Linear response of a nonlinear stochastic oscillator

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A nonlinear oscillator driven by both a coherent external field and a (weak) fluctuation-dissipation process of standard type is studied. It is shown that in the extremely-low-friction regime an additional random process comes into play which makes the linear-response theory work in spite of the scarce amount of standard fluctuation. This additional random process depends on the interplay of the nonlinear character of the oscillator under study and the weak fluctuation-dissipation process of standard type, but it is of transverse type, i.e., it does not involve any energy-dissipation process. On the theoretical side, this result rests on the joint use of a procedure of elimination of "irrelevant" variables and a rotating-wave-approximation technique which makes the role played by the transverse relaxation-process transparent. These predictions are checked by means of an analog experiment. Furthermore, light is shed onto an incorrect conclusion on the range of validity of the linear-response theory arrived at in an earlier report. This incorrect conclusion is now traced back to an ambiguous feature of the experimental results. The frequency position of the absorption peak does not change significantly upon increase of the excitation field up to a certain value of the excitation field. After this initial plateau the frequency of the absorption peak exhibits a rapid increase with increasing the intensity of the radiation field. Then the increase of the linewidth of the absorption peak becomes slower again with a parallel slow increase of the linewidth of the absorption spectrum, the shape of which, furthermore, does not show any sign of saturation, thereby generating the false impression, when the experimental investigation is carried out in this region, of a complete breakdown of the linear-response theory.

I. INTRODUCTION: THE DUFFING OSCILLATOR

The microscopic foundation of the linear-response theory (LRT) (Ref. 1) has been subject of vivacious debates.[2–8] We would like to mention some major contributions. First of all, we must quote the well-known paper of van Kampen.[2] This work points out that the linearity of microscopic motion is entirely different from macroscopic linearity. The macroscopic linearity is the result of a process of statistical linearization provoked by the nonlinear character itself of the microscopic interactions.[9]

The van Kampen criticism stimulated some attempts towards a more careful foundation of the LRT.[3–8] The basic idea of van Vliet[3] is that the LRT should rely either on a master equation approach such as that introduced by van Hove[10] or a projective method like that developed by Zwanzig.[11] This should make a stochastic content appear prior to its application for the calculation of transport coefficients.[3] It is also worth quoting the attempt made by Kenkre[4] to justify the LRT via Zwanzig's projection technique, since the procedure of elimination of irrelevant variables used in this paper is somewhat related to that. Further work has been done, based on the computer simulation of the microscopic processes behind the LRT.[5,6] Finally, we would like to mention the contributions given to the development of the LRT by Oppenheim and his co-workers.[7]

Of special relevance to the subject of the present paper are the arguments in favor of the LRT recently pointed out by Rhodes.[8] By using a model previously developed to study the effects of adiabatic modulation of the spectrally active modes of a molecule by the background modes of the molecule and its medium,[12] Rhodes showed that the statistical effects necessary to make the LRT valid stem naturally from within the LRT itself. This adiabatic modulation acts as a transverse relaxation process, i.e., one resulting in dephasing effects which do not affect the rate of energy dissipation. The role played by the nonlinear interactions in generating precisely a transverse relaxation process will be the major purpose of this paper.

We would like to remark that these arguments fit the results of the investigation on the validity of the equations of the Fokker-Planck type.[13] It can be shown that a microscopic nonlinear Hamiltonian results in the standard Gaussian and Markovian Fokker-Planck equation for the variable of interest under the requirement alone that a large time-scale separation between the variable of interest and its "thermal bath" is available.[13] In other words, the standard Fokker-Planck equation is often a good approximation to the description of macroscopic properties.[13] On the other hand, in the ideal case of a Gaussian system, the LRT would be generally valid regardless of the intensity of the excitation fields (see Appendix A).

It is therefore interesting to study the effect of external excitation on a non-Gaussian model. We shall refer, therefore, our discussion to the model of the Duffing oscillator, which has already been the subject of a paper by our group.[14] This model reads

$$\dot{x}=v\ ,$$
$$\dot{v}=-\omega_0^2 x-\beta x^3-\gamma v+f(t)\ . \tag{1.1}$$

This model can be thought of as a result of a preliminary contraction over infinite freedom degrees so as to produce a standard fluctuation-dissipation process, that according to which the white-Gaussian noise $f(t)$ and the friction parameter γ are related to one another via the relationship

$$\langle f(0)f(t)\rangle_{eq}=2D\delta(t)=2\gamma\langle v^2\rangle_{eq}\delta(t)\ . \tag{1.2}$$

The current research on the rigorous derivation of the fluctuation-dissipation process from a Hamiltonian picture are dependent on the subtle assumption that the system is found in a canonical distribution. This means that the very intimate sources for the appearance of stochastic forces are introduced via an *ad hoc* hypothesis, whereas a widely shared conviction is that the studies on the deterministic transition to chaos should find the true source for the appearance of stochastic properties in the nonlinear nature of the microscopic interaction.[13] According to this interpretation, the standard fluctuation-dissipation process of Eq. (1.2) will contribute to support the LRT without shedding further light into the transition from the nonlinear microscopic world to the macroscopic one. Note, however, that the deterministic drift of Eq. (1.1) is nonlinear. To make more transparent the role of precisely this nonlinear interaction, we shall study this system in an extremely underdamped regime, where the interplay between the nonlinear interaction and the stochastic force $f(t)$, albeit the latter will be assumed to be very weak, will be shown to produce significant effects.

The theoretical tools used in this paper are precisely the same as those developed in two related papers,[14,15] which do not consider, however, the problem of the validity of the LRT.

We shall study the system of Eq. (1.1) under the influence of an external radiation field. The method used in Ref. 15 to eliminate the freedom degrees of the stochastic oscillator simulating the radiation field will lead us to a diffusion equation with an effective temperature which depends on the energy absorbed per unit of time. The formal expression of this absorption as a function of the frequency of the radiation field is the same as that provided by the LRT. In the linear case this prediction is certainly of unlimited validity (i.e., completely independent of the intensity of the radiation field). However, a further prediction of this diffusion equation, the Boltzmann distribution of energy, even in the linear case requires radiation fields of very weak intensity. In the nonlinear case, on the contrary, the range of validity of this latter prediction is more extended due to the fact that a transverse relaxation mechanism stemming from the interplay of the stochastic force $f(t)$ and the anharmonic interaction, see Eq. (1.1), comes into play.

Reference 16 seems to indicate a breakdown of the LRT in the nonlinear case when the extremely underdamped regime is attained. We shall show that the transverse relaxation mechanism stemming from the joint work of noise and anharmonic interaction makes the LRT valid for a fairly extended interval of the radiation field intensity.

The outline of this paper is as follows. In Sec. II we shall study the Duffing oscillator in a moderately inertial regime. The corresponding calculations are carried out by using two methods of calculation developed in Ref. 15, which in principle, bypass the limitations of the LRT. Section III is devoted to developing an analytical approach to the absorption line shape. We build up a simple equation which allows us to determine quantitatively the range of validity of the LRT in the extremely underdamped limit. The absorption spectrum is evaluated via an average on a distribution of energy, whose "excited" temperature is determined by the intensity of the radiation field. In Sec. IV we check our theoretical prediction via an "experiment" of analog simulation. The concluding remarks of Sec. V, as well as Sec. IV, are in part devoted to amend a preliminary short note of our group[16] from a wrong statement on the range of validity of the LRT, the motivation of which is traced back to some misleading features of the experiment done in Ref. 16. In Appendix A we show that in the Gaussian case the prediction of the LRT on the response to an external perturbation is valid regardless of the intensity of the excitation field. This affords us a straightforward way of extending the LRT to the case of a completely deterministic radiation field.

II. ABSORPTION SPECTRUM IN THE MODERATELY INERTIAL REGIME: THE ROTATING-WAVE APPROXIMATION

The Duffing oscillator in the presence of a radiation field reads

$$\begin{aligned}&\dot{x}=v\ ,\\ &\dot{v}=-\omega_0^2x-\beta x^3-\gamma v+f(t)+\omega_f^2y\ ,\\ &\dot{y}=w\ ,\\ &\dot{w}=-\omega_R^2y-\lambda w+f_R(t)\ .\end{aligned} \tag{2.1}$$

The stochastic force $f_R(t)$ is a white-Gaussian noise defined by

$$\langle f_R(0)f_R(t)\rangle_{eq}=2\lambda\langle w^2\rangle_{eq}\delta(t)\ . \tag{2.2}$$

The stochastic linear oscillator with space coordinate y and velocity w mimics the influence of a noncoherent radiation field, the coherence time duration of which is $\tau_c\equiv 1/\lambda$. A completely deterministic description of the radiation field can be recovered by assuming

$$\begin{aligned}&\lambda=0\ ,\\ &f_R(t)=0\ ,\end{aligned} \tag{2.2'}$$

which is precisely the physical condition adopted in this section. In Ref. 15 this case has been studied without having recourse to the LRT. In this section we use precisely Eq. (3.30) and Eq. (3.42) of Ref. 15, both equations relying on the assumption alone that the linewidth of the absorption curve is much smaller than the dominant oscillation frequency of the anharmonic oscillator (rotating-wave approximation). These two equations allow us to evaluate the total amount of energy $\langle E(\Delta\omega)\rangle$ stored in the system as a function of the detuning parameter $\Delta\omega$ defined by

$$\Delta\omega=\omega_R-\omega_0\ . \tag{2.3}$$

The intensity S of the interaction between system and radiation field is defined by

$$S=(S_1^2+S_2^2)^{1/2}\,, \tag{2.4}$$

where

$$S_1=\frac{\omega_I^2 w(0)}{2\omega_R(2k_BT)^{1/2}}\,, \tag{2.5}$$

$$S_2=\frac{\omega_I^2\omega_R y(0)}{2\omega_R(2k_BT)^{1/2}}\,. \tag{2.5'}$$

A further parameter of significant interest is

$$\alpha=\frac{3\beta k_BT}{4\omega_R^3}\,. \tag{2.6}$$

The physical meaning of this parameter has been widely illustrated in Ref. 14. This can be thought of as being the linewidth corresponding to a relaxation process of transverse nature resulting from the interplay of the stochastic force $f(t)$ of Eq. (1.1) and the anharmonic nature of the Duffing oscillator. In Ref. 16 it has been shown that the regime

$$\gamma<\alpha\ll\omega_0 \tag{2.7}$$

is of special interest since the result of analog simulation there reported seems to imply the breakdown of the LRT in this extremely underdamped region. The main purpose of this paper, on the contrary, is to show that when the condition of Eq. (2.7) applies, that associated with α becomes the leading mechanism of relaxation and this is enough to render valid the prediction of the LRT.

Via numerical calculations we found that Eqs. (3.30) and (3.42) of Ref. 15, when applied to the regime of Eq. (2.7), are fraught with severe problems of slow numerical convergence. This obliged us to explore only the more conventional regime described by the condition ($\gamma\ll\omega_0$)

$$\gamma>\alpha\,. \tag{2.8}$$

The comparison with the prediction of the LRT was done by noticing that the LRT relies on the Laplace transform of $\langle v(0)v(t)\rangle_{\rm eq}$, which is the velocity autocorrelation function in the absence of external radiation field ($\omega_I=0$). This Laplace transform, in turn, was evaluated by using the continued-fraction procedure of Ref. 17.

Figure 1 shows that the range of intensities of the interaction with the radiation field which can be studied by using Eqs. (3.30) and (3.42) of Ref. 15 produces absorption line shapes indistinguishable from the prediction of the LRT. Both equations involve problems of numerical convergence for intensities of radiation coupling higher than $S\simeq 0.15$ when $\alpha=0.001$ and $\gamma=0.2$. In the linear case ($\alpha=0$) on the contrary, both methods show a fast convergence to a Lorentzian line shape, regardless of the intensity of the radiation field (this agrees with Appendix A). This result can be understood on the ground of the theory developed in Ref. 15. Let us consider, for instance, the former method of calculation [Eq. (3.30) of Ref. 15]. This consists of expanding the Fokker-Planck operator associated to Eqs. (2.1) and (2.2), and supplemented by the

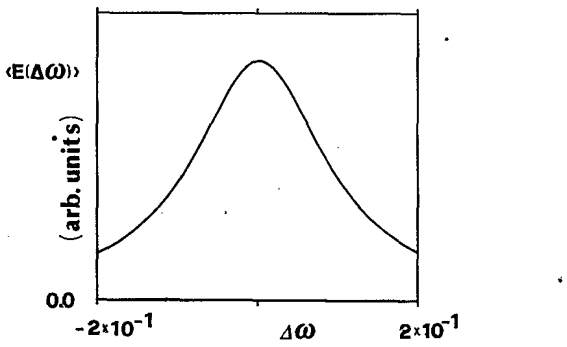

FIG. 1. $\langle E(\Delta\omega)\rangle$, the total amount of energy stored in the system as a function of the detuning parameter $\Delta\omega\equiv\omega_R-\omega_0$, as resulting from the two equivalent formulas of Eqs. (3.30) and (3.42) of Ref. 15, and application of the LRT. In this scale the three results are indistinguishable from each other for S ranging from $S=0$ to $S=0.15$ (with $\gamma=0.2$ and $\alpha=0.001$).

rotating-wave approximation, over a basis set $|nm\rangle=|n\rangle\,|m\rangle$, where $|n\rangle$ and $|m\rangle$ denote the Hermite polynomial concerning two suitable adimensional variables, ξ and η, proportional to x and v, respectively. The observable $\langle E(\Delta\omega)\rangle$ only depends on the expansion coefficients of the equilibrium state over the manifold $n+m=2$. In the case $\alpha=0$ these expansion coefficients only depend on the interaction with the two lower manifolds via the coupling between system and radiation field (and can be evaluated by means of analytical expressions). Thus, provided that the matrix to be diagonalized is given the minimum size 6×6, these coefficients of expansion are completely independent of the size of the expansion basis set. In the nonlinear case, on the contrary, the anharmonic interaction makes the second-order manifold also dependent on the higher-order ones, thereby generating the aforementioned convergence problems. Similar remarks apply also to the latter method of calculation [Eq. (3.42) of Ref. 15].

Note that if the diagonalization procedure were used to determine the excited energy distribution in the presence of strong radiation fields (see also Ref. 18), the linear case would not exhibit any relevant computational advantage compared to the nonlinear one because this observable would depend on manifolds of increasing order with the increase in intensity of the radiation field.

We can conclude this section by saying that the methods of calculation used here do not work when the critical region

$$\alpha\lesssim\gamma \tag{2.9}$$

is reached. Therefore, in principle, it is not possible to assess whether the LRT holds in this critically underdamped region, which has been explored before only by using the analog simulation method.[16] Note that Ref. 15 explored this critical region by making the basic assumption that the LRT holds. We plan to settle this basic issue by a joint use of the theory and analog simulation (the latter having been widely improved compared to Ref. 16). This will be the major purpose of the next two sections.

III. ABSORPTION SPECTRUM IN THE EXTREMELY LOW-FRICTION REGIME

In two previous papers,[14,15] it has been assessed that, with decreasing γ while keeping the other parameters unchanged, a region is met at about

$$\gamma \sim \alpha ,$$

where the standard procedure of statistical linearization[19] is completely invalidated. A preliminary experiment of analog simulation[19] led us to believe that this region is also characterized by a complete breakdown of the LRT. A subsequent more careful investigation[14] made it clear that in this new regime the leading mechanism for producing chaos is no longer that associated with the standard fluctuation-dissipation process of Eq. (1.2): This is replaced by a frequency modulation process, reminiscent of those pointed out by Rhodes[8,12] within the context of his picture of stochastic modulation in the radiative decay of large molecules.

We are now in a position to show by analytical arguments that this transverse relaxation process, contrarily to our previous point of view,[16] allows the LRT to be fulfilled even in the extremely underdamped regime. This will be the major purpose of the present section. In Sec. IV we shall check these predictions with a new, much more accurate experiment of analog simulation.

The Fokker-Planck equation associated with the system of Eq. (2.1), can be written as follows:

$$\frac{\partial}{\partial t}\rho(x,v,y,w;t)=(\mathscr{L}_a+\mathscr{L}_b+\mathscr{L}_1)\rho(x,v,y,w;t) , \tag{3.1}$$

where $[V'\equiv(\partial/\partial x)V]$

$$\mathscr{L}_a\equiv -x\frac{\partial}{\partial v}+V'\frac{\partial}{\partial v}+\gamma\left[\frac{\partial}{\partial v}v+k_BT\frac{\partial^2}{\partial v^2}\right] , \tag{3.2}$$

$$\mathscr{L}_b\equiv -w\frac{\partial}{\partial y}+\omega_R^2 y\frac{\partial}{\partial V}+\lambda\left[\frac{\partial}{\partial w}V+\langle w^2\rangle_{eq}\frac{\partial^2}{\partial w^2}\right] , \tag{3.3}$$

$$\mathscr{L}_1\equiv\omega_I^2 y\frac{\partial}{\partial v} . \tag{3.4}$$

In this section we shall study the limit $\lambda\to 0$ of the stochastic description of the radiation field. In Appendix B it is shown that this coincides with the purely deterministic description $\lambda=0, f(t)=0$ ($\langle w^2\rangle_{eq}=0$).

The first step of the approach of elimination of irrelevant variables developed by Grigolini[20] consists in writing Eq. (3.1) in the interaction picture, thereby leading to

$$\frac{\partial}{\partial t}\tilde{\rho}(x,v,y,w;t)=\mathscr{L}_1(t)\tilde{\rho}(x,v,y,w;t) , \tag{3.5}$$

where

$$\tilde{\rho}(x,v,y,w;t)=e^{\mathscr{L}_0 t}\tilde{\rho}(x,v,y,w;t) , \tag{3.6}$$

$$\mathscr{L}_1(t)=e^{-\mathscr{L}_0 t}\mathscr{L}_1 e^{\mathscr{L}_0 t} , \tag{3.7}$$

$$\mathscr{L}_0=\mathscr{L}_a+\mathscr{L}_b . \tag{3.8}$$

The second step consists in applying to Eq. (3.5) the projection operator method of Zwanzig[11] with the projection operator P defined by ($a\equiv(x,v)$; $b\equiv(y,w)$)

$$P\rho(a,b;t)=\rho_{eq}(b)\sigma(a;t)\equiv\rho_{eq}(b)\int db\,\rho(a,b;t) , \tag{3.9}$$

$$\mathscr{L}_b\rho_{eq}(b)=0 , \tag{3.10}$$

$$\int db\,\rho_{eq}(b)=1 . \tag{3.11}$$

We thus obtain

$$\frac{\partial}{\partial t}\sigma(a;t)=\mathscr{L}_a\sigma(a;t)+\int_0^t K(t-\tau)\sigma(a;\tau)d\tau$$
$$=\mathscr{L}_a\sigma(a;t)+\int_0^t K(\tau)\sigma(a;t-\tau)d\tau , \tag{3.12}$$

where ($\overleftarrow{\exp}$ denotes[20] a time-ordered exponential)

$$K(t-\tau)=\rho_{eq}^{-1}(b)e^{-\mathscr{L}_a t}P\mathscr{L}_1(t)\overleftarrow{\exp}\left[\int_\tau^t dt'(1-P)\mathscr{L}_1(t')\right](1-P)\mathscr{L}_1(\tau)P\rho_{eq}(b)$$
$$=\rho_{eq}^{-1}(b)P\mathscr{L}_1 e^{(\mathscr{L}_a+\mathscr{L}_b)t}\overleftarrow{\exp}\left[\int_\tau^t dt'(1-P)\mathscr{L}_1(t')\right](1-P)e^{-(\mathscr{L}_a+\mathscr{L}_b)\tau}\mathscr{L}_1 P e_{eq}(b) . \tag{3.13}$$

We follow the spirit of the LRT. This means that the contributions of an order higher than $\mathscr{L}_1^2$ are disregarded and $K(t-\tau)$ can be approximated by

$$K(t-\tau)=\rho_{eq}^{-1}(b)P\mathscr{L}_1 e^{(\mathscr{L}_a+\mathscr{L}_b)t}(1-P)$$
$$\times e^{-(\mathscr{L}_a+\mathscr{L}_b)\tau}\mathscr{L}_1 P\rho_{eq}(b) . \tag{3.14}$$

It must be stressed that higher-order contributions in $\mathscr{L}_1$ might stem from the non-Markovian character of Eq. (3.12). In fact by developing $\sigma(t-\tau)$ into a Taylor power series around $\tau=0$ we obtain [from Eq. (3.12)]

$$\frac{\partial}{\partial t}\sigma=\mathscr{L}_a\sigma+\sum_{r=0}^{\infty}\frac{1}{r!}\int_0^t d\tau K(\tau)(-\tau)^r\frac{\partial^r\sigma}{\partial t^r} . \tag{3.15}$$

This equation can be solved by an iterative procedure generating both terms of the order $\mathscr{L}_a^r$ ($r>0$) and terms of the order $\mathscr{L}_1^n$ with $n\geq 4$. We keep disregarding the latter kind of contributions. Thus from Eq. (3.15) we get

$$\frac{\partial}{\partial t}\sigma(a;t)=\mathscr{L}_a\sigma(a;t)+\sum_{r=0}^{\infty}\frac{1}{r!}\int_0^t dt\,K(\tau)(-\tau)^r\mathscr{L}_a^r\sigma(a;t)$$
$$=\mathscr{L}_a\sigma(a;t)+\int_0^t dt\,K(\tau)e^{-\mathscr{L}_a\tau}\sigma(a;t) . \tag{3.16}$$

Let us consider the operator $W(\tau)$ defined by

$$K(\tau)=W(\tau)e^{\mathcal{L}_a\tau}. \tag{3.17}$$

When Eq. (3.17) is replaced into Eq. (3.16) we obtain

$$\frac{\partial}{\partial t}\sigma(a;t)=\mathcal{L}_a\sigma(a;t)+\left[\int_0^t W(\tau)d\tau\right]\sigma(a;t). \tag{3.18}$$

In the present paper we are interested in studying the steady state. Therefore we shall replace Eq. (3.18) with

$$\frac{\partial}{\partial t}\sigma(a;t)=\mathcal{L}_a\sigma(a,t)+\hat{W}(0)\sigma(a;t), \tag{3.19}$$

where

$$\hat{W}(0)=\int_0^\infty W(\tau)d\tau. \tag{3.20}$$

The operator $W(\tau)$ can be given an explicit analytical expression by using a technique recently developed by Grigolini.[21] By using this technique we obtain

$$\hat{W}(0)=\frac{\partial}{\partial v}Q_1(x,v)\frac{\partial}{\partial v}+\frac{\partial}{\partial v}Q_2(x,v)\frac{\partial}{\partial x}. \tag{3.21}$$

We do not write the explicit (and complicated) expressions of Q_1 and Q_2, because the joint use of a mean-field approximation and the LRT assumption allow us to use a much simpler expression for $\langle Q_1\rangle$ ($\langle Q_2\rangle$ is not involved in the energy diffusion equation).

The energy absorbed from the radiation field per unit of time, P, can be evaluated from the operator W as follows [$S\equiv\frac{1}{2}\omega_I^2(\langle y^2\rangle_{\rm eq}/k_BT)^{1/2}$]:

$$P=\left[\frac{d\langle v^2\rangle}{dt}\right]_{\text{from }W}=\int_0^\infty d\tau\int dy\int dv\frac{v^2}{2}W(\tau)\sigma=\omega_I^4\int_0^\infty d\tau\int dy\int dv\frac{v^2}{2}y\frac{\partial}{\partial v}e^{(\mathcal{L}_a+\mathcal{L}_b)\tau}y\frac{\partial}{\partial v}e^{-\mathcal{L}_a\tau}\rho_{\rm eq}(y,w)\sigma$$

$$=4S^2\mathrm{Re}\left[\int_0^\infty\langle v(0)v(t)\rangle\exp(i\omega_Rt)dt\right], \tag{3.22}$$

thereby providing a result coincident with the prediction of the LRT.[1] This result is obtained by assuming σ to be very close to the equilibrium distribution ($\mathcal{L}_a\sigma=0$) so that $\exp(-\mathcal{L}_at)\sigma$ on the right-hand side of Eq. (3.22) can be replaced by σ.

The next step consists in approximating $\hat{W}(0)$ as follows (mean-field approximation):

$$\hat{W}(0)=\langle Q_1\rangle\frac{\partial^2}{\partial v^2}+\langle Q_2\rangle\frac{\partial}{\partial x}\frac{\partial}{\partial v}. \tag{3.23}$$

Let us use this approximated expression of $\hat{W}(0)$ to determine the energy absorbed from the external field per unit of time. Thus we obtain

$$\langle Q_1\rangle=P. \tag{3.24}$$

P, in turn, is determined from Eq. (3.22).

By applying the Stratonovich method[15] to Eq. (3.19) supplemented by Eqs. (3.23) and (3.24), we obtain

$$\frac{\partial}{\partial t}\sigma(E;t)=\left[\gamma\frac{\partial}{\partial E}E+(P+\gamma k_BT)\frac{\partial}{\partial E}E\frac{\partial}{\partial E}\right]\sigma(E;t), \tag{3.25}$$

which results in the steady-state distribution

$$\sigma_{\rm exc}\propto\exp\left[-E\Big/\left[\frac{P}{\gamma}+k_BT\right]\right],$$

$$\left[T_{\rm eff}\equiv T+\frac{P(\Delta\omega)}{k_B\gamma}\right]. \tag{3.26}$$

On the other hand, it has been shown[14,15] that in the absence of the interaction with the radiation field

$$\mathrm{Re}\langle v(0)v(t)\rangle_{\rm eq}e^{i\omega_Rt}=\tfrac{1}{2}\mathrm{Re}\left\{\int_0^\infty dE\,E\sigma_{\rm eq}(E)\int_0^\infty dt\exp\left[\left[\frac{3i\beta E}{4\omega_0^3}-i\Delta\omega-\frac{\gamma}{2}\right]t\right]\right\}, \tag{3.27}$$

with

$$\sigma_{\rm eq}(E)\propto e^{-E/k_BT}. \tag{3.28}$$

We are thus led to keep unchanged the LRT structure of our result while replacing in Eq. (3.27) $\sigma_{\rm eq}$ with $\sigma_{\rm exc}$. From Eqs. (3.22) and (3.27) we then obtain

$$P(\Delta\omega)=2S^2\left[1+\frac{P(\Delta\omega)}{\gamma k_BT}\right]^{-1}\mathrm{Re}\left\{\int_0^\infty dE\,E\sigma_{\rm exc}(E)\int_0^\infty dt\exp\left[\left[\frac{3i\beta E}{4\omega_0^3}-i\Delta\omega-\frac{\gamma}{2}\right]t\right]\right\}. \tag{3.29}$$

The novelty of this result compared to Refs. 14 and 15 is that $P(\Delta\omega)/S^2$ is no longer independent of the radiation field intensity. We evaluated $P(\Delta\omega)$ with an iterative numerical procedure and the corresponding results will be compared in Sec. IV with those derived from the analog simulation experiment.

Equation (3.29), supplemented by Eq. (3.26), suggests analytical arguments on the range of validity of the LRT. First of all from Eq. (3.26) we obtain that the effective temperature is close to the unperturbed one if the condition

$$P(\Delta\omega)/\gamma k_B T \ll 1 \tag{3.30}$$

is fulfilled. From Eq. (3.29) we derive that (when $\gamma \ll \alpha$)

$$P(\Delta\omega) \sim S^2 k_B T/\alpha \, . \tag{3.31}$$

By replacing Eq. (3.31) into Eq. (3.30) we obtain

$$S^2/\alpha\gamma \ll 1 \, . \tag{3.32}$$

This is a significant result. It is reminiscent of the analog relationships which ensure conditions of negligible saturation in the field of magnetic resonance phenomena.[22] The transverse, T_2, and longitudinal, T_1, relaxation involved by these processes are replaced here by $1/\gamma$ and $1/\alpha$, respectively. If we take into account the fact that $\gamma \ll \alpha$, from Eq. (3.32) we derive that the range of validity of the LRT extends well beyond the limits predicted by the seemingly more intuitive relationship

$$S \ll \gamma \, . \tag{3.33}$$

Note that Eq. (3.29) also shows that the increase in peak frequency accompanies with the increase in width of the absorption spectrum. In other words, the linewidth of the absorption spectrum undergoes negligible changes if the peak frequency is given weak changes. In Sec. IV these predictions will be compared to the results of analog simulation.

The fact that in the linear case the absorption spectrum is given the Lorentzian shape predicted by the LRT regardless of the intensity of the excitation field must not be confused with the problem of the range of validity of Eq. (3.25). In Ref. 18 it has been remarked that Eq. (3.25) corresponds to an incoherent distribution of the variables x and v of the Duffing oscillator and that by increasing the intensity of the radiation field a transition to a coherent state can be provoked. In the linear case this change from a coherent to an incoherent state does not affect the shape of the absorption spectrum. This is so for the reasons already illustrated in Sec. II. Note also that in a linear system the time behavior of an excited autocorrelation function of x or v is independent of the intensity of the excitation (see Appendix A).

It is worth mentioning that Eq. (3.27) seems to belong to the same class of expressions as that found by Visscher,[5] i.e., formally equivalent to the LRT but containing also higher-order contributions of the external field. In Eq. (3.29) the higher-order contributions stem from the weighting function which depends on an effective temperature, which, in turn, depends on the excitation process. This formula therefore allows us to assess the range of validity of the LRT simply by determining the effects provoked by replacing T with T_{eff}.

The approach followed by Kenkre,[4] on the contrary, formally recovers the LRT without justifying the corresponding approximation. We are therefore led to conclude that the present approach shares the theoretical background of Kenkre,[4] namely, the Zwanzig projection approach,[11] without giving room to any criticism. Note, indeed, that according to van Velsen,[9] Kenkre's approach is basically equivalent to Kubo's approach.

IV. COMPARISON BETWEEN THEORY AND ANALOG SIMULATION

The analog simulation apparatus used in this paper is basically the same as that used in Refs. 14 and 16. The main difference with respect to Ref. 16 is that attention is now focused on the determination of $P(\Delta\omega)$ rather than $\langle E(\Delta\omega)\rangle$. This provides extremely more precise results, which are illustrated in Fig. 2.

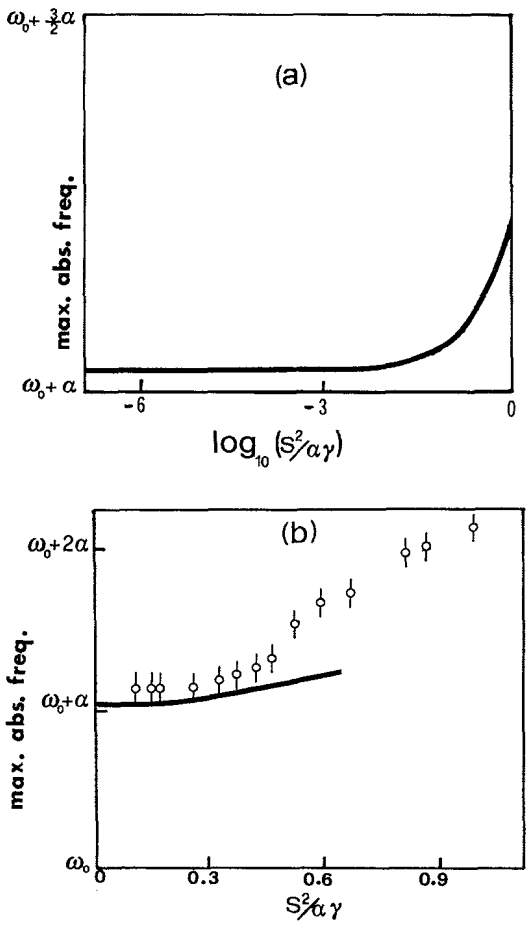

FIG. 2. Frequency of maximum absorption as a function of the field intensity S. (a) Theoretical prediction on the frequency of the maximum absorption as a function of $\log_{10}(S^2/\alpha\gamma)$. (b) Comparison between theoretical (full line) and experimental results (circles). The parameters used are $\omega_0 = 11.4 \times 10^3$ (c/sec); $\beta = 5.10^6$ (c/sec$\times$V)^2; $\alpha = 0.118\omega_0$; $\gamma = 0.010\omega_0$.

We are now in a position to settle two basic questions: (a) Is the LRT valid or not in the extremely underdamped regime $\gamma \ll \alpha$? (b) If the LRT of valid even in this critical region, how much extended is its range of validity? Does Eq. (3.33) or Eq. (3.32) hold?

The joint use of theory and analog simulation allows us to explore a fairly wide range of values of the radiation intensity. Note indeed that for technical reasons we could not explore with the analog simulation the region $S^2/\alpha\gamma \sim 0.1$, where the theoretical treatment of Sec. IV applies without difficulty. For $S^2/\alpha\gamma \sim 0.5$, on the contrary, the predictions of the theoretical treatment are not more completely correct and we must rely on the results of analog simulation. A proper joint use of the theoretical finding and analog simulation results (which are in a good agreement with each other in the region $0.1 < S^2/\alpha\gamma < 0.5$) leads us to the following picture. The peak of the absorption peak is placed at $\Delta\omega \sim \alpha$ for a wide interval of the radiation field intensity, ranging from $S^2/\alpha\gamma = 0$ to about $S^2/\alpha\gamma \sim 0.5$. Then the frequency position of the absorption peak increases more rapidly and reaches a new region of slow increase (a sort of "new plateau") at about $S^2/\alpha\gamma \sim 0.9$ with peak frequency $\Delta\omega \sim 2\alpha$.

We can thus settle the question (a) by stating that the LRT is certainly fulfilled if radiation fields of enough weak intensity are used. Furthermore the above remarks enable us to account for the wrong conclusions of Ref. 16. The radiation field intensity of that experiment corresponds indeed to the new plateau. We could assess that even in the region of this new plateau the linewidth of the absorption spectrum is proportional to the frequency position of the absorption peak [in the region of the first plateau this property fits the theoretical predictions of Eq. (3.27)]. As a consequence of this, the slow increase of the frequency of the absorption peak reflects in an almost negligible change of the line shape of the absorption spectrum, generating therefore the false impression that, in a region well far from saturation, the absorption peaks settles at $\Delta\omega \sim 2\alpha$. This frequency is well far from the prediction of the LRT, which, as also confirmed by the investigation of the present paper, predicts the absorption peak to settle at $\Delta\omega = \alpha$. Note that $\Delta\omega = 2\alpha$ is precisely the frequency position where, according to the "quantization rules" of Ref. 15 one should expect an intense resonance effect if the transverse relaxation properties stemming from the joint work of fluctuation and anharmonic interaction were neglected. The role of this relaxation process (disregarded in Ref. 16) in making the LRT valid has been widely stressed in Sec. III. In accordance with these theoretical results, the more accurate "analog experiment" of this paper shows that with decreasing the intensity of the parameter S, after an apparent settlement at $\Delta\omega = 2\alpha$, the absorption peak eventually attains the frequency position $\Delta\omega = \alpha$.

As to question (b), Fig. 2 shows that the generalized version of the LRT of Sec. III fits fairly well with the experimental results up to $S^2/\alpha\gamma \sim 0.5$. The slight discrepancy between theory and analog simulation throughout the interval $0.1 < S^2/\alpha\gamma \sim 0.5$ probably has to be ascribed to a systematic error related to the uncertainty affecting the experimental determination of the parameters characterizing the system under study. Both theory and experiment exhibit a small increase of the peak frequency with increasing S in the high-field part of this region. This increase is so small as to affect the predictions of the standard LRT by an error less than 10%. *This means that the predictions of the LRT is fulfilled beyond the seemingly natural limit $S = \gamma$, i.e., the relationship of Eq. (3.33) does not hold.* We have indeed found that at $S^2/\alpha\gamma \sim 0.5$ the radiation field intensity is $S = 0.0245$ to be compared to $\gamma = 0.01\omega_0$ ($\alpha = 0.118\omega_0$).

Equation (3.27) predicts slow increase of the peak frequency of the absorption spectrum with increasing S. This slow increase parallels fairly well the experimental results up to $S^2/\alpha\gamma \sim 0.5$. Beyond this region the experimental increase becomes much faster than the theoretical prediction and then, as already remarked above, a sort of new plateau is reached. This cannot be reproduced by the theory of this paper which takes into account the field effect only via the excited distribution of energy of Eq. (3.26) while retaining the Born approximation behind Eq. (3.14). The iterative method of solution of Eq. (3.29), furthermore, produces a slower and slower numerical convergence with increasing S, thereby preventing this method from being applied for $S^2/\alpha\gamma > 0.6$. The regions of even higher intensity require that the incoherent state description of this paper is replaced by the coherent state description by Nitzan and Carmeli.[23(a),18]

V. CONCLUDING REMARKS

The major results of this paper are found in Secs. III and IV. First of all the results illustrated there mean that we are obliged to leave the point of view expressed in Ref. 16. This short note[16] relies completely on a quite preliminary result of analog simulation which shows both the standard inertial regime ($\gamma > \alpha$) and the extremely underdamped one ($\gamma < \alpha$) to be characterized by an absorption peak at $\Delta\omega = 2\alpha$. We found the surprising result that the absorption spectrum, certainly far from the saturation region and rescaling with the square of the field intensity, peaks at about $\Delta\omega = 2\alpha$ even in the region where our calculations based on the LRT predict a peak at about $\Delta\omega = \alpha$. We arrived, therefore, at the wrong conclusion that the extremely underdamped regime is characterized by both the breakdown of the statistical linearization procedure[19] (which leads everywhere to the renormalized harmonic frequency $\omega_0 + 2\alpha$) and that of the LRT itself.

We are now in a position to conclude that the breakdown of the statistical linearization[19] is not accompanied by that of the LRT. On the contrary, the range of validity of the LRT is proven to be made wider by the nonlinear character of the potential, due to the fact that when the intensity of the random process tends to vanish, a transverse relaxation process (generated by the anharmonic interaction) comes into play.

The additional broadening, which in this paper is proven to be responsible for the more extended range of validity of the LRT, has also been found by other authors.[24,25] This broadening is therefore a well-settled property. To the best of our knowledge, however, this paper is the first

relating, with theoretical arguments, this property to an observable spectroscopic effect such as the initial plateau of Fig. 2. A further element of interest is the fact that these effects were investigated via an experiment of analog simulation.

The problem discussed in this paper is closely related to that of resonantly activated processes, which are presently the subject of a wide investigative effort.[15,23] Note that the validity of the theory illustrated in Sec. III does not rely on the stochastic description of the radiation field used there. In Appendix A we show indeed that the same results can be found by adopting a completely deterministic picture of the radiation field [$\lambda=0$, $f(t)=0$] provided that the intensity

$$S=\frac{1}{2}\frac{\omega_I^2}{\omega_R}\left[\frac{\omega_R^2 y_+^2+w^2}{2k_BT}\right]^{1/2}$$

of the deterministic description replaces the parameter

$$S=\tfrac{1}{2}\omega_I^2\left[\frac{\langle y^2\rangle_{\rm eq}}{k_BT}\right]^{1/2}$$

of the stochastic description.

This means that if the radiation field is small enough the diffusion equation of Eq. (3.18) leading to the Boltzmann-like distribution of Eq. (3.26) is certainly valid either if the excitation field is stochastic (with $\lambda=0$) or coherent and deterministic [$\lambda=0$, $f(t)=0$]. The regime of validity of this Boltzmann-like distribution in the nonlinear case is certainly more extended than in the linear one. This is so because the transverse relaxation mechanism produced by the anharmonic interaction, becoming predominant at $\alpha>\gamma$, makes available that mechanism of fast erasure of the relative phase (difference between the system and the driving field phase) which, according to Carmeli and Nitzan[23(a)] is indispensable to generate the Boltzmann-like distribution of energy with a resonant effective temperature [see Eq. (3.26)].

This is in a merely apparent contrast with the statement of Appendix A that the range of validity of the LRT is unlimited in the linear case. It must be stressed indeed that these are two completely different predictions. In spite of the fact that in the linear case the absorption of energy keeps its Lorentzian form regardless of the intensity of the radiation field, at certain intensities of the radiation field the distribution of energy deviates significantly from the Boltzmann distribution. By using analog simulation it has been possible to show[18] that with increasing α, and leaving unchanged the other parameters, the system makes a transition from the coherent to the incoherent state distribution [the latter as predicted by Eq. (3.25)], in complete agreement with the findings of this paper.

APPENDIX A: ON THE GENERALIZATION OF THE LRT

The major aim of this appendix is to support, with additional theoretical arguments, the statements made throughout the present paper that in the linear and Gaussian case the range of validity of the LRT would be unlimited. This affords also a straightforward way of generalizing this theory.

One of the most appealing aspects of the LRT is that the behavior of the system of interest after a weak excitation is traced back via this popular theoretical approach to some relevant equilibrium properties of the system. Let us consider, for instance, a system described by the equation of motion

$$\frac{\partial}{\partial t}\rho(a,b;t)=\mathscr{L}\rho(a,b;t)\ , \tag{A1}$$

where a denotes the variable of interest and b the set of the remaining variables. The operator $\mathscr{L}$ either denotes a rigorous Liouvillian or a Fokker-Planck operator, i.e., a sort of effective Liouvillian.

A significant equilibrium property is the correlation function

$$\Phi_a(t)\equiv\frac{\langle a(0)a(t)\rangle_{\rm eq}}{\langle a^2\rangle_{\rm eq}}\ , \tag{A2}$$

where

$$\langle Q\rangle_{\rm eq}\equiv\int da\int db\, a(e^{\mathscr{L}^\dagger t}a)e_{\rm eq}(a,b)\ , \tag{A3}$$

$\mathscr{L}^\dagger$ is the operator adjoint to that of Eq. (A1), and $\rho_{\rm eq}(a,b)$ is the equilibrium distribution of the whole system, defined by

$$\mathscr{L}\rho_{\rm eq}(a,b)=0\ . \tag{A4}$$

The LRT makes a wise use of the information provided by $\phi_a(t)$. If the variable a is slightly removed from its equilibrium state, which is assumed being characterized by

$$\langle a\rangle_{\rm eq}=0\ , \tag{A5}$$

the corresponding regression to equilibrium is determined by that information via

$$\langle a(t)\rangle_{\rm exc}=\langle a(0)\rangle_{\rm exc}\Phi_2(t)\ . \tag{A6}$$

It is well known[26] that in the linear and Gaussian case, the equilibrium correlation function of Eq. (A2) provides a fully exhaustive description of the dynamical properties of the system under study. For instance, the four-times equilibrium correlation function

$$\langle a(t_1)a(t_2)a(t_3)a(t_4)\rangle_{\rm eq}$$

can be traced back to $\Phi_2(t)$ via the definition itself of Gaussian variable which results in ($t_1\geq t_2\geq t_3\geq t_4$)

$$\langle a(t_1)a(t_2)a(t_3)a(t_4)\rangle_{\rm eq}=\langle a^2\rangle_{\rm eq}^2[\Phi_a(t_1-t_2)\Phi_a(t_3-t_4)+\Phi_2(t_1-t_3)\Phi_a(t_2-t_4)+\Phi_a(t_1-t_4)\Phi_2(t_2-t_3)]\ . \tag{A7}$$

When the variable a is not Gaussian, the detection of the four-times correlation function provides information on the system, additional to that associated with $\Phi_a(t)$. Now the question arises of how to exploit such additional information as wisely as the LRT does via Eq. (A6). This appendix is also devoted to extend this appealing aspect of the LRT.

First of all, let us assume that the equilibrium distribution reads

$$\rho_{eq}(a,b) \propto e^{-(a^2/2\langle a^2\rangle_{eq})}\psi_{eq}(b) . \tag{A8}$$

Note that the fact that a is given a Gaussian equilibrium distribution is not incompatible with the breakdown of the dynamical Gaussian property illustrated by Eq. (A7).[27] Let us now consider an excited distribution expressed by

$$\rho_{ex}(a,b)=\sigma_{exc}(a)\psi_{eq}(b) . \tag{A9}$$

$\sigma_{exc}(b)$ can be expressed via the following expansion:

$$\sigma_{exc}(a)=\sigma_{eq}(a)\sum_{n=0}^{\infty} a_n H_n\left[\frac{a}{\sqrt{2}\langle a^2\rangle^{1/2}}\right] , \tag{A10}$$

where

$$\sigma_{eq}(a)=\frac{1}{(2\pi a)^{1/2}}e^{-(a^2/2\langle a^2\rangle_{eq})} \tag{A11}$$

and the $H_n(\xi)$ denote Hermite polynomials with the normalization properties ($\xi \equiv a/\langle a^2\rangle^{1/2}$)

$$\frac{1}{\sqrt{2\pi}}\int_{-\infty}^{+\infty} d\xi\, e^{-\xi^2/2}H_n\left[\frac{\xi}{\sqrt{2}}\right]H_n\left[\frac{\xi}{\sqrt{2}}\right] = \begin{cases} 0 , & n\neq m \\ 2^n n!\equiv N_n , & n=m . \end{cases} \tag{A12}$$

The expansion coefficients are defined by

$$a_0=1 , \quad a_1=\frac{1}{\sqrt{2}}\frac{\langle a\rangle_{exc}}{\langle a^2\rangle_{eq}^{1/2}} , \qquad a_n=\frac{1}{N_n}\frac{1}{\langle a^2\rangle_{eq}^{1/2}}\int da\,\sigma_{exc}(a)H_n \times\left[\frac{a}{2\langle a^2\rangle_{eq}}\right] , \quad n>1 . \tag{A13}$$

We are thus in a position to evaluate the regression to equilibrium of the variable a. By applying the relationship

$$\langle a(t)\rangle = \int\int (e^{\mathscr{L}^\dagger t}a)\rho_{exc}(a,b)da\,db = \int\int a e^{\mathscr{L}t}\rho_{exc}(a,b)da\,db , \tag{A14}$$

which enables us to change from the "Heisenberg" to the "Schrödinger" representation, and by making use of Eq. (A10) we get

$$\langle a(t)\rangle_{exc}=\sum_{n=0}^{\infty} a_n\left\langle H_n\left[\frac{a}{(2\langle a^2\rangle_{eq})^{1/2}}\right]a(t)\right\rangle_{eq} . \tag{A15}$$

This is an elegant generalization of a former expansion.[27] Note that due to parity conservation the contribution coming from odd n vanishes. In the Gaussian case [when Eq. (A7) and the corresponding higher-order relationships hold], all the contributions with $n>1$ are proven to vanish and Eq. (A15) becomes coincident with Eq. (A6). *In other words in the Gaussian case the range of validity of the LRT is* unlimited. Note that the validity of the LRT in the non-Gaussian case, on the contrary, relies on the assumption that the expansion in power series of Eq. (A10) is possible and may be truncated to the lowest significant order. This is equivalent to saying that the excitation process itself, necessary to produce these deviations from equilibrium, is weak enough as to render valid the LRT.

We divided the excitation-relaxation processes into two well-separated regimes. The excitation regime concerns times from $t=-\infty$ up to $t=0$. At $t=0$, the excitation field is suddenly turned off, and we start monitoring the decay process. The details of the excitation process are completely ignored and replaced by a proper initial distribution [see Eq. (A9)]. This, of course, could raise the widely discussed question[28] of how much the irrelevant

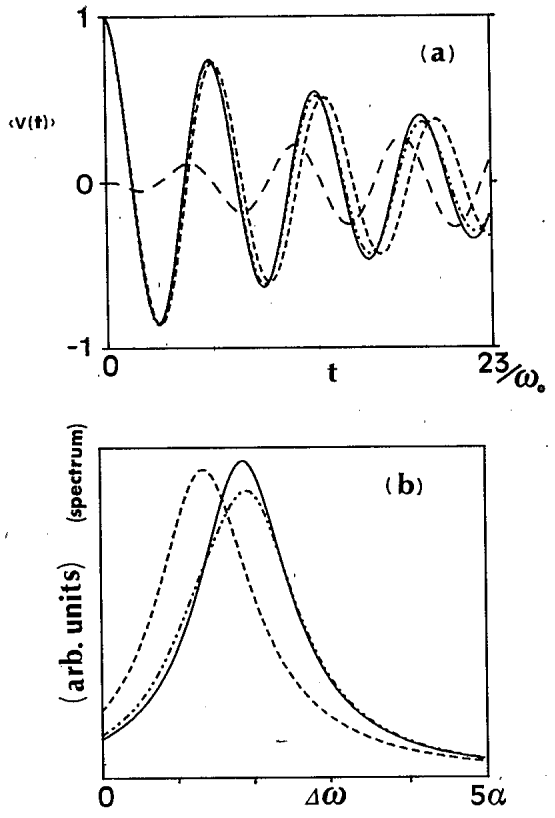

FIG. 3. Time evolution of the excited mean value $\langle v(t)\rangle$, Eq. (A15) with $a=v$, (a), and the corresponding spectrum, (b). The form of the excited distribution is $\rho_{exc}(v)=\exp[-(v-u)^2/2(1+\epsilon)k_BT]$. (a) -----, unperturbed case; ——, excited case; —··—, unperturbed term plus the first correction term; and — — —, the first correction. (b) -----, unperturbed case; ——, excited case; and —··—, unperturbed term plus the first correction term. The parameters used are $\alpha=0.0075$, $\gamma=0.1$, $\epsilon=0.5$: $u=0.1$, $\omega_0=1$.

variables b themselves are driven far from the equilibrium state during the excitation process. In the non-Markovian case, the regression to equilibrium of the "irrelevant" variables produces significant effects which in the present appendix are deliberately ignored via the *ad hoc* choice of Eq. (A9).

By a computer calculation based on the continued-fraction procedure of Ref. 17 we are in a position to show (see Fig. 3) that the predictions of Eq. (A15) are correct. It is interesting to remark that the shift towards high frequencies of the spectrum of the excited observable is fairly well reproduced by using only the first two nonvanishing contributions to Eq. (A15) (i.e., the standard one plus the first correction term).

APPENDIX B

The main aim of this appendix is to show that Eq. (3.25) can also be derived within a purely deterministic description of the radiation field. Let us consider the case

$$\begin{aligned} &\dot{x}=v\ , \\ &\dot{v}=-\frac{\partial V}{\partial x}-\gamma v+f(t)+A\cos(\omega_R t)\ , \end{aligned} \tag{B1}$$

where $f(t)$ is the same stochastic force as that defined by Eq. (1.2). In such a case we must deal with the time-dependent Fokker-Planck equation

$$\begin{aligned} \frac{\partial}{\partial t}\rho(x,v;t)&=\mathscr{L}(t)\rho(x,v;t) \\ &\equiv[\mathscr{L}_a+\mathscr{L}_1(t)]\rho(x,v;t)\ , \end{aligned} \tag{B2}$$

where

$$\mathscr{L}_a\equiv -v\frac{\partial}{\partial x}+\frac{\partial V}{\partial x}\frac{\partial}{\partial v}+\gamma\left[\frac{\partial}{\partial v}v+k_BT\frac{\partial^2}{\partial v^2}\right]\ , \tag{B3}$$

$$\mathscr{L}_1(t)\equiv -A\cos(\omega_R t)\frac{\partial}{\partial v}\ . \tag{B4}$$

Let us write Eq. (B2) in the interaction picture

$$\frac{\partial}{\partial t}\tilde{\rho}=\tilde{\mathscr{L}}_1(t)\tilde{\rho}\ , \tag{B5}$$

$$\tilde{\rho}=e^{-\mathscr{L}_a t}\rho\ , \tag{B6}$$

$$\tilde{\mathscr{L}}_1(t)\equiv e^{-\mathscr{L}_a t}\mathscr{L}_1(t)e^{\mathscr{L}_a t}\ , \tag{B7}$$

then, as usual,[13] let us apply to Eq. (B5) the Zwanzig projection procedure. The only minor difference with respect to Sec. III is the fact that the projection operator is now defined by

$$\begin{aligned} \sigma(x,v;t)&\equiv P\rho(x,v;t) \\ &=P\sum_{\mu=\pm1,0}e^{i\mu\omega_R t}\rho\mu(x,v;t) \\ &=\rho_0(x,v;t)\ . \end{aligned} \tag{B8}$$

From now on the same assumptions as those in Sec. III can be made and precisely the same approach can be followed. We thus obtain

$$\frac{\partial}{\partial t}\sigma(x,v;t)=\mathscr{L}_a\sigma(x,v;t)+\frac{A^2}{2}\mathrm{Re}\left[\int_0^t d\tau\frac{\partial}{\partial v}e^{(\mathscr{L}_a+i\omega_R)(t-\tau)}\frac{\partial}{\partial v}e^{-\mathscr{L}_a(t-\tau)}\sigma(x,v;t)\right]\ , \tag{B9}$$

which coincides with Eq. (3.16) if we assume

$$\frac{A^2}{2}=\omega_I^4\langle y^2\rangle_{\mathrm{eq}}\ . \tag{B10}$$

The derivation of the diffusion equation of Eq. (3.25) is then precisely the same as that followed in Sec. III.

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