu_y(t)\mu_z(t)\rangle-\frac{2k_BT\alpha}{\Gamma}\langle\mu_y(t)\rangle\ ,\\
&\langle\dot{\mu}_z(t)\rangle=\alpha\langle\mu_x(t)\mu_y(t)\rangle-\alpha\frac{\omega_0}{\Gamma}\langle\mu_y^2(t)\rangle\\
&\qquad-\frac{2k_BT\alpha}{\Gamma}\langle\mu_z(t)\rangle\ .
\end{aligned}\tag{3.15}$$

The linear relaxation process typical of conventional theories of spin relaxation is due to the last term in the second and third equations of this set. The third term on the right-hand side (rhs) of the second equation and the second on the rhs of the third equation (the terms proportional to $\alpha\omega_0$) are the nonlinear dissipation terms discussed by Kubo and Hashitsume,[3] Seshadri and Lindenberg,[4] and Grigolini and Roncaglia.[6] These terms vanish in the adiabatic limit. In addition to these nonlinear terms, we also have to take into account the role of the renormalization of the part of interest, viz., the second term on the rhs of the second equation and the first term on the rhs of the third equation of this set.

Unfortunately, these equations are the first of an infinite hierarchy. This is so because, due to thermal fluctuations, the first moments depend on higher-order moments. To truncate this hierarchy, Kenkre and Grigolini[19] proposed the following approximated set of equations:

$$\begin{aligned}
&\langle\dot{\mu}_x(t)\rangle=\omega_0\langle\mu_y(t)\rangle\ ,\\
&\langle\dot{\mu}_y(t)\rangle=-\omega_0\langle\mu_x(t)\rangle-\alpha\langle\mu_x(t)\rangle\langle\mu_z(t)\rangle\\
&\qquad+\alpha\frac{\omega_0}{\Gamma}\langle\mu_y(t)\rangle\langle\mu_z(t)\rangle-\frac{2k_BT\alpha}{\Gamma}\langle\mu_y(t)\rangle\ ,\\
&\langle\dot{\mu}_z(t)\rangle=\alpha\langle\mu_x(t)\rangle\langle\mu_y(t)\rangle-\alpha\frac{\omega_0}{\Gamma}\langle\mu_y(t)\rangle^2\\
&\qquad-\frac{2k_BT\alpha}{\Gamma}(\langle\mu_z(t)\rangle-\langle\mu_z^{\rm eq}\rangle)\ ,
\end{aligned}\tag{3.16}$$

where the quantity $\langle\mu_z^{\rm eq}\rangle$ should be determined using the equilibrium distribution of Eq. (3.10). Note that for T going to infinity, the conventional (and linear) relaxation process becomes the dominant process and we expect that equilibrium is reached via an exponential-like decay. At lower temperatures, on the other hand, the relaxation towards equilibrium takes place in two distinctive stages. Initially, the system relaxes towards one of the two quasiequilibrium states, characterized by a nonzero value for $\langle\mu_x\rangle$. Subsequently, the system tends to leave this state and relaxes towards the final equilibrium state characterized by $\langle\mu_x\rangle=0$. This is a result of a subtle interplay between thermal fluctuations and nonlinearity, and, unfortunately, within the factorization assumptions leading to Eq. (3.16), we cannot satisfactorily describe the dynamics. This is confirmed by the comparison between the prediction of Eq. (3.16) and the result of digital simulations (see Fig. 5). At relatively small temperatures [Fig. 5(a)], the dipole reaches a quasistationary state, quite distinct from the final equilibrium, with a damped oscillatory behavior pretty well reproduced by Eq. (3.16). The relaxation towards the final equilibrium state, on the other hand, has little to do with the theoretical predictions. However, as the temperature is increased, the agreement between theory and experiment gets better and better, until the whole relaxation process is fairly well reproduced [see Fig. 5(c)]. We should add here that when solving Eqs. (3.16), we have really imposed that $\langle\mu_z^{\rm eq}\rangle\equiv0$, contrary to Ref. 19. This assumption, together with a thorough discussion of the relaxation towards equilibrium of the dynamics implied by Eqs. (3.15) and (3.16) at large temperatures and of the Arrhenius-like behavior at small temperatures will be the subject of future work.[23]

IV. CONCLUDING REMARKS

The digital simulation of the spin-relaxation process of the model of Eq. (2.1) shows that, if the reaction field is fully taken into account and a large-coupling condition is explored, then the behavior of the relaxation process has properties completely different from the conventional weak-coupling theories. We have been able to explain some of the equilibrium properties via the renormalization of the Hamiltonian, which stems from having taken into account the contribution of the reaction field.

The theory of Ref. 9 has strongly benefited from the

research work of Kenkre and co-workers on the nonlinear dimer.[15–19] The interpretation of the results of the digital simulation of this paper, in turn, significantly benefited from the theory of Ref. 9, whose major predictions are proved to be correct. We have discovered a seemingly bistable character of the energy distribution. Further research work is under way to improve our understanding of how the joint action of the renormalization of the Hamiltonian and of the nonlinear fluctuation-dissipation process affect the dynamical properties of this system.

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