Non-Markovian theory of activated rate processes. V. External periodic forces in the low-friction limit

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(Received 12 April 1985)

The escape of a particle from a potential well under the influence of both thermal (generalized Langevin) noise and friction and an external periodic driving force is studied in the low-friction limit. We consider three models: (a) additive thermal noise and a completely coherent driving force; (b) additive thermal noise and a phase-diffusing driving force; (c) coherent driving force and multiplicative random noise. The last two models are characterized by dephasing which affects the escape dynamics both qualitatively and quantitatively. In all three cases the escape rate is resonantly enhanced; however, while the first case is characterized by a finite energy peak in the steady-state distribution function, the presence of strong dephasing in the other two cases leads to a generalized Boltzman distribution with an effective temperature which depends resonantly on the external pumping. The relevance of this work to recent experimental results on the resonant activation of a Josephson junction out of its zero-voltage state is discussed.

I. INTRODUCTION

In a recent series of articles[1] we have generalized Kramers's treatment[2] of escape of a Brownian particle from a potential well to the whole friction range,[1(a)–1(c)] to non-Markovian situations,[1(b)–1(f)] to situations which include the effects of coupled nonreactive modes,[1(g)] and to the presence of multiplicative random noise (position-dependent friction).[1(h)] Many authors have also over the years extended Kramers's theory in these and other directions.[3]

A related, relatively unexplored problem is the escape of a Brownian particle out of a potential well in the presence of an external periodic force. Processes such as multiphoton dissociation and isomerization of molecules in high-pressure gas or in condensed phases,[4] laser-assisted desorption,[5] and transitions in current-driven Josephson junctions under the influence of microwaves[6–8] may be described with such a model, where the periodic force results from the radiation field.

Some time ago we present a solution for the case of a truncated harmonic well in the low-friction Markovian limit.[6] For this case the escape rate is given by the inverse mean first-passage time for a particle starting with an energy sampled from the steady-state distribution to reach the critical escape energy. The equation of motion for this case is

$$\ddot{x}+\gamma\dot{x}+\omega^2 x=A\cos(\omega_R t)+(1/M)R(t)\ , \quad (1)$$

where x is the coordinate of the harmonic particle characterized by a mass M, frequency ω, and friction coefficient γ. A is the amplitude of the external force divided by M, of frequency ω_R. The thermal noise R satisfies

$$\langle R\rangle=0,\ \ \langle R(t_1)R(t_2)\rangle=2\gamma k_B T\delta(t_1-t_2)\ , \quad (2)$$

where T is the temperature and k_B the Boltzmann constant. The steady-state distribution for this model was found to be

$$P_{SS}(E)=N_E\exp\left[-\frac{2\omega_R^2}{\omega^2+\omega_R^2}\frac{(E^{1/2}-\bar{E}^{1/2})^2}{k_B T}\right]\ , \quad (3)$$

where

$$\bar{E}=\frac{M(\omega^2+\omega_R^2)A^2}{4[(\omega^2-\omega_R^2)^2+\gamma^2\omega_R^2]} \quad (4)$$

and the mean first-passage time to reach the threshold energy E_B is (for $\omega=\omega_R$)

$$\tau=(\gamma k_B T)^{-1}\times\int_{\bar{E}}^{E_B}\frac{dy}{y}\exp\left[\frac{y^{1/2}-\bar{E}^{1/2}}{k_B T}\right]\times\int_0^y dz\exp\left[-\frac{(z^{1/2}-\bar{E}^{1/2})^2}{k_B T}\right]\ . \quad (5)$$

For high barriers or low temperatures ($E_B-\bar{E}\gg k_B T$) Eq. (5) reduces to

$$\tau=\frac{1}{\gamma}\left[\frac{4\pi\bar{E}}{k_B T}\right]^{1/2}\frac{E_B-\bar{E}}{E_B}\exp\left[\frac{(E_B^{1/2}-\bar{E}^{1/2})^2}{k_B T}\right]\ . \quad (6)$$

For anharmonic potential surfaces the problem becomes much more complicated: The underlying deterministic equation (zero-temperature limit) may have several locally stable-state solutions corresponding to resonances of the external force with higher harmonics and subharmonics of the potential well. These give rise to the phenomenon of steps in the current-voltage characteristics of the microwave-driven Josephson junction.[9] For a strong external driving force the deterministic motion becomes chaotic and it seems impossible to identify a slow dynamic variable for a convenient reduction of the problem. [The simplicity of the solutions (3)—(5) stems from the fact that in the low-friction limit the energy near steady state varies much more slowly than the phase.] An analytical treatment is therefore possible only in the weak-oscillating-force limit.[10]

In many physical systems the situation becomes simpler due to the inherent stochastic nature of the driving field itself. To see the possible significance of this effect consider a conventional CO_2-laser pulse with duration of 10 nsec and bandwidth of 1 cm^{-1} incident on a diatomic molecule characterized by an environment-induced energy relaxation time of ~ 100 nsec. The laser pulse is obviously not uncertainty limited and its width is associated with the random fluctuations in its phase and/or amplitude. For simplicity we consider random phase fluctuations, whence the external field is

$$F(t)=F_0\cos\left[\omega_R t+\int^t dt'\phi(t')\right] \tag{7}$$

with ϕ being a Gaussian random variable

$$\langle\phi(t)\rangle=0,\quad \langle\phi(t_1)\phi(t_2)\rangle=2\Gamma\delta(t_1-t_2) \tag{8}$$

so that

$$\langle F(t_1)F(t_2)\rangle=F_0^2\exp(-\Gamma\,|\,t_1-t_2\,|\,)\cos[\omega(t_1-t_2)]\ . \tag{9}$$

Γ^{-1} is the inverse correlation time and Γ may be shown to be an additive part of the beam spectral width. For the present example τ_c is thus 10^{-11} sec. This is much shorter than the energy relaxation time, so that in this respect the radiation field is similar to a thermal bath. In particular, phase coherence necessary to generate deterministic chaos does not exist in this situation.

In Sec. II we consider a model which is the anharmonic and non-Markovian analog of Eq. (1),

$$\ddot{x}+\int_0^t d\tau Z(t-\tau)+\frac{1}{M}\frac{\partial V(x)}{\partial x}=F(t)+\frac{1}{M}R(t)\ , \tag{10}$$

$$\langle R(t)\rangle=0,\quad \langle R(t_1)R(t_2)\rangle=Mk_BTZ(t_1-t_2)\ , \tag{11}$$

where the external driving force satisfies Eqs. (7)—(9). We consider this system in the low-friction limit $[\hat{Z}(\omega)\ll\omega$ where $\hat{Z}(\omega)$ is the Fourier transform of $Z(t)]$, and show that the assumption of rapid phase randomization of the driving field makes it possible to derive a reduced Fokker-Planck equation for the system's energy, which in turn is used to obtain a simple equation for the low-friction escape rate. This should be contrasted with the deterministic limit ($\Gamma\rightarrow 0$, $T\rightarrow 0$) of the same model where the energy and phase variables vary on the same time scale, giving rise to multiple field-drive steady states and, for strong enough driving, to deterministic chaotic motion.

The observation that rapid dephasing of the driving force results in a simpler description of the stochastic dynamics suggests that, since the phase of interest is presumably the relative (difference between the system and the driving field) phase, similar simplification will arise from dephasing processes associated with the motion of the system. This leads us to consider (in Sec. III) such dephasing processes which arise from the presence of the thermal noise $R(t)$. In this context we find it useful to generalize Eq. (10) to include multiplicative noise terms $g(x)R(t)$. In the presence of such terms [and also for the case of purely additive noise if the potential $V(x)$ is anharmonic] it is possible, in analogy to quantum statistical mechanics, to distinguish between pure dephasing (T_2) processes and dephasing associated with energy relaxation (T_1) processes. We confirm in Sec. IV our expectation that in the presence of strong internal dephasing processes reduction to a simple energy Fokker-Planck equation is possible and an expression for the low-friction escape rate easily follows.

The validity of this model to describe laser-induced reactions in condensed phase is uncertain, even though such models have been used for this purpose before.[10,11] The reason for this is that many laser-induced processes are better modeled as a quantum-mechanical system involving only the limited number of discrete states which resonantly interact with the radiation field. In other cases where a classical model is advantageous because the number of quantum states is too large we should be concerned about rapid thermal relaxation and consequently heating of the environment, in contrast to the model assumption of fixed temperature. Provided that this assumption holds, the present model should be useful for quick estimates of the effects of oscillating driving fields on escape processes. In this respect it should be mentioned that some of the current experimental work on laser-induced desorption[5] mostly uses the laser as a fast heating device. It is of great interest to perform similar experiments using the laser to excite directly the adsorbate-substrate bond.[12] Such a process will be directly related to the calculation described below.

A recent experiment by Devoret *et al.*[8] has demonstrated that the transition from the zero-voltage state to the nonzero-voltage state of an underdamped current-biased Josephson junction may be resonantly activated by a weak microwave perturbation. The model discussed in Sec. IV is directly relevant to this experiment. We further discuss this application in Sec. V.

A recent paper by Faetti and Grigolini[12] discusses the effect of an external radiation field on the escape of a particle from a potential well. These authors discuss the high-friction (diffusion) limit in which the well motion is overdamped leading to elimination of resonance phenomena. Here we are concerned with the opposite low-friction limit and with the resonant response of the activation process.

II. PHASE-DIFFUSING DRIVING FIELD IN THE LOW-FRICTION LIMIT

We start from the Langevin equations

$$\dot{x}=v\ , \tag{12}$$

$$\dot{v}=-\frac{1}{M}\frac{dV(x)}{dx}-\int_0^t d\tau Z(t-\tau)v(\tau)+\frac{1}{M}R(t)+\frac{1}{M}F(t)\ , \tag{13}$$

where R is a Gaussian random function satisfying $\langle R\rangle=0$ and

$$\langle R(t_1)R(t_2)\rangle = Mk_BTZ(t_1-t_2)\ , \tag{14}$$

$$\langle F(t_1)F(t_2)\rangle = MY(t_1-t_2)\ . \tag{15}$$

The correlation functions $Z(t)$ and $Y(t)$ decay to zero on time scales τ_c and τ_c^y respectively. We assume that the time scales characterizing the process satisfy

$$\omega(E)^{-1} \ll \tau_c, \tau_c^y \ll (d\ln\langle E\rangle/dt)^{-1}\ , \tag{16}$$

where $\omega(E)$ is the frequency associated with the potential $V(x)$ and with the particle's mass M and energy E. Under these conditions we can use a reduction procedure identical to that described for the purely thermal case [Ref. 1(e), henceforth referred to as I] to derive a Smoluchowski equation for the energy. To this end we transform (x,v) to the action-angle coordinates (J,ϕ) and expand

$$x(J,\phi) = \sum_{n=-\infty}^{\infty} x_n(J)e^{in\phi}\ , \tag{17}$$

$$v(J,\phi) = \sum_{n=-\infty}^{\infty} v_n(J)e^{in\phi} \tag{18}$$

[note that $v_n(J) = in\omega(J)x_n(J)$, $\omega(J) = \partial E(J)/\partial J$].

Using the method in I (this method involves calculation of moments of the form $\langle[\Delta J_t(\tau)]^m[\Delta\phi_t(\tau)]^l\rangle$ where $\Delta J_t(\tau) = \int_t^{t+\tau} dt' J(t')$. In this calculation we neglect, as usual, terms of order τ^n, $n>1$, and also terms of order $[\hat{Z}(\omega)/\omega]^n$, $n\geq 1$. $\hat{Z}(\omega)$ is the Fourier transform of $Z(t)$ and the latter approximation corresponds to the low-friction limit) we obtain the following Fokker-Planck equation for $P(J,\phi,t)$:

$$\frac{\partial P(J,\phi,t)}{\partial t} = L_0P + L_1P\ , \tag{19}$$

$$L_0 = \frac{\partial}{\partial J}\epsilon(J)\left[k_BT\frac{\partial}{\partial J} + \omega(J)\right] + \Gamma(J)\frac{\partial^2}{\partial\phi^2} - \Omega(J)\frac{\partial}{\partial\phi}\ , \tag{20}$$

$$L_1 = \frac{\partial}{\partial J}\mu(J)\frac{\partial}{\partial J} + \lambda(J)\frac{\partial^2}{\partial\phi^2} + \eta(J)\frac{\partial}{\partial\phi}\ , \tag{21}$$

where[13]

$$\epsilon(J) = 2M\sum_{n=1}^{\infty} n^2x_n^2\hat{Z}_n^c\ , \tag{22}$$

$$\Gamma(J) = 2Mk_BT\sum_{n=1}^{\infty}\left[\frac{dx_n}{dJ}\right]^2\hat{Z}_n^c + Mk_BT\left[\frac{dx_0}{dJ}\right]^2\gamma\ , \tag{23}$$

$$\Omega(J) = \omega(J) - M\left[k_BT\frac{d}{dJ} - \omega(J)\right]\sum_{n=1}^{\infty} n\frac{d(x_n^2)}{dJ}\hat{Z}_n^s\ , \tag{24}$$

$$\mu(J) = 2M\sum_{n=1}^{\infty} n^2x_n^2\hat{Y}_n^c\ , \tag{25}$$

$$\lambda(J) = 2M\sum_{n=1}^{\infty}\left[\frac{dx_n}{dJ}\right]^2\hat{Y}_n^c\ , \tag{26}$$

$$\eta(J) = M\frac{d}{dJ}\sum_{n=1}^{\infty}\frac{d(x_n^2)}{dJ}\hat{Y}_n^s\ , \tag{27}$$

$$\gamma = \int_0^\infty dt\,\hat{Z}(t) = \hat{Z}_0^c\ , \tag{28}$$

and where $Z_n^c, Z_n^s, Y_n^c, Y_n^s$ are cos and sin Fourier transforms, e.g.,

$$Z_n^c(\omega) = \int_0^\infty dt\, Z(t)\cos(n\omega t)\ ,$$

$$Z_n^s(\omega) = \int_0^\infty dt\, Z(t)\sin(n\omega t)$$

and depend on the action J through the J dependence of ω. Integrating over ϕ and using the fact that P has to be periodic in ϕ we get [setting $P(J) = \int_0^{2\pi} d\phi P(J,\phi)$] the Smoluchowski equation

$$\frac{\partial P(J,t)}{\partial t} = \frac{\partial}{\partial J}\left[\epsilon(J)\left[k_BT\frac{\partial}{\partial J} + \omega(J)\right]P(J,t)\right] + \frac{\partial}{\partial J}\mu(J)\frac{\partial}{\partial J}P(J,t)\ , \tag{29a}$$

where J is the action variable. We can get the corresponding energy equation using the transformation $J\to J(E)$, $dJ = dE/\omega(E)$. This leads to

$$\frac{\partial P(E,t)}{\partial t} = \frac{\partial}{\partial E}\left[D_1(E)\left[k_BT\frac{\partial}{\partial E} + 1\right]\omega(E)P(E,t)\right] + \frac{\partial}{\partial E}\left[D_2(E)\frac{\partial}{\partial E}[\omega(E)P(E,t)]\right] \tag{29b}$$

with

$$D_1(E) = \omega(E)\epsilon(E)\ , \tag{30a}$$

$$D_2(E) = \omega(E)\mu(E)\ . \tag{30b}$$

The results (29) and (30) are a generalization of the energy-diffusion equation obtained in the absence of the external phase-diffusing driving force ($D_2=0$). It is interesting to note that (29b) may be written in the form

$$\frac{\partial P(E,t)}{\partial t} = \frac{\partial}{\partial E}\left[D_1(E)\left[k_BT_{\text{eff}}(E)\frac{\partial}{\partial E} + 1\right]\omega(E)P(E,t)\right]\ , \tag{31}$$

where the energy-dependent effective temperature is given by

$$T_{\text{eff}}(E) = T + \frac{D_2(E)}{k_BD_1(E)}\ . \tag{32}$$

The general steady-state solution ($\partial P_{\text{SS}}/\partial t = 0$) of Eq. (31) is

$$P_{\text{SS}}(E) = \frac{A_1}{\omega(E)}\exp\left[-\int_0^E dE'\beta(E')\right] - \frac{A_2}{\omega(E)}\int_0^E \frac{dE'\beta(E')}{D_1(E')}\exp\left[-\int_{E'}^E dE''\beta(E'')\right]\ , \tag{33}$$

where

$$\beta(E)=[k_B T_{\rm eff}(E)]^{-1} , \tag{34}$$

and where A_1 and A_2 are constants. One of them may be determined given a normalization condition and the other is easily shown to be the steady-state current

$$j_{\rm SS}=-D_1(E)\left[\beta^{-1}(E)\frac{d}{dE}+1\right]\omega(E)P_{\rm SS}(E)=A_2 . \tag{35}$$

The equilibrium solution (in the presence of the driving field) corresponds to zero current,

$$P_{\rm eq}(E)=\frac{A_1}{\omega(E)}\exp\left[-\int_0^E dE'\beta(E')\right] . \tag{36}$$

In the very-low-friction limit the barrier energy E_B may be considered to be an absorbing boundary for the diffusion motion described by (31), i.e., $P_{\rm SS}(E_B)=0$. This and Eq. (33) imply

$$A_1=A_2\int_0^{E_B}dE'\frac{\beta(E')}{D_1(E')}\exp\left[\int_0^{E'}dE''\beta(E'')\right] . \tag{37}$$

The rate is given by

$$k=j_{\rm SS}\Big/\int_0^{E_B}dE\,P_{\rm SS}(E) \tag{38}$$

which, using (33)–(38), results in

$$k=\left[\int_0^{E_B}dE\frac{\beta(E)}{\omega(E)D_1(E)}[P_{\rm eq}(E)]^{-1}\int_0^E dE'P_{\rm eq}(E')\right]^{-1} . \tag{39}$$

The following observations can be made concerning these results.

(a) When $\beta(E)$ is replaced by $(k_BT)^{-1}$ the result (39) becomes identical to the low-viscosity rate obtained in the purely thermal case (see paper I).

(b) The effect of the phase-diffusion driving force enters through the (generally energy-dependent) effective temperature $T_{\rm eff}(E)$. To see this effect more explicitly we may consider the harmonic-oscillator case together with the simple choices

$$Z(t)=\frac{\gamma}{\tau_c}e^{-t/\tau_c} , \tag{40}$$

$$Y(t)=\bar{F}^2e^{-\Gamma t}\cos(\omega_R t) . \tag{41}$$

τ_c and $\tau_c^y=\Gamma^{-1}$ are the thermal and the driving-force correlation times, respectively. Equations (22) and (25) become [in the harmonic-oscillator case only the term $n=1$ appears and $x_1=(J/2M\omega)^{1/2}=\omega^{-1}(E/2M)^{1/2}$]

$$\epsilon(J)=\frac{J}{\omega}\frac{\gamma}{1+(\omega\tau_c)^2} , \tag{42}$$

$$\mu(J)=\frac{J\bar{F}^2}{2}\frac{\Gamma}{\Gamma^2+(\omega_R-\omega)^2} \tag{43}$$

(in (43) we disregard a small term proportional to $\Gamma[\Gamma^2+(\omega_R+\omega)^2]^{-1}$). Using Eqs. (30), (32), (42), and (43) we get

$$k_BT_{\rm eff}=k_BT+\frac{\bar{F}^2}{2\Gamma\gamma}\frac{1+(\omega\tau_c)^2}{1+[(\omega_R-\omega)/\Gamma]^2} . \tag{44}$$

We see that $T_{\rm eff}$ goes through a maximum near the resonance condition $\omega_R=\omega$ (ω is the oscillator frequency). The phase-diffusing driving force results in a thermallike kinetics with a renormalized temperature which is resonantly enhanced relative to the bare temperature.

(c) The result just discussed is in sharp contrast to that obtained for a coherent incident radiation field. For this case in the harmonic limit the equilibrium distribution is given by Eq. (3) and has a maximum for $E=\bar{E}$ where resonant behavior enters through the resonant nature of $\bar{E}$, Eq. (4). In the present case the driving field enters through the effective temperature. Physically the difference between the two situations arises from the fact that in the former one the deterministic ($T\rightarrow 0$) system has a well-defined phase relative to the radiation field while here, because of the field's phase diffusion, this coherence is lost and the field operates in this respect as a temperature source. Remarkably, *this effective-temperature source still maintains its resonance properties.*

(d) As in other calculations of the escape rate a first-order kinetics characterized by a constant rate is valid only when the well depth is much larger than k_BT so that the long-time dynamics reflects the establishment of a quasisteady distribution in the well. Note that the mean first passage time to go from some initial energy E_0 to E_B is given by

$$\tau(E_B,E_0)=\int_{E_0}^{E_B}\frac{dE\beta(E)}{\omega(E)D_1(E)}[P_{\rm eq}(E)]^{-1}\int_0^E dE'P_{\rm eq}(E') \tag{45}$$

and that another common expression for the steady-state rate is

$$k=\left[\int_0^{E_B}dE\,P_{\rm SS}(E)\tau(E_B,E)\right]^{-1} . \tag{46}$$

For sufficiently deep wells, $\tau(E_B,E_0)$ is a very weak function of E_0 up to energies close to E_B so that for a temperature sufficiently small relative to E_B the results (46) and (39) are practically identical.

(e) The qualitative difference observed between the effect of a fully deterministic and a phase-diffusing periodic force has been attributed to the rapid elimination of the relative phase between the particle motion and the driving force. This suggests also that fully deterministic driving forces may lead to stochastic dynamics similar to that obtained here (i.e., governed by a field-dependent effective temperature), provided that the thermal fluctuations affecting our system result in a rapid erasure of the same relative phase. In the harmonic-oscillator example of Ref. 6 (reviewed above) such a dephasing process is absent (due to linearity of both the system's deterministic motion and of its coupling to the bath). As we shall see below, when strong dephasing prevails (usually a consequence of multiplicative random noise and/or strong anharmonicity of the deterministic motion) the energy-diffusion equation is again controlled by a field-dependent effective temperature. Before turning to this we shall briefly discuss the origin of dephasing effects in classical stochastic dynamics.

III. DEPHASING IN THE LOW-FRICTION LIMIT

Dephasing process are usually discussed in their quantum-mechanical context, referring to the destruction of the quantum-mechanical phase of a system due to its coupling to the thermal environment. It is customary to distinguish between pure (proper) dephasing (T_2) processes, which lead to phase destruction without energy change, and total dephasing, which includes also the effect of energy-changing (T_1) interactions. In quantum mechanics pure dephasing is associated with the diagonal (in the system energy eigenstate representation) part of the system—thermal-environment interaction. As we show below, the analogous process in classical mechanics is often (but not necessarily) associated with the presence of multiplicative random noise.

Several authors[14–17,1(h)] have recently discussed the effect of multiplicative random noise on the escape of a Brownian particle out of a potential well. For the low-friction limit of the non-Markovian problem modeled by

$$\dot{x}=v \,, \tag{47}$$

$$\dot{v}=-\frac{1}{M}\frac{dV(x)}{dx}-f(x(t))\int_0^t d\tau Z(t-\tau)f(x(\tau))v(\tau)+\frac{1}{M}f(x(t))R(t)\,, \tag{48}$$

$$\langle R(t)\rangle=0,\quad \langle R(t_1)R(t_2)\rangle=Mk_BTZ(t_1-t_2) \tag{49}$$

we have shown[1(h)] that the energy distribution evolves according to the Smoluchowski-like equation

$$\frac{\partial P(E,t)}{\partial t}=\frac{\partial}{\partial E}\left[D(E)\left[k_BT\frac{\partial}{\partial E}+1\right]\omega(E)P(E,t)\right]\,, \tag{50}$$

where

$$D(E)=2M\omega(E)\sum_{n=1}^{\infty}n^2\,|\,G_n(E)\,|\,^2Z_n^c(\omega(E))\,, \tag{51}$$

and where $G_n(E)=G_n(J(E))$ is related to $G(x)=\int^x dx'f(x')$ by [using the transformation $(x,v)\rightarrow(J,\phi)$]

$$G(J,\phi)=\sum_{n=-\infty}^{\infty}G_n(J)\exp(in\phi)\,. \tag{52}$$

Note that $D_1(E)$ of Eq. (30a) is a special case of Eq. (51) in which $f(x)=1$ and $G(x)=x$.

It should be noted that Eq. (48) may be derived from a microscopic model[18,19] only for the special case where the friction kernel does not depend on the particle's velocity. This is not generally the case and a rigorous derivation of reduced stochastic equations describing the motion of a subsystem coupled nonlinearly to its thermal environment leads to more-complicated equations (see Refs. 18—21 for further discussions of this issue). Equation (48) may still be derived for special cases. Here we use this equation as a model for a simple demonstration of the dephasing process.

Pure classical dephasing may be understood in terms of the general equations of motion for the particle in the action-angle representation

$$\dot{J}=-\frac{\partial H}{\partial\phi}=-\frac{\partial V_{\rm SB}}{\partial\phi}\,, \tag{52$'$}$$

$$\dot{\phi}=\frac{\partial H}{\partial J}=\frac{\partial(H_S+V_{\rm SB})}{\partial J}\,, \tag{52$''$}$$

where H is the total Hamiltonian (including the thermal environment), H_S the system's Hamiltonian, and $V_{\rm SB}$ the system-bath interaction. Expanding

$$V_{\rm SB}=\sum_{n=-\infty}^{\infty}V_n^{(\rm SB)}(J)e^{in\phi}\,, \tag{53}$$

where $V_n^{(\rm SB)}(J)$ depends on J and on the thermal bath coordinates, it is obvious that the $n=0$ term of (53) leads to a time dependence of ϕ due to the system-bath coupling which is not associated with a corresponding time dependence of J. The $n=0$ term in (53) is thus a source of pure dephasing ("T_2 processes") while the $n\neq0$ terms induce both phase and energy relaxation.

For a harmonic oscillator [where $x(J,\phi)=\sqrt{2J/M\omega}\cos\phi$] pure dephasing interactions can result only from contributions to $V_{\rm SB}$ which are nonlinear in x. In general, however, even linear system-bath coupling may lead to pure dephasing. For example, for the Morse oscillator ($V(x)=D\{\exp[-(x-\bar{x})/\alpha]-1\}^2$) it may be shown that

$$x_0(J)=\bar{x}-\alpha\ln\frac{2\lambda^2}{1+\lambda}\,, \tag{54}$$

where

$$\lambda=1-J/(2MD\alpha^2)^{1/2} \tag{55}$$

so that even linear (in x) coupling leads to a pure dephasing contribution. In order to apply these observations to the problem described by Eqs. (47)—(49) we follow the procedures of Refs. 1(b) (paper I) and 1(h). Defining

$$G(x)=\int^x dx'f(x')\,, \tag{56a}$$

$$\dot{G}=f\dot{x} \tag{56b}$$

and transforming $G(x,\dot{x})\rightarrow G(J,\phi)$, $\dot{G}(x,\dot{x})\rightarrow\dot{G}(J,\phi)$ according to

$$G(J,\phi)=\sum_{n=-\infty}^{\infty}G_n(J)e^{in\phi}\,, \tag{57a}$$

$$\dot{G}(J,\phi)=\sum_{n=-\infty}^{\infty}in\omega(J)G_n(J)e^{in\phi} \tag{57b}$$

[note that $\dot{G}_n(J)=in\omega(J)G_n(J)$], we get [in analogy to Eqs. (29) and (39) of I]

$$\dot{J}=M\sum_{n=-\infty}^{\infty}\sum_{n'=-\infty}^{\infty}nn'\omega G_nG_{n'}\hat{Z}_{n'}e^{i(n+n')\phi}+iR(t)\sum_{n=-\infty}^{\infty}nG_ne^{in\phi}\,, \tag{58}$$

$$\dot{\phi}=\omega+M\sum_{n=-\infty}^{\infty}\sum_{n'=-\infty}^{\infty} in'\omega G_{n'}\frac{dG_n}{dJ}\hat{Z}_{n'}e^{i(n+n')\phi}$$
$$-R(t)\sum_{n=-\infty}^{\infty}\frac{dG_n}{dJ}e^{in\phi}\ . \tag{59}$$

Here $\hat{Z}_n(\omega)=\int_0^{\infty}dt\, e^{in\omega t}Z(t)dt$. Obviously the $n=0$ terms contribute to $\dot{\phi}$ but not to J and may be therefore viewed as sources of pure dephasing.

In Ref. 1(h) Eqs. (58) and (59) were shown to lead to the following Fokker-Planck equation for $P(J,\phi,t)$:

$$\frac{P(J,\phi,t)}{t}=\frac{\partial}{\partial J}\left[\epsilon(J)\left[k_BT\frac{\partial}{\partial J}+\omega(J)\right]P(J,\phi,t)\right]$$
$$+\Gamma(J)\frac{\partial^2 P}{\partial\phi^2}-\Omega(J)\frac{\partial p}{\partial\phi}\ , \tag{60}$$

where $\epsilon(J)$, $\Gamma(J)$, and $\Omega(J)$ are given by equations similar to (22)—(24) in which x_n is replaced by G_n everywhere. In particular

$$\Gamma(J)=2Mk_BT\sum_{n=1}^{\infty}\left[\frac{dG_n}{dJ}\right]^2\hat{Z}_n^c(\omega(J))+Mk_BT\gamma\left[\frac{dG_0}{dJ}\right]^2 \tag{61}$$

[where $\gamma=\hat{Z}^c(\omega=0)$] may be identified, by inspecting Eq. (60), with the phase diffusion rate. In accord with the discussion above we also identify the $n=0$ term

$$\Gamma_0(J)=Mk_BT\gamma\left[\frac{dG_0}{dJ}\right]^2 \tag{62}$$

with the pure dephasing rate. In many cases, in particular when the characteristic system frequency is much larger than the typical frequency (e.g., the Debye frequency) of the thermal bath, γ is much larger than $Z_n^c(\omega)$, $n>0$, and consequently

$$\Gamma_0 \gg |\Gamma(J)-\Gamma_0|\ .$$

Similarly Γ_0 is often much larger than the energy relaxation rate (measured by[1(h)] $[(d\ln E/dt)_{T=0}=\omega D(E)/E]$). These points are further discussed in Appendix A.

As is well known from other (mostly quantum-mechanical) treatments, when the dephasing is much faster than the energy relaxation rate, it constitutes the major contribution to the linewidth of the spectrum associated with the well motion. This point is demonstrated for the present classical model in Appendix B, and serves to further justify the identification of Γ_0 with the pure dephasing rate.

In Sec. IV we investigate the motion of a generalized (non-Markovian) Brownian particle under the effect of external periodic force. The assumption that the driving field is chaotic in the sense defined in Sec. II is dropped. Instead we assume that pure dephasing is so fast that at any time t the phase distribution is uniform [i.e., $P(\phi)=1/2\pi$]. This assumption is satisfied in the low-friction limit in the absence of the external field, for the coarse-grained time scale on which the energy diffusion equation is valid. In the presence of the external driving field it implies that the phase diffuses due to the thermal interactions faster than the field manages to restore it. To obtain a more quantitative criterion for the validity of this assumption we anticipate results of Sec. IV and consider the following simplified form for the time evolution of the phase, ϕ, in the presence of an external field which adds a term $\mu(x)\cos(\omega_R t)$ to the Hamiltonian

$$\dot{\phi}=\omega-\sqrt{\Gamma_0}\rho(t)-\frac{d\mu_1}{dJ}\cos\phi\cos(\omega_R t)\ , \tag{63}$$

where $\langle\rho\rangle=0$ and $\langle\rho(0)\rho(t)\rangle=2\delta(t)$; Γ_0 is given by (62) and $\mu_1(J)$ is a coefficient in the expansion $\mu(x)=\sum_n\mu_n(J)e^{in\phi}$. Equation (62) is a simplified version of Eq. (73b) where we keep only the lowest-order terms in the driving force and in the thermal noise and where we focus on the Markovian limit.

Equation (63) may be simplified and solved for small detuning ($\omega\simeq\omega_R$). This is also the case most relevant to our issue because we expect the external field to oppose the thermal erasure of the phase most efficiently near resonance. Denoting $x=\phi-\omega_R t$ we derive in Appendix C the steady-state distribution $P_{SS}(x)$ associated with Eq. (63)

$$P_{SS}(x)=Ne^{(\eta x-\mu_1'\sin x)/\Gamma_0}\times\left[1+(e^{-2\pi\eta/\Gamma_0}-1)\times\frac{\int_0^{x}d\bar{x}\exp(-\eta\bar{x}-\mu_1'\sin\bar{x})/\Gamma_0}{\int_0^{2\pi}d\bar{x}\exp(-\eta\bar{x}-\mu_1'\sin\bar{x})/\Gamma_0}\right], \tag{64}$$

where $\eta=\omega-\omega_R$ and $\mu_1'=d\mu_1'/dJ$. $P_{SS}(x)$ is easily shown to be periodic in x as should be. In Fig. 1 we show this function (at resonance, $\eta=0$) for different values of the parameters μ_1'/Γ_0. When this parameter is much smaller than 1 we indeed get a practically uniform distribution. In what follows we take the condition for a uniform phase distribution to be

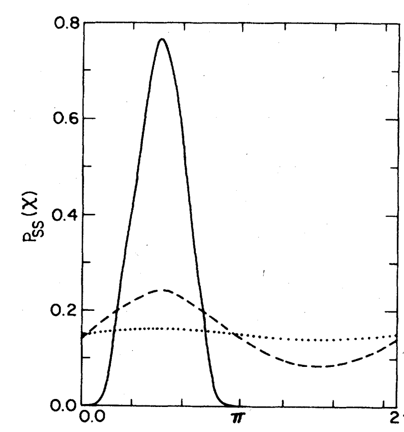

FIG. 1. $P_{SS}(x)$ plotted as a function of x for the resonance case, $\eta=0$. Solid line, $\mu_1'/\Gamma_0=4.0$; dashed line, $\mu_1'/\Gamma_0=2.5$; dotted line, $\mu_1'/\Gamma_0=0.0625$.

$$\frac{d\mu_1}{dJ}/\Gamma_0 \ll 1 , \tag{65}$$

where we should keep in mind that $d\mu_1/dJ$ and Γ_0 appear here as leading orders of more complicated expressions.

IV. EXTERNAL OSCILLATING FORCE IN THE FAST-THERMAL-DEPHASING LIMIT

We now focus on the model described by Eqs. (47)—(49), now supplemented by the presence of an oscillating force

$$\dot{x} = v , \tag{66}$$

$$\dot{v} = -\frac{1}{M}\frac{dV(x)}{dx} - f(x(t))\int_0^t d\tau Z(1-\tau)f(x(\tau))v(\tau) + \frac{1}{M}f(x(t))R(t) - \frac{1}{M}\frac{d\mu(x)}{dx}\cos\psi , \tag{67}$$

$$\dot{\psi} = \omega_R . \tag{68}$$

Here we have taken the particle—external-field interaction to be represented by the potential $\mu(x)\cos\psi$ where ψ satisfies (68) and where $\mu(x)$ is some function of the particle's coordinate which also depends on the external field intensity. Transforming to the particle's action-angle variables (J,ϕ) associated with the deterministic undamped motion in the potential $V(x)$ (using the procedure of paper I), Eqs. (66)—(68) are replaced by

$$\dot{J} = -M\sum_{n,n'} inG_n(t)e^{in\phi(t)}\int_0^t d\tau Z(t-\tau)e^{in'\phi(\tau)}\dot{G}_{n'}(\tau) + \cos\psi\sum_n in\mu_n e^{in\phi} + R(t)\sum_n inG_n e^{in\phi} , \tag{69}$$

$$\dot{\phi} = \omega(J) + M\sum_{n,n'}\left[\frac{dG_n}{dJ}\right]_t e^{in\phi(t)} \times \int_0^t d\tau Z(t-\tau)e^{in'\phi(\tau)}\dot{G}_{n'}(\tau) - \cos\psi\sum_n \frac{d\mu_n}{dJ}e^{in\phi} - R(t)\sum_n \frac{dG_n}{dJ}e^{in\phi} , \tag{70}$$

$$\dot{\psi} = \omega_R , \tag{71}$$

where G_n is defined by (52) and where[1(h)] $\dot{G}_n = in\omega(J)G_n$. As in paper I we focus on the low-friction and short environment-correlation-time limit

$$\omega(J) \gg \tau_c^{-1} \gg \gamma = \int_0^\infty dt\, Z(t) \tag{72a}$$

and in addition we assume that the external force is not too strong so that[22]

$$\omega(J) \gg d\mu_n/dJ \quad (\text{all } n) . \tag{72b}$$

In addition we shall make use of the fast-dephasing assumption (65).

Our aim is to reduce Eqs. (69)—(71) to a Smoluchowski-type equation for the action variable J. To this end we first follow the procedure which leads to Eqs. (29) and (30) of paper I, eliminating the memory terms. This leads to

$$\dot{J} = -\sum_{n,n'} B_{nn'}^{(J)}e^{i(n+n')\phi} + \cos\psi\sum_n \mu_n^{(J)}e^{in\phi} + R(t)\sum_n \sigma_n^{(J)}e^{in\phi} , \tag{73a}$$

$$\dot{\phi} = \omega + \sum_{n,n'} B_{nn'}^{(\phi)}e^{i(n+n')\phi} - \cos\psi\sum_n \mu_n^{(\phi)}e^{in\phi} - R(t)\sum_n \sigma_n^{(\phi)}e^{in\phi} , \tag{73b}$$

where

$$\sigma_n^{(J)} = inG_n , \tag{74a}$$

$$\sigma_n^{(\phi)} = \frac{dG_n}{dJ} , \tag{74b}$$

$$B_{nn'}^{(J)} = -nn'M\omega G_n G_{n'}\hat{Z}_{n'} , \tag{75a}$$

$$B_{nn'}^{(\phi)} = in'M\omega\frac{dG_n}{dJ}G_{n'}\hat{Z}_{n'} , \tag{75b}$$

$$\mu_n^{(J)} = in\mu_n , \tag{76a}$$

$$\mu_n^{(\phi)} = \frac{d\mu_n}{dJ} , \tag{76b}$$

and where, as usual, $\hat{Z}_n = \int_0^\infty dt\, e^{in\omega t}Z(t)$. Note that $\sigma_n^{(J)}$ and $\sigma_n^{(\phi)}$ are equivalent to σ_n and μ_n while $B_{nn'}^{(J)}$ and $B_{nn'}^{(\phi)}$ are equivalent to $B_{nn'}$ and $C_{nn'}$ of paper I.

To reduce Eq. (73) to a Fokker-Planck equation we use an iteration procedure similar to that used to perform a similar task in paper I. Here we give only an outline of the derivation. The iteration equation's equivalent to (40) and (41) of I are

$$\Delta J_t^{(l)}(\tau) = \epsilon\omega\tau + \sum_n \int_t^{t+\tau} du\, R(u)\left[\sigma_n^{(J)} + \frac{d\sigma_n^{(J)}}{dJ}\Delta J(u)\right]\exp\{in[\phi + \Delta\phi^{(l-1)}(u)]\}$$

$$+ \left[\tfrac{1}{2}e^{i\psi}\int_t^{t+\tau} du\, e^{i\omega_R(u-t)}\sum_n\left[\mu_n^{(J)} + \frac{d\mu_n^{(J)}}{dJ}\Delta J^{(l-1)}(u)\right]\exp\{in[\phi + \Delta\phi^{(l-1)}(u)]\} + \text{c.c.}\right] , \tag{77}$$

$$\Delta\phi_t^{(l)}(\tau)=\int_t^{t+\tau}du\left[\omega+\frac{d\omega}{dJ}\Delta J^{(l-1)}(u)\right]-\sum_n\int_t^{t+\tau}du\,R(u)\left[\sigma_n^{(\phi)}+\frac{d\sigma_n^{\phi}}{dJ}\Delta J^{(l-1)}(u)\right]\exp\{in[\phi+\Delta\phi^{(l-1)}(u)]\}-\left[\tfrac{1}{2}e^{i\psi}\int_t^{t+\tau}du\,e^{i\omega_R(u-t)}\sum_n\left[\mu_n^{(\phi)}+\frac{d\mu_n^{(\phi)}}{dJ}\Delta J^{(l-1)}(u)\right]\exp\{in[\phi+\Delta\phi^{(l-1)}(u)]\}+\text{c.c.}\right]. \quad (78)$$

In these equations nonargumented variables are evaluated at time t (i.e., the first term on the right-hand side of (78) is $\int_t^{t+\tau}du\{\omega[J(t)]+(d\omega/dJ)_t\Delta J^{(l-1)}(u)\}$ and the increments $\Delta J_t(\tau)$ and $\Delta\phi_t(\tau)$ are defined by $\Delta J_t(\tau)=J(t+\tau)-J(t)$, etc.). Also in Eq. (77) we have anticipated a simplifying feature (see Appendix B of I) by replacing a term involving $\sum_{n,n'}B_{nn'}^{(J)}$ by $\epsilon\omega\tau$ where

$$\epsilon(J)=M\sum_{n=-\infty}^{\infty}n^2|G_n|^2\hat{Z}_n=2M\sum_{n=1}^{\infty}n^2|G_n|^2\hat{Z}_n^{(c)},$$

while in (78) we have neglected the term $\sum_{n,n'}B_{nn'}^{(\phi)}e^{i(n+n')\phi}$ which is small compared to ω (or else we could have added its diagonal part to ω as a small frequency shift).

The iteration procedure used here differs from that in I by one important detail: there we have followed the usual procedure of expanding $e^{in\Delta\phi}=1+in\Delta\phi$ using the fact that the coarse-graining time τ of Eqs. (77) and (78) is much smaller than a typical oscillator period $(2\pi\omega^{-1})$. Here this is still true; however, following the same procedure we encounter potentially divergent denominators like $(\omega_R-n\omega)^{-1}$. This divergence has the same source as that encountered in treating coupling between a (classical or quantum) oscillator and the radiation field in low order. To eliminate it we are forced to keep $\Delta\phi$ in the exponent, which leads in the course of the iteration procedure to the appearance of $\exp(\int R)$ terms (R is the random force). The latter are handled by cumulant averaging techniques which are easily carried out since R is a Gaussian variable. It should be noted however that while in I we perform a systematic iteration procedure to third order (while showing that higher-order contributions are negligible), here we keep higher-order contributions when necessary to renormalize ω (thus avoiding the divergencies mentioned above). We should also note that in addition to neglecting terms small in the thermal interactions (using $\gamma/\omega\ll 1$) as well as contributions of order τ^l, $l>1$ to $\langle\Delta J\rangle$ and $\langle\Delta J^2\rangle$ (see paper I) we also use repeatedly the inequality (72b). Furthermore, in performing the averages that lead to $\langle\Delta J\rangle$ and $\langle\Delta J^2\rangle$ we use the strong dephasing approximation, assuming that $\phi(t)$ is uniformly distributed in $0,\ldots,2\pi$.[23] We thus take an average over the ϕ distribution $[(2\pi)^{-1}\int_0^{2\pi}d\phi]$ together with an average over the thermal noise $R(t)$.

With these additional complications the evaluation of $\langle\Delta J\rangle$ and $\langle\Delta J^2\rangle$ proceeds as in paper I and will not be reproduced here. We only give in Appendix D an example (evaluating the average of one of the terms that contribute to $\langle\Delta J\rangle$) in order to demonstrate our method and approximations. The final result is

$$\langle\Delta J\rangle=\tau\left[-\omega(J)\epsilon(J)+k_BT\frac{d\epsilon}{dJ}+k_BT\frac{d\lambda}{dJ}+\eta(J)\right], \quad (79)$$

$$\langle\Delta J^2\rangle=2k_BT\tau[\epsilon(J)+\lambda(J)], \quad (80)$$

where

$$\epsilon(J)=2M\sum_{n=1}^{\infty}n^2|G_n|^2\hat{Z}_n^c, \quad (81)$$

$$\lambda(J)=\frac{1}{2}\sum_{n=1}^{\infty}\frac{n^4|\mu_n|^2\Gamma_0/k_BT}{(n\omega-\omega_R)^2+n^4\Gamma_0^2}, \quad (82)$$

$$\eta(J)=\frac{1}{2}\sum_{n=1}^{\infty}n^4|\mu_n|^2\frac{d\Gamma_0}{dJ}\frac{n^4\Gamma_0^2-(n\omega-\omega_R)^2}{[n^4\Gamma_0^2+(n\omega-\omega_R)^2]^2}, \quad (83)$$

and where $\Gamma_0(J)$ is given by (62). The corresponding Fokker-Planck equation for $P(J)$ is

$$\frac{\partial P(J,t)}{\partial t}=\frac{\partial}{\partial J}\left[(\epsilon+\lambda)\left[k_BT\frac{\partial}{\partial J}+\frac{\epsilon\omega-\eta}{\epsilon+\lambda}\right]P(J,t)\right]. \quad (84)$$

Transforming from J to E and defining

$$D(E)=(\epsilon\omega-\eta)_{J=J(E)}, \quad (85)$$

$$\beta(E)=(k_BT)^{-1}\left[\frac{\epsilon-\eta/\omega}{\epsilon+\lambda}\right]_{J=J(E)} \quad (86)$$

we get

$$\frac{\partial P(E,t)}{\partial t}=\frac{\partial}{\partial E}\left[D(E)\left[\frac{1}{\beta(E)}\frac{\partial}{\partial E}+1\right]P(E,t)\right]. \quad (87)$$

Equation (87) is identical in form to Eq. (31) and leads to equations of the form (33), (35), (36), and (39) for the steady-state distribution and flux, the equilibrium distribution, and the steady-state escape rate. In particular, the latter is given by

$$k=\left[\int_0^{E_B}dE\frac{\beta(E)}{\omega(E)D(E)}[P_{\rm eq}(E)]^{-1}\int_0^E dE'P_{\rm eq}(E')\right]^{-1}, \quad (88)$$

where $\beta(E)$ and $D(E)$ are given by Eqs. (85) and (86) and where

$$P_{\rm eq}(E)=\frac{A_1}{\omega(E)}\exp\left[-\int_0^E dE'\beta(E')\right]. \quad (89)$$

[A_1 is determined by normalization. Note that its value does not affect the result (88).]

In the absence of the external field ($\eta=\lambda=0$) Eq. (89) reduces to the purely thermal result $P_{\rm eq}(E) \sim [A_1/\omega(E)]e^{-E/k_BT}$. In the presence of the field we expect the effective temperature $\beta^{-1}(E)$ to be larger than the actual temperature. From (86) this implies

$$\frac{\epsilon-\eta/\omega}{\epsilon+\lambda}<1\ , \tag{90}$$

i.e.,

$$\omega>\eta/\lambda\ . \tag{91}$$

Using Eqs. (82) and (83) it is easy to show that $\eta(J)/\lambda(J)<k_BT\,d\ln\Gamma_0(J)/dJ$. Thus a sufficient condition for (91) to hold is

$$\omega>k_BT\frac{d\ln\Gamma_0}{dJ} \tag{92}$$

or, integrating over J,

$$E(J)>k_BT\ln\frac{\Gamma_0(J)}{\Gamma_0(0)} \tag{93}$$

which holds for all relevant $E(J)$. Thus indeed the field-induced effective temperature is larger than the actual temperature.

More explicit results may be obtained by treating special cases. Consider, for example, a harmonic oscillator $[(1/M)dV/dx=\omega^2x$ in (67)] with

$$f(x)=1+\frac{x}{\xi} \tag{94}$$

and

$$\mu(x)=ax\ . \tag{95}$$

The parameter ξ measures the strength of the multiplicative stochastic noise while a is proportional to the amplitude of the external field. Using $G(x)=x+x^2/2\xi$ we get

$$G_0=\frac{J}{2M\omega\xi},\ G_{\pm1}=\left[\frac{J}{2M\omega}\right]^{1/2},\ G_{\pm2}=\frac{J}{4M\omega\xi}\ , \tag{96}$$

$G_n=0$ for $|n|>2$.

From (62) we have

$$\Gamma_0=\frac{k_BT\gamma}{4M\omega^2\xi^2} \tag{97}$$

so that η of Eq. (83) vanishes. Using for the noise correlation function $Z(t)=(\gamma/\tau_c)e^{-t/\tau_c}$, i.e., $\hat{Z}_n^c=\gamma/[1+(n\omega\tau_c)^2]$, we obtain from (81) (using also $E=\omega J$)

$$\epsilon=\frac{E\gamma}{\omega^2}\left[\frac{1}{1+(\omega\tau_c)^2}+\frac{E}{2M\omega^2\xi^2}\frac{1}{1+(2\omega\tau_c)^2}\right] \tag{98}$$

and from (82)

$$\lambda=\frac{\Gamma_0}{2k_BT}\frac{a^2E/2M\omega^2}{(\omega-\omega_R)^2+\Gamma_0^2}\ . \tag{99}$$

Equations (85) and (86) yield

$$D(E)=\epsilon\omega\ ,$$
$$\beta(E)=\frac{1}{k_BT}\left[1+\frac{\lambda}{\epsilon}\right]^{-1}\ . \tag{100}$$

In particular, using (98)—(100) leads after a little algebra to

$$\int_0^E dE'\beta(E')=\frac{1}{k_BT}\left[E-c_1\ln\left[1+\frac{E}{c_1+c_2}\right]\right]\ , \tag{101}$$

where

$$c_1=\frac{a^2[1+(2\omega\tau_c)^2]}{8M}\frac{1}{(\omega-\omega_R)^2+\Gamma_0^2} \tag{102}$$

while

$$c_2=\frac{k_BT\gamma}{2\Gamma_0}\frac{1+(2\omega\tau_c)^2}{1+(\omega\tau_c)^2}\ . \tag{103}$$

The presence of the oscillating external field obviously make the higher-energy states more probable. This effect is characterized by a resonance behavior and is maximized for $\omega=\omega_R$. Unlike the result obtained in the absence of dephasing (Ref. 6 and Sec. I) the equilibrium distribution does not peak sharply at any $E>0$ but, more like the chaotic external-field case (Sec. II) may be characterized by an effective temperature. This can be clearly seen at resonance, where for energy sufficiently small $[E/(c_1+c_2)\ll 1]$ Eq. (101) may be approximated by

$$\int_0^E dE'\beta(E')\simeq\frac{1}{k_BT}\left[1-\frac{c_1}{2(c_1+c_2)}\right]E\ , \tag{104}$$

so that $P_{\rm eq}(E)$ is a Boltzmann-like distribution characterized by the temperature

$$T_{\rm eff}=\left[1-\frac{c_1}{2(c_1+c_2)}\right]^{-1}T\ . \tag{105}$$

V. NUMERICAL RESULTS AND DISCUSSION

In what follows we shall refer to the model reviewed in Sec. I (no dephasing) as model I, to the external-field dephasing model of Sec. II as model II and to the intrinsic dephasing model of Sec. IV as model III. These three models are compared in the Markovian limit for a harmonic well (with a cutoff at $E=E_B$) in Figs. 2 and 3. Figure 2 shows the escape rate k [expressed in terms of $\ln(k/k_0)$ where k_0 is the corresponding rate in the absence of the external field] as a function of the external-field frequency ω_R (in units of the well frequency ω). In Fig. 3 the equilibrium distribution $P_{\rm eq}(E)$ is displayed as function of E for the three models in the presence of the external field and for the purely thermal system (no external field). The parameters used in these calculations are $\gamma=0.01$, the phase-damping rate in model II $\Gamma=0.025$, the intrinsic dephasing rate in model III $\Gamma_0=0.025$, all in units of the harmonic-well frequency ω, $E_B=8.0$ (in units of k_BT), and the external-force amplitude $a=0.03$ [in

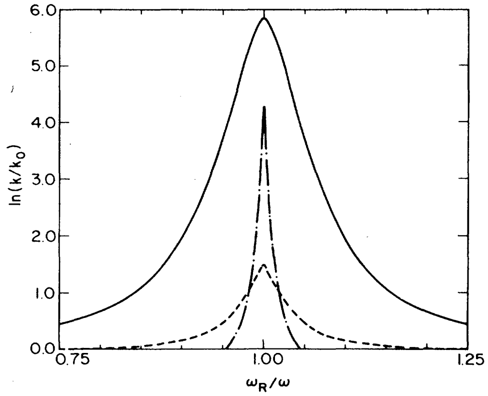

FIG. 2. $\ln(k/k_0)$ vs ω_R/ω for the harmonic well models discussed in Sec. I (no dephasing, dashed-dotted line), Sec. II (external field dephasing, solid line), and Sec. IV (intrinsic dephasing, dashed line). Parameters used in this calculation are given in the text.

units of $(k_B T M^2)^{1/2}$]. The results of Fig. 3 are for the resonance situation $\omega_R=\omega$. The full lines in these figures correspond to model II, the dashed lines to model III, and the dotted-dashed lines to model I. The dotted line in Fig. 3 corresponds to the purely thermal (no driving) case. The following points should be noted about these results.

(1) In Fig. 2 the resonances are symmetric and centered about the harmonic well frequency. Their widths are determined by the choice of γ and Γ or Γ_0. All these features are characteristic of the harmonic situation.

(2) The sharpest resonance in Fig. 2 is that associated with model I. This results from the absence of a dephasing contribution to the width in this model.

(3) The resonance enhancement of the escape rate in model III is seen in Fig. 2 to be substantially smaller than that of model II. The cause for this difference may be traced to the difference between the effects of the external-field dephasing and the intrinsic dephasing associated with the multiplicative random noise. The latter affects the higher-energy states of the oscillator much more strongly than the former, effectively taking these states out of resonance with the driving field, thus reducing the resonance enhancement.

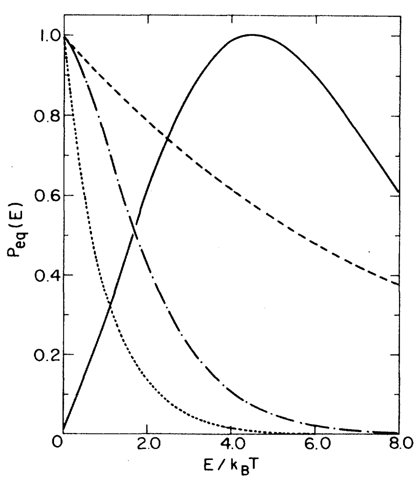

FIG. 3. $P_{\text{eq}}(E)$ vs E/k_BT for the harmonic well models displayed in Fig. 2. Line notation is as in Fig. 2. The dotted line represents the distribution obtained in the absence of the driving field. Parameters used in the calculation are given in the text.

(4) While the three models yield qualitatively similar results for k/k_0 (namely, resonance enhancement of the escape rate), the effects of external driving on the energy distribution function (Fig. 3) lead to qualitatively different results: As discussed in preceding sections and as seen in Fig. 3, in the absence of dephasing an external periodic force leads to a peak at finite energy in $P_{\text{eq}}(E)$ while in the presence of strong dephasing this peak is absent and the enhancement is best associated with a (generally E-dependent) effective temperature.

(5) The rates displayed in Fig. 2 were calculated from the low-friction expressions [(5) for model I, (39) for model II, and (88) for model III]. Alternatively we could invoke the transition-rate-theory assumption (valid for intermediate thermal relaxation rate within the well) that the rate is proportional to the equilibrium population $P_{\text{eq}}(E_B)$ near the threshold energy, and use Eqs. (3), (36), and (89) for models I, II, and III, respectively. As seen below, both assumptions lead to qualitatively similar results. (For sufficiently deep wells these results should become identical; see paper I.)

Resonant enhancement of activated rate processes is a common phenomenon in multiphoton photochemistry where in many cases we have enough spectroscopical data to make a quantum treatment possible. Recently Devoret *et al.*[8] have observed resonant activation of the transition from the zero- to the finite-voltage state of a current-driven Josephson junction induced by a weak microwave perturbation. This phenomenon provides a potentially important tool for determination of the junction parameters. For such an application, however, the origin of friction and the possibility of dephasing have to be considered.

The main qualitative difference between the results displayed in Fig. 2 and the experimental results of Ref. 8 lies in the details of the line shape. The resonance observed in Ref. 8 is asymmetric with a tail on the low-frequency side, and its peak is shifted to the red relative to the estimated well-bottom frequency. Both these features have been previously observed in multiphoton dissociation of large molecules and are associated with the anharmonic nature of the well, namely the existence of lower frequencies associated with higher-energy states in the well.

In Figs. 4 and 5 we present calculations done with an anharmonic model—a Morse oscillator. The choice of this model makes the calculations described in Secs. II and IV particularly easy because the functions $E(J)$ and $\omega(J)$ and the coefficients of the expansion $x=\sum_n x_n(J)e^{in\phi}$ are known analytically (see, e.g., paper I). Focusing on model III we have thus taken in Eq. (67)

$$V(x)=E_B(e^{-2x/\alpha}-2e^{-x/\alpha}) ,$$

$$f(x)=1+\frac{x}{\xi} ,$$

and

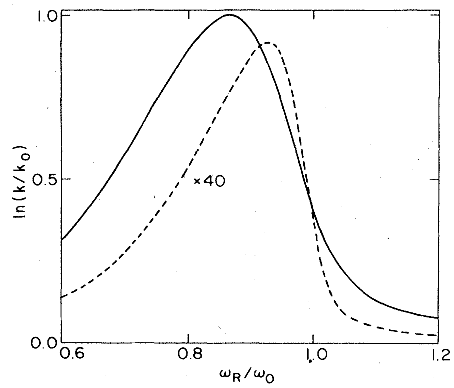

FIG. 4. $\ln(k/k_0)$ vs ω_R/ω_0 for model III with Morse potential. Parameters used for the solid line are $a\alpha/k_BT=0.45$, $E_B/kT=8.0$, $\xi/\alpha=0.42$, and $\gamma=0.0225$. Dashed line is obtained when the external-field strength a is reduced by a factor of 10.

$$\mu(x)=ax \ ,$$

The parameter α is related to the well bottom frequency ω_0 by $\alpha=(M\omega_0^2/2E_B)^{1/2}$ and the energy dependence of the frequency is $\omega(E)=\omega_0(-E/E_B)^{1/2}$. The parameters defining the Morse potential were taken so that the parameters E_B ($=8kT$) and ω_0 were the same as those given in Ref. 8. In Figs. 4 and 5 we show the results obtained for k/k_0 [k and k_0 calculated from Eq. (88)] and for $P_{\text{eq}}(E_B)/P_{\text{eq}}^0(E_B)$ [calculated from Eq. (89)] where k_0 and P_{eq}^0 correspond to the purely thermal (no driving) limit. As discussed above, the former result corresponds to the extreme low-friction limit while the latter describes the rate ratio expected under conditions of fast thermal relaxation in the well so that the escape rate is proportional to the equilibrium population near the threshold energy. In both cases we have chosen the parameters a, ξ, and γ to yield the experimentally observed yield and red shift.

The results of both calculations are in qualitative agreement with the observations: the red shift of the peak and the asymmetric red tail are clearly observed. The resonance width is about twice as large as the width observed experimentally. At the temperature considered, this width is not very sensitive to the choice of γ and ξ and is mostly determined by the frequency spectrum of the potential (Morse in our case, cosine in the actual system). This is seen by the narrowing observed when the external force is reduced (dashed lines of Figs. 4 and 5). In this case the resonance is governed by the field-induced dynamics closer to the well bottom. This leads to a smaller red shift of the peak and to a narrow line. The asymmetry becomes more pronounced because the main contribution (near ω_0) now lies to the blue of all other frequencies.

Similar calculations on model II yield qualitatively similar results.

We should end this discussion by noticing that there is no evidence that dephasing effects play an important role in the experiment of Devoret *et al.*[8] Such an evidence can be obtained in principle by monitoring the well distribution function [point (4) above] but this is not a feasible measurement. The microwave source used in Ref. 8 is likely to be very coherent. On the other hand, intrinsic dephasing always exists in an anharmonic system even in the absence of a multiplicative noise[24] and when the external driving is weak enough (as may be the case here), may lead to the strong dephasing limit discussed here and in Sec. IV.

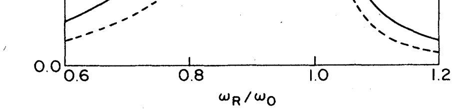

FIG. 5. $\ln[P_{\text{eq}}(E_B)/P_{\text{eq}}^0(E_B)]$ vs ω_R/ω_0 for the same model as in Fig. 4. Parameters used for the solid line are $a\alpha/k_BT=0.35$, $E_B/kT=8.0$, $\xi/\alpha=0.25$, and $\gamma=0.0225$. Dashed line is obtained when $a\alpha/k_BT$ is reduced by a factor of 10.

VI. CONCLUSIONS

We have treated a classical non-Markovian model of activated rate process in the presence of an external periodic force. In particular we found that dephasing effects (both those associated with the driving field and intrinsic dephasing associated with the coupling between the system and its thermal environment) may lead to qualitative changes in the escape process. Our results may be relevant to recent experiments[8] on microwave-induced resonant enhancement of the activation out of the zero-voltage state of a current-biased Josephson junction.

ACKNOWLEDGMENTS

This research was supported by the U.S.-Israel Binational Science Foundation and by the Israel Academy of Sciences. We thank Professor D. Bergman and Professor E. Ben Jacob for very useful discussions. One of us (A.N.) also thanks Dr. R. Landauer and Dr. G. Schon for helpful discussions, and Professor P. Hänggi for useful correspondence.

APPENDIX A

Here we consider the case of a damped harmonic oscillator with a simple multiplicative noise

$$\ddot{x}+\omega^2x+a^2x(t)\int_0^t d\tau Z(t-\tau)x(\tau)v(\tau)+\frac{a}{M}x(t)R(t) \ , \tag{A1}$$

where a is a constant of dimension of inverse length and where Z and R satisfy Eq. (49). For $Z(t)=(\gamma/\tau)e^{-t/\tau}$ we get $\hat{Z}_n^c(\omega)=\gamma(1+n^2\omega^2\tau^2)^{-1}$. Using

$x_n=(J/2M\omega)^{1/2}\delta_{|n|,1}$ and $G(x)=\frac{1}{2}ax^2$ we get [from Eq. (61)]

$$\Gamma=\Gamma_0+\frac{k_BTa^2}{16M\omega^2}Z_2^c(\omega) \tag{A2}$$

and [from Eq. (62)]

$$\Gamma_0=\frac{k_BTa^2}{8M\omega^2}\gamma \ . \tag{A3}$$

Similarly, from Eq. (51) we obtain

$$D(E)=8M\omega\,|\,G_2\,|^2Z_2^c(\omega)=\frac{a^2E^2}{2M\omega^3}Z_2^c(\omega) \ . \tag{A4}$$

The energy relaxation rate is related to $D(E)$ by[1(h)]

$$\left[\frac{1}{E}\frac{dE}{dt}\right]_{T=0}=\frac{\omega D(E)}{E}=\frac{Ea^2}{2M\omega^2}Z_2^c(\omega) \ . \tag{A5}$$

We see from Eqs. (A2) and (A3) that the pure dephasing rate Γ_0 is larger than the "improper" contribution to the dephasing provided that $\gamma \gg Z_2^c(\omega)$ (i.e., $\omega\tau \gg 1$) and is larger than the energy relaxation rate provided that

$$\frac{k_BT}{E}\frac{\gamma}{Z_2^c(\omega)} \gg 1 \ . \tag{A6}$$

In most chemical rate processes k_BT/E is of the order of 10^{-1}—10^{-2} while $\gamma/Z_2^c(\omega)$ is of the order of $\omega\tau$. If we take for ω a typical value of a molecular frequency (10^{-1} eV) and for τ^{-1} a typical cutoff frequency for liquids ($\sim 10^{-3}$ eV) we get $\omega\tau\sim 10^2$. This is however an underestimate. For ω larger than the cutoff frequency ω_D of the medium $Z_n^c(\omega)$ falls off with ω much faster than ω^{-2}: a more realistic estimate is given by the "energy-gap law"

$$Z_n^c(\omega)\sim\gamma\exp(-n\omega/\omega_D) \ . \tag{A7}$$

Using (A7) we find that the inequality (A6) is satisfied for most realistic systems.

APPENDIX B

Here we show that when the dephasing rate is much faster than the energy relaxation rate it constitutes the major contribution to the spectral width of the motion in the well. The spectrum is taken to be the cosine Fourier transform of the correlation function

$$\begin{aligned} C(t)&=\langle x(0)x(t)\rangle \\ &=\sum_n\sum_{n'}x_n(J)x_{n'}(J)\langle e^{in\phi(0)}e^{in'\phi(t)}\rangle \ , \end{aligned} \tag{B1}$$

where in the rhs of (B1) we have assumed that the energy E (and the action J) do not change on the time scale of interest. The time evolution of the phase ϕ is governed by the ϕ-dependent part of Eq. (60)

$$\frac{\partial P(\phi,t)}{\partial t}=-\Omega\frac{\partial P}{\partial\phi}+\Gamma\frac{\partial^2P}{\partial\phi^2} \ . \tag{B2}$$

Taking $\Delta\phi=\phi-\Omega t$ we find for the probability distribution $\overline{P}(\Delta\phi)$ of $\Delta\phi$

$$\frac{\partial\overline{P}(\Delta\phi,t)}{\partial t}=\Gamma\frac{\partial^2\overline{P}(\Delta\phi,t)}{\partial(\Delta\phi)^2} \ . \tag{B3}$$

This leads to

$$P(\Delta\phi,t\mid\Delta\phi_0,0)=\frac{1}{2\pi}\sum_{n=-\infty}^{\infty}\exp[in(\Delta\phi-\Delta\phi_0)-n^2\Gamma t] \ . \tag{B4}$$

Using (B4) we get

$$\begin{aligned} \langle e^{in\phi_0}e^{in'\phi(t)}\rangle &=e^{in'\Omega t}\langle e^{in\Delta\phi_0}e^{in'\Delta\phi(t)}\rangle \\ &=\frac{1}{2\pi}e^{in'\Omega t}\int_0^{2\pi}d(\Delta\phi_0)\int_0^{2\pi}d(\Delta\phi)P(\Delta\phi,t\mid\Delta\phi_0)e^{in\Delta\phi_0+in'\Delta\phi} \\ &=\delta_{n,-n'}e^{-in\Omega t-n^2\Gamma t} \ , \end{aligned} \tag{B5}$$

and from Eqs. (B1) and (B5) we get

$$L(\omega)=\int_0^\infty d\omega\cos(\omega t)C(t)\sim\sum_n\frac{|\,x_n\,|^2}{(\omega-n\Omega)^2+n^2\Gamma} \ . \tag{B6}$$

APPENDIX C

Here we obtain the steady-state distribution associated with Eq. (63). It is convenient to rewrite Eq. (63) in terms of two variables, ϕ and ψ:

$$\begin{aligned} \dot\phi&=\omega-\frac{d\mu_1}{dJ}\cos\phi\cos\psi-\sqrt{\Gamma_0}\rho(t) \ , \\ \dot\psi&=\omega_R \ . \end{aligned} \tag{C1}$$

For $\omega\sim\omega_R$ we replace $\cos\phi\cos\psi$ by its rotating-wave approximation

$$\cos\phi\cos\psi\rightarrow 2\cos(\phi-\psi) \ . \tag{C2}$$

Transforming to the detuning variable $x=\phi-\psi$ we get (setting $\eta=\omega-\omega_R$)

$$\dot x=\eta-2\frac{d\mu_1}{dJ}\cos x-\sqrt{\Gamma_0}\rho(t) \tag{C3}$$

which is equivalent to the Fokker-Planck equation

$$\begin{aligned} \frac{\partial P(x,t)}{\partial t}=&-\frac{\partial}{\partial x}\left[\left[\eta-2\frac{d\mu_1}{dJ}\cos x\right]P(x,t)\right] \\ &+\Gamma_0\frac{\partial^2P(x,t)}{\partial x^2} \ . \end{aligned} \tag{C4}$$

A general steady-state solution of Eq. (C4) is obtained from ($\mu_1'=d\mu_1/dJ$)

$$(\eta-2\mu_1'\cos x)P_{SS}(x)+\Gamma_0\frac{P_{SS}(x)}{\partial x}=C\ , \tag{C5}$$

where C is any constant. This yields

$$P_{SS}(x)=N\exp\left[\frac{\eta x-2\mu_1'\sin x}{\Gamma_0}\right]\times\left[1+\frac{C}{\Gamma_0}\int_0^x dx'\exp\left[-\frac{\eta x'-2\mu_1'\sin x'}{\Gamma_0}\right]\right]. \tag{C6}$$

N is determined from the normalization requirement. The constant C is determined by imposing periodic boundary conditions

$$P_{SS}(x)=P_{SS}(x+2\pi)\ . \tag{C7}$$

This leads to

$$C=\Gamma_0\frac{e^{-2\pi\eta/\Gamma_0}-1}{\int_0^{2\pi}dx\exp[-(\eta x-\mu_1'\sin x)/\Gamma_0]} \tag{C8}$$

which, inserting into (C6), yields Eq. (64).

APPENDIX D

Here we evaluate the term $k_BT(d\epsilon/dJ)$ appearing in Eq. (79), and comment on the calculation of the other terms appearing there. Repeatedly using Eqs. (77) and (78) in the iteration procedure discussed above we arrive at the following expression for $\Delta J_t^{(l)}(\tau)$ where l is very large:

$$\begin{aligned}\Delta J_t(\tau)=&-\tau\omega(J)\epsilon(J)+\sum_n\int_0^\tau du\,R(u)\sigma_n^{(J)}\exp[in\phi(u)]\\&+\sum_n\int_0^\tau du\,R(u)\frac{d\sigma_n^{(J)}}{dJ}\exp[in\phi(u)]\left[\int_0^u ds\,R(s)f^{(J)}(s)+\frac{1}{2}\left[e^{i\psi}\int_0^u ds\,e^{i\omega_R s}g^{(J)}(s)+\text{c.c.}\right]\right]\\&+\frac{1}{2}\left[e^{i\psi}\int_0^\tau du\,e^{i\omega_R u}\sum_n\mu_n^{(J)}\exp[in\phi(u)]+\text{c.c.}\right]\\&+\frac{1}{2}\left\{e^{i\psi}\int_0^\tau du\,e^{i\omega_R u}\sum_n\frac{d\mu_n^{(J)}}{dJ}\exp[in\phi(u)]\right.\\&\qquad\left.\times\left[\int_0^u ds\,R(s)f^{(J)}(s)+\frac{1}{2}\left[e^{i\psi}\int_0^u ds\,e^{i\omega_R s}g^{(J)}(s)+\text{c.c.}\right]\right]+\text{c.c.}\right\}\ ,\end{aligned} \tag{D1}$$

where

$$\phi(u)=\phi+\omega u-\int_0^u ds\,R(s)F(u,s)-\frac{1}{2}\left[e^{i\psi}\int_0^u ds\,e^{i\Omega s}G(u,s)+\text{c.c.}\right]\ , \tag{D2}$$

$$F(u,s)=f^{(\phi)}(s)-(u-s)\frac{d}{dJ}f^{(J)}(s)\ , \tag{D3a}$$

$$G(u,s)=g^{(\phi)}(s)-(u-s)\frac{d}{dJ}g^{(J)}(s)\ , \tag{D3b}$$

$$f^{(W)}(s)=\sum_n\sigma_n^{(W)}\exp[in\phi(s)]\ , \tag{D4a}$$

$$g^{(W)}(s)=\sum_n\mu_n^{(W)}\exp[in\phi(s)] \tag{D4b}$$

and where W stands for either J or ϕ and $\sigma_n^{(\phi)}$, $\sigma_n^{(J)}$, $\mu_n^{(\phi)}$, and $\mu_n^{(J)}$ are given in Eqs. (74) and (76), respectively. Note that Eq. (D2) is a self-consistent equation for $\phi(u)$ which may be solved by iterations. However, one may expect the iterations to converge very rapidly in the low-friction limit (i.e., $\omega\gg F,G$) and in fact we are going to use

$$\phi(s)\simeq\phi+\omega s \tag{D5}$$

in Eqs. (D4) whenever the divergencies of the type $(\omega_R-n\omega)^{-1}$ (which were discussed in Sec. IV) are not expected to appear. In particular we invoke this approximation for Eq. (D2) but we do not use it for $f^{(W)}(s)$ and $g^{(W)}(s)$ appearing explicitly in Eq. (D1).

Next we calculate the second S_2 and the third S_3 terms of Eq. (D1). For the second term we have

$$S_2=\sum_n\sigma_n^{(J)}\int_0^\tau du\exp\left[in\phi+in\omega u-\frac{in}{2}\left[e^{i\psi}\int_0^u ds\,e^{i\omega_R s}G(u,s)+\text{c.c.}\right]\right]\left\langle R(u)\exp\left[-in\int_0^u ds R(s)F(u,s)\right]\right\rangle\ , \tag{D6}$$

using the properties of Gaussian variables we get

$$\left\langle R(u)\exp\left[-in\int_0^u ds\,R(s)F(u,s)\right]\right\rangle=-in\int_0^u ds\,F(u,s)\langle R(u)R(s)\rangle\exp\left[\frac{n^2}{2}\int_0^u ds\,F(u,s)\int_0^u ds'F(u,s')\right]\langle R(s)R(s')\rangle\ . \tag{D7}$$

Using Eq. (D5) for $F(u,s)$ we get

$$\int_0^u ds\, F(u,s) \int_0^u ds' F(u,s') \langle R(s)R(s')\rangle$$

$$= Mk_BT \sum_{n,n'} \exp[i(n+n')(\phi+\omega u)] \int_0^u ds \left[\frac{d}{dJ}[G_n \exp(-in\omega s)]\right] \int_0^u ds \left[\frac{d}{dJ}[G_{n'} \exp(-in'\omega s')]\right] Z(s-s')$$

$$= 2uMk_BT \sum_n \left[\left[\frac{dG_n}{dJ}\right]^2 \hat{Z}_n(\omega) + 2G_n \frac{dG_n}{dJ}\frac{d\hat{Z}_n(\omega)}{dJ} + G_n^2 \frac{d^2\hat{Z}_n(\omega)}{dJ^2} \right]$$

$$= 2u\Gamma_0 + 4uMk_BT \sum_{n=1}^{\infty} \left[\left[\frac{dG_n}{dJ}\right]^2 \hat{Z}_n(\omega) + \frac{d(G_n^2)}{dJ}\frac{d\hat{Z}_n(\omega)}{dJ} + G_n^2 \frac{d\hat{Z}_n(\omega)}{dJ} \right] . \tag{D8}$$

In the fast dephasing limit we assume that the term $2u\Gamma_0$ is much bigger than the rest of the rhs of Eq. (D8). Thus we only consider this term whenever the double integral of the left-hand side of Eq. (D8) appears. Equation (D6) may now be written as

$$S_2 = -iMk_BT \sum_n n\sigma_n^{(J)} \int_0^\tau du \exp\left[in\phi + in\omega u - n^2\Gamma_0 u - \frac{in}{2}\left[e^{i\psi} \int_0^u ds\, e^{i\omega_R s} G(u,s) + \text{c.c.} \right]\right] \int_0^u ds\, F(u,s) Z(u-s) . \tag{D9}$$

Next we neglect the terms containing $G(u,s)$ and Γ_0 with respect to ω in the spirit of the weak-interaction approximation and we find

$$S_2 = -iMk_BT \sum_n n\sigma_n^{(J)} \int_0^\tau du \exp(in\phi + in\omega u) \int_0^u ds\, Z(u-s) \sum_{n'} \exp(in'\phi + in'\omega s) \left[\sigma_{n'}^{(\phi)} - (u-s)\frac{d}{dJ}\sigma_{n'}^{(J)}\right] , \tag{D10}$$

where we have used Eqs. (D3a), (D4a), and (D5) for $F(u,s)$. Taking the average over the phase and using

$$\frac{1}{2\pi} \int_0^{2\pi} d\phi\, e^{i(n+n')\phi} = \delta_{n,-n'} \tag{D11}$$

we obtain from Eq. (D10)

$$S_2 = -iMk_BT \sum_n n\sigma_n^{(J)} \left[\sigma_{-n}^{(\phi)} \int_0^\tau du \int_0^u ds \exp[in\omega(u-s)] Z(u-s) - \sigma_{-n}^{(J)} \frac{d\omega}{dJ} \int_0^\tau du \int_0^u ds \exp[in\omega(u-s)](u-s) Z(u-s) \right] . \tag{D12}$$

Using $\sigma_{-n}^{(\phi)} = \sigma_n^{(\phi)} = dG_n/dJ$ and $-\sigma_{-n}^{(J)}(d\omega/dJ)\exp[in\omega(u-s)](u-s) = G_n(d/dJ)\exp[in\omega(u-s)]$ we rearrange Eq. (D12) to get

$$S_2 = Mk_BT \sum_n n^2 G_n \frac{d}{dJ}\left[G_n \int_0^\tau du \int_0^u ds \exp[in\omega(u-s)] Z(u-s) \right]$$

$$\cong \tau Mk_BT \sum_n n^2 G_n \frac{d}{dJ}[G_n \hat{Z}_n(\omega)] . \tag{D13}$$

Turning now to the third term of Eq. (D1), using the Gaussian properties of the stochastic variables and taking only average terms contributing to the lowest order in the interactions, we find for this term

$$S_3 = \sum_n \frac{d\sigma_n^{(J)}}{dJ} \int_0^\tau du \exp\left[in\phi + in\omega u - \frac{in}{2}\left[e^{i\psi} \int_0^u ds\, e^{i\omega_R s} G(u,s) + \text{c.c.} \right] - n^2\Gamma_0 u \right] \times \int_0^u ds \sum_{n'} \sigma_{n'}^{(J)} \exp(in'\phi + in'\omega s) \langle R(u)R(s)\rangle . \tag{D14}$$

Ignoring as before the terms containing $G(u,s)$ and Γ_0 relative to ω and taking the average over the phase, we obtain from Eq. (D14)

$$S_3 = Mk_B T \sum_n \frac{d\sigma_n^{(J)}}{dJ} \sigma_{-n}^{(J)} \int_0^\tau du \int_0^u ds \exp[in\omega(u-s)] Z(u-s) = \tau M k_B T \sum_n n^2 \frac{dG_n}{dJ} G_n \hat{Z}_n(\omega) . \quad \text{(D15)}$$

Combining Eqs. (D13) and (D15) we obtain

$$S_2 + S_3 = \tau M k_B T \frac{d}{dJ} \left[\sum_n n^2 G_n^2 Z_n(\omega) \right] = 2\tau M k_B T \frac{d}{dJ} \left[\sum_{n=1}^{\infty} n^2 G_n^2 \hat{Z}_n(\omega) \right] = \tau k_B T \frac{d\epsilon(J)}{dJ} , \quad \text{(D16)}$$

where Eq. (81) has been used to get the last equality.

Note that besides the weak-coupling limit [$\omega \gg \hat{Z}_n(\omega)$ and $\omega \gg d\mu_n/dJ$] we need not use in the derivations described any other approximation. Thus we could safely neglect Γ_0 compared to ω. The situation is different however when we evaluate the rest of the summations appearing in Eq. (D1). For example, in the fourth term $\exp[in\phi(u)]$ does not appear alone as before but it is rather accompanied by $\exp(\pm i\omega_R u)$. In this case we are not allowed to neglect Γ_0 relative to $\omega_R - n\omega$ since the last quantity may be very small under resonance conditions. Taking this into account, in addition to the weak-interaction approximation, we arrive at Eqs. (79) and (80).

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