Resonant Directed Diffusion in Nonadiabatically Driven Systems

M. I. Dykman,[1] H. Rabitz,[2] V. N. Smelyanskiy,[1] and B. E. Vugmeister[2]
[1]*Department of Physics and Astronomy, Michigan State University, East Lansing, Michigan 48824*
[2]*Department of Chemistry, Princeton University, Princeton, New Jersey 08544*
(Received 5 May 1997)

We show that a high-frequency field may *resonantly* decrease the activation energy of escape from a potential well. For systems in spatially periodic potentials, the effect is different for the transitions in opposite directions, which gives rise to resonantly directed diffusion (DD). DD arises in both asymmetric and symmetric periodic potentials. It depends exponentially strongly on the field magnitude, and its direction can be controlled by varying the field spectrum. [S0031-9007(97)03880-5]

PACS numbers: 05.40.+j, 02.50.–r, 05.20.–y, 05.60.+w

Much attention has been given recently to the occurrence of unidirectional motion of systems which fluctuate in a periodic potential. This motion is superimposed on diffusion and arises if the system is away from thermal equilibrium. It is substantially due to fluctuations and can be viewed as a directed diffusion (DD). The effect was initially considered for potentials asymmetric within the period (ratchets) [1,2]. It then became clear that DD may arise also in symmetric potentials [3]. The interest in DD is stimulated by its relevance to a broad class of processes, from atomic diffusion in crystals, solid surfaces, and optical lattices to phase diffusion in Josephson junctions and the motion of proteins along biopolymers. For the most part, the analysis of DD has been limited to systems driven by nonequilibrium noise or by thermal noise and an *adiabatically slow* driving field where the diffusion rate is determined by the instantaneous value of the field [1–3]. The adiabatic picture does not apply if the field period τ_F is less than the characteristic relaxation time of the system $t_{\rm rel}$. One might expect that DD will be "averaged out" with the decreasing τ_F. We show that, on the contrary, the rate of DD may display *resonant peaks* as a function of the field frequency.

For small fluctuation intensity, the rate of directed diffusion j in a periodic potential $U(q)$ with period l is determined by the difference between the probabilities W^+, W^- of the transitions from a potential minimum to the right and to the left,

$$j = l(W^+ - W^-). \tag{1}$$

The analysis of resonant DD requires general nonadiabatic theory of escape rates $W^{\pm}$. It should differ qualitatively from the theories where the effect of a high-frequency field is described in terms of field-enhanced diffusion over energy [4]. Such diffusion gives rise to the correction to the distribution over energy which is quadratic in the field amplitude, whereas the rates W^+ and W^- would still coincide with each other.

In the present paper we provide a nonadiabatic theory of directed diffusion for the nontrivial and important case of moderately strong driving fields. We show that, even for the amplitude of forced vibrations about the minima of the potential $U(q)$ being *small* compared to the period l, the probabilities of transitions between potential wells depend on the field magnitude *exponentially* strongly. We also show that both the direction and speed of DD depend on spectral characteristics of the driving field.

Interwell transitions require large fluctuations and happen occasionally, for small fluctuation intensity. The probability densities for the system to move in a transition along different paths are very different, and with overwhelming probability it moves along the most probable escape path (MPEP); see [5–7].

Qualitatively, the effect of a moderately strong driving field on the escape rates can be thought of in terms of the work the field does while the system is moving along the MPEP. This work changes the activation energy of escape S_a. Clearly, the change $\delta S_a^{\pm}$ may be resonantly large, for the appropriate field frequencies. Since the MPEPs for the escape to the right and to the left are different so may also be the changes $\delta S_a^{\pm}$. This happens if a periodically driven system lacks the spatiotemporal symmetry $q \to -q, t \to t + \tau_F/2$ considered earlier in the context of quasienergy states of driven quantum systems [8]; for DD induced by an additive driving force the criterion was earlier found in [3]. We note that the quantities $\delta S_a^{\pm}$ are *time independent* in the nonadiabatic theory, in contrast to periodic modulation of the activation energies that occurs in the adiabatic theory [1,3].

The analysis of MPEPs in classical systems driven by Gaussian noise has much in common with the instanton theory [9]. Both types of trajectories provide the minima to certain functionals which can be associated with mechanical action $S[q(t)]$. The minimum of the action S gives the logarithm of the escape probability W. In the problem of activated escape of thermal nonequilibrium systems, the form of the functional $S[q]$ depends on the system dynamics and the properties of the noise [5,6].

To illustrate the occurrence of DD and the possibility of controlling it we will consider the simplest but nontrivial

 0031-9007/97/79(7)/1178(4)$10.00

case of a Brownian particle with a unit mass, which is moving in a periodic potential $U(q)$ and is additionally driven by a time-dependent force $F(t)$:

$$\Pi(\ddot{q},\dot{q},q;t)=\xi(t),\quad \Pi=\ddot{q}+2\Gamma\dot{q}+U'(q)-F(t),$$
$$\langle\xi(t)\xi(t')\rangle=4\Gamma kT\delta(t-t'),\quad U(q+l)=U(q). \tag{2}$$

By generalizing the results known for stationary systems [5–7] one can show that, for small noise intensities, i.e., for low temperatures T, the escape probabilities $W^{\pm}$ are described by the activation law $W^{\pm}\propto\exp(-S_a^{\pm}/kT)$. The activation energies $S_a^{\pm}$ are given by the solution of the following variational problem:

$$S_a^{\pm}=\min S[q(t)],\quad S[q(t)]=\frac{1}{8\Gamma}\int_{-\infty}^{\infty}dt\,\Pi^2(\ddot{q},\dot{q},q;t),$$
$$q(t)\to q_{\rm st}(t)\quad \text{for } t\to-\infty, \tag{3}$$
$$q(t)\to q_{\rm u}^{\pm}(t)\quad \text{for } t\to\infty.$$

Here, $q_{\rm st}(t)$ is the stable state of forced vibrations about a given minimum of $U(q)$ in the absence of noise, and $q_u^{+}(t)$, $q_{\rm u}^{-}(t)$ are the unstable periodic states of the vibrations about the local maxima of $U(q)$ to the right and to the left from this minimum, respectively.

The variational problem (3) defines the most probable paths $q^{\pm}(t)$ for escape over the nearest right or left potential maximum. These paths start in the vicinity of the stable periodic state for $t\to-\infty$ and approach the appropriate unstable periodic state as $t\to\infty$.

In the absence of periodic force, i.e., for $F(t)=0$, fluctuations of the Brownian particle satisfy detailed balance conditions. In this case the MPEPs are time-reversed paths from the corresponding maxima to the minimum of the potential in the absence of noise [10]. They satisfy the equation

$$q(t)_{F=0}\equiv Q(t),\qquad \ddot{Q}^{\pm}(t)-2\Gamma\dot{Q}^{\pm}(t)+U'(Q^{\pm})=0 \tag{4}$$

with appropriate boundary conditions. One can see from Eq. (3) that, for the solution (4), the activation energies $S_a^{+}=S_a^{-}$ are given by the difference ΔU of the periodic potential $U(q)$ in its maximum and minimum, $\Delta U=U(q_{\rm u}^{\pm})-U(q_{\rm st})$. The characteristic duration of motion along the paths $Q^{\pm}(t)$ is of the order of the characteristic relaxation time of the system $t_{\rm rel}=\max\{\Gamma^{-1},\Gamma/U''(q_{\rm st})\}$.

The effect of the periodic field on the escape rates depends substantially on the interrelation between $t_{\rm rel}$ and the field period τ_F. For $\tau_F\gg t_{\rm rel}$ the transitions can be considered as occurring for a given instantaneous value of the field $F(t)$ in a biased potential $U(q)-F(t)q$, and the activation energies $S_a^{\pm}$ periodically depend on time.

In the opposite case of nonadiabatic driving, $\tau_F<t_{\rm rel}$, once the system has approached the vicinity of an unstable periodic state $q_{\rm u}^{\pm}(t)$ as a result of a large fluctuation, it will stay there for a time that exceeds τ_F and perform small fluctuations with an amplitude $\propto(kT)^{1/2}$. Eventually it will either come back to the stable state from which the large fluctuation started or make a transition to another stable state, with a probability $\sim1/2$. Therefore the transition probability is not synchronized with the field and does not depend on time, to logarithmic accuracy.

In contrast to the transition probabilities, the MPEPs *are* synchronized with the field. It is clear from the variational problem (3) that, for a periodic field $F(t)$, there is a set of the MPEPs $q^{\pm}(t-n\tau_F)$ which repeat each other with the period τ_F. This set is discrete, in contrast to the set of the MPEPs in the absence of driving $Q^{\pm}(t-t_c)$ (4) which is continuous (t_c can be arbitrary).

For a moderately strong driving field, the leading-order correction to S_a is linear in F. It can be found from Eq. (3) by integrating along the unperturbed trajectory $Q^{\pm}(t-t_c)$ (4) the term in Π^2 which is linear in $F(t)$.

It is clear from the above arguments that, since for a driven system there is generically only one MPEP per period of $F(t)$, the value of t_c should be such that the trajectory $Q^{\pm}(t-t_c)$ was close to it. Therefore generically there is only one $t_c[\mathrm{mod}\,\tau_F]$ that gives the "right" escape trajectory $Q^{\pm}(t-t_c)$. It was shown in [11] that the corresponding t_c is the one that provides *the minimum* to the field-dependent correction $\delta S^{\pm}(t_c)$ to the activation energy evaluated along the path $Q^{\pm}(t-t_c)$. To first order in F we have

$$S_a^{\pm}=\Delta U+\delta S_a^{\pm},\qquad \delta S_a^{\pm}=\min_{t_c}\delta S^{\pm}(t_c),$$
$$W^{\pm}(F)=W^{\pm}(0)\exp[-\delta S_a^{\pm}/kT], \tag{5}$$

$$\delta S^{\pm}(t_c)=\int_{-\infty}^{\infty}dt\,\chi^{\pm}(t-t_c)F(t),\quad \chi^{\pm}(t)=-\dot{Q}^{\pm}(t). \tag{6}$$

The quantities $\chi^{\pm}(t)$ describe changes of the *logarithms* of the field-dependent escape rates $W^{\pm}(F)$, which are linear in the driving field F, and can be called *logarithmic susceptibilities* [11]. Logarithmic susceptibilities are expressed in terms of the characteristics of the system in the absence of driving, as are also "conventional" susceptibilities. It is seen from Eq. (5) that the linear correction to the escape rate corresponds to the *maximal* work done by the force $F(t)$ on the escaping particle.

For a monochromatic field $F(t)=F_1\cos\Omega t$ the corrections $\delta S_a^{\pm}$ take the form

$$\delta S_a^{\pm}=-|\hat{\chi}^{\pm}(\Omega)|F_1,\qquad \hat{\chi}^{\pm}(\Omega)=\int dt\,e^{i\Omega t}\chi^{\pm}(t). \tag{7}$$

In the static limit $\Omega\to0$ the spectral densities $\hat{\chi}^{\pm}(\Omega)$ approach $|q_{\rm u}^{\pm}-q_{\rm st}|$ (cf. Fig. 1), and Eqs. (5) and (7) give the minimal adiabatic activation energies over the period. In general, the shape of $|\hat{\chi}^{\pm}(\Omega)|$ depends on interrelation between the friction coefficient Γ and the characteristic frequencies of intrawell vibrations of the Brownian particle in the absence of noise. For small damping, the function

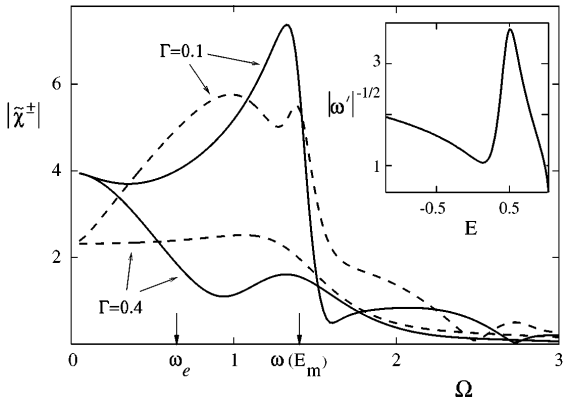

FIG. 1. The spectral densities $|\chi^+(\Omega)|$ (solid curves) and $|\chi^-(\Omega)|$ (dashed curves) for the modulation of the rate of escape to the right and to the left from the minima of the potential $U(q) = \sin q + 0.3\sin(2q + 0.4)$. The upper and lower sets of curves refer to $\Gamma = 0.1$ and $\Gamma = 0.4$, respectively. Inset: The function $|\omega'(E)|^{-1/2}$ for the same potential.

$|\hat{\chi}^{\pm}(\Omega)|$ displays sharp peaks, and in fact it may have multiple-peak structure as shown in Fig. 1. This structure can be understood if one writes the velocity on the MPEP in the form

$$\dot{Q}^{\pm}(t) \equiv P^{\pm}(t) = \mathrm{Re}\sum_{n>0} P_n[E^{\pm}(t)]\exp[-in\varphi^{\pm}(t)],$$
$$\dot{E}^{\pm} \approx 2\Gamma\omega(E^{\pm})I(E^{\pm}), \qquad \dot{\varphi}^{\pm} \approx \omega(E^{\pm}), \tag{8}$$

where $\omega(E)$ is the eigenfrequency of the vibrations with an energy E in the absence of dissipation, $I(E)$ is the action for these vibrations, and $P_n(E)$ is the amplitude of the nth overtone of the momentum P. We dropped the terms $\propto\Gamma$ in the equation for the phase and fast-oscillating terms in the equation for the energy.

Resonant contributions to $\hat{\chi}^{\pm}(\Omega)$ come from vibrations with energies $E_n(\Omega)$ for which $n\omega(E_n) = \Omega$. In the range of Ω where the contribution from an overtone with a given n is much larger than from other overtones, we may approximate $\hat{\chi}^{\pm}(\Omega)$ by the function $\hat{\chi}_n(\Omega)$ calculated from Eqs. (7) taking into account in (8) the term with one n only. Evaluating the integral over time in (7) by the steepest descent method, we obtain

$$|\hat{\chi}_n(\Omega)| = \left[|P_n(E)|\left|\frac{2\Gamma n I(E)}{\pi}\frac{d\omega^2}{dE}\right|^{-1/2}\right]_{E=E_n(\Omega)}. \tag{9}$$

The amplitude $|P_n(E)|$ is usually largest for the fundamental mode $n = 1$, and the peak of $|\hat{\chi}_1(\Omega)|$ should be dominating in the spectrum. In a general case where, for E close to its minimal value $E_m = U(q_{\mathrm{st}})$ the frequency $\omega(E)$ decreases and $|\omega'(E)|$ increases with the increasing energy E, this peak is sharp and strongly asymmetric,

$$|\hat{\chi}_1(\Omega)| = A(1 - Bx)\theta(x), \quad x = \omega(E_m) - \Omega, \tag{10}$$
$$A = [\pi/2\Gamma|\omega'|]^{1/2}, \quad 2\omega' B = -(|P_2/P_1|^2)' - \omega''/\omega'$$

(here, the derivatives over E are evaluated for $E = E_m$). For $\Gamma = 0.1$, such a toothlike peak is seen in Fig. 1. Its shape near the maximum is well described by Eq. (10). Similar peaks arise also from the overtones with $n > 1$, but their widths are larger and the heights are much smaller; cf. the spectrum for $\Omega > \omega(E_m)$ in Fig. 1.

It follows from (9) that $|\hat{\chi}_n(\Omega)|$ may also display peaks near the frequencies $n\omega(E)$ for which $|\omega'(E)|^{-1/2}$ has a narrow maximum (see inset in Fig. 1); the occurrence of the peaks in conventional susceptibility due to vanishing of $\omega'(E)$ was considered in [12].

In the range where the contributions from several overtones are substantial it is necessary to allow for their *interference*. The phase shifts between resonating overtones with $n\omega(E_n) = \Omega$ are different for $|\hat{\chi}^+(\Omega)|$ and $|\hat{\chi}^-(\Omega)|$ in the case of an asymmetric potential $U(q)$. The phases of the MPEPs for the escape to the right and to the left differ by π, and in particular the cross-terms from the overtones $n = 1$ and $n = 2$ in $|\hat{\chi}^+(\Omega)|$, $|\hat{\chi}^-(\Omega)|$ have opposite signs. For the potential in Fig. 1 we have $\omega(E_m) \approx 2\omega_e$. This gives rise to strong interference of the peak of $\hat{\chi}_1(\Omega)$ near $\omega(E_m)$ and the peak of $\hat{\chi}_2(\Omega)$ near $2\omega_e$. As a result, the peak of $|\chi^+(\Omega)|$ near $\omega(E_m)$ is enhanced, and there occurs a dip in $|\chi^-(\Omega)|$ near $2\omega_e$. Detailed results on the spectroscopy of the exponential modulation of escape rates will be given elsewhere. The structure of the spectrum is smeared with the increasing dissipation.

It is seen from Fig. 1 and from Eqs. (5) and (7) that the field-induced modulation of the transition rates $W^{\pm}$ in an asymmetric potential is most effective if the system is underdamped and the field is resonantly tuned. Since the direction of the diffusion (1) depends on which of the rates is larger, it is possible to change it just by varying the field frequency.

In the case of a symmetric potential $U(q)$ the MPEPs $Q^{\pm}(t)$ are mirror symmetrical, $\dot{Q}^+(t) = -\dot{Q}^-(t)$, and DD does not arise for a monochromatic field. This is a consequence of the spatiotemporal symmetry of the driven system. If $F(t + \tau_F/2) = -F(t)$, which is true for a monochromatic field, it follows from Eq. (6) that $\delta S^+(t_c) = \delta S^-(t_c - \tau_F/2)$. Clearly, the activation energies given by the minimal values of $\delta S^+(t_c)$, $\delta S^-(t_c)$ coincide with each other, and therefore $W^+ = W^-$.

In general, for a periodic nonmonochromatic field with Fourier components F_n we have from (6) and (7)

$$\delta S^{\pm}(t_c) = \sum_n \hat{\chi}^{\pm}(n\Omega)F_{-n}\exp(in\Omega t_c). \tag{11}$$

For a symmetric potential we have $\chi^+(\omega) = -\chi^-(\omega)$ and $\delta S^-(t_c) = -\delta S^+(t_c)$. If the force $F(t)$ has *both* odd *and* even harmonics, then the field-induced terms in the activation energy $\delta S_a^+ \equiv \min \delta S^+(t_c)$ and $\delta S_a^- = -\max \delta S^+(t_c)$ are not equal to each other. Respectively, the escape rates $W^{\pm}$ are different and there arises DD.

Each of the functions $\delta S^{\pm}(t_c)$ (11) may have several local minima. The physically interesting quantities $\delta S_a^{\pm}$

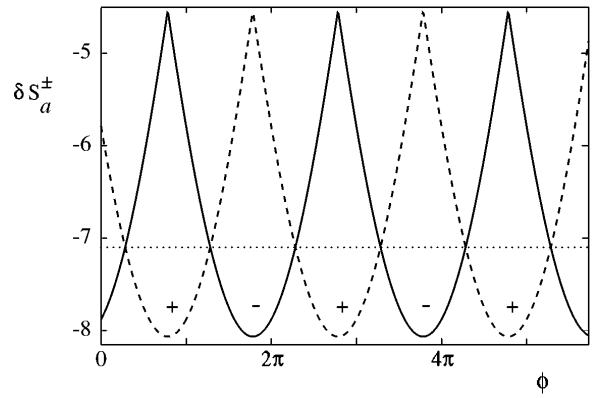

FIG. 2. The activation energies δS_a^- (solid curve) and δS_a^+ (dashed curve) for the escape to the right and to the left from the minima of the symmetrical potential $U(q) = \sin q$ in the field $F(t) = 2\cos\Omega t + 0.73\cos(2\Omega t + \phi)$, for $\Gamma = 0.1$, $\Omega = 0.44$. The ranges where the dc current flows to the right and to the left are shown by "+" and "–," respectively. The dotted line shows the value of $\delta S_a^{\pm}$ where the current reversal occurs.

are determined by the absolute minima of the respective $\delta S^{\pm}(t_c)$. Therefore, with the varying spectrum of the field $F(t)$, the field dependence of $\delta S_a^{\pm}$ will change discontinuously from that for one local minimum of $\delta S^{\pm}(t_c)$ to that for another. This is similar to what happens at a first-order phase transition, with $\delta S_a^{\pm}$ and t_c being analogs of the free energy and the order parameter.

In Fig. 2 we illustrate the activation energies $\delta S_a^{\pm}$ and the onset of DD in a symmetric potential for a field with two harmonics, $F(t) = F_1 \exp(i\Omega t) + F_2 \exp(2i\Omega t) +$ cc. It follows from Eqs. (5) and (11) that the quantities $\delta S_a^{\pm} \equiv \delta S_a^{\pm}(\phi)$ are periodic in the phase shift between the field harmonics $\phi = \arg(F_2/F_1)$, and also that $\delta S_a^+(\phi) = \delta S_a^-(\phi + \pi)$. Therefore the curves $\delta S_a^+(\phi), \delta S_a^-(\phi)$ intersect each other twice within the period 2π. At each intersection the direction of diffusion changes to that for which the activation energy is smaller. The rate of DD is maximal where $\delta S_a^{\pm}(\phi)$ are at their minima. The abrupt changes in the slopes of $\delta S_a^{\pm}(\phi)$ are also seen in Fig. 2. However, they occur for $\delta S_a^+(\phi)$ in the range of ϕ where $\delta S_a^+(\phi) > \delta S_a^-(\phi)$ [and similarly for $\delta S_a^-(\phi)$]. We note that, for the chosen frequency $\Omega \sim \omega(E_m)/2$, the maximal values of $|\delta S_a^{\pm}|$ in Fig. 2 exceed those in the adiabatic approximation by a factor of 3. Equations (5) and (6) make it possible to optimize the shape of the periodic field for control purposes.

In conclusion, we have shown that, even for comparatively weak driving fields, the rate of activated escape from a potential well, and, consequently, the rate of directed diffusion in a spatially periodic potential, depend exponentially on the field amplitude. The rates display resonant behavior as functions of the period of the field. This behavior is determined by the system dynamics far from equilibrium positions in the absence of driving and can be used to investigate this dynamics. The direction of DD can be controlled by varying the field spectrum.

The work of M. I. D. was supported by NSF Grant No. PHY-9722057.

[1] M. Magnasco, Phys. Rev. Lett. **71**, 1477 (1993).
[2] R. D. Astumian and M. Bier, Phys. Rev. Lett. **72**, 1766 (1994); J. Prost, J.-F. Chauwin, L. Peliti, and A. Ajdari, Phys. Rev. Lett. **72**, 2652 (1994); C. Doering, W. Horsthemke, and J. Riordan, Phys. Rev. Lett. **72**, 2984 (1994); M. M. Millonas and M. I. Dykman, Phys. Lett. A **183**, 65 (1994); M. M. Millonas, Phys. Rev. Lett. **74**, 10 (1995); F. Marchesoni, Phys. Rev. Lett. **77**, 2364 (1996); A.-L. Barabási *et al.* (to be published).
[3] A. Ajdari, D. Mukamel, L. Peliti, and J. Prost, J. Phys. I (France) **4**, 1551 (1994); M. C. Mahato and A. M. Jayannavar, Phys. Lett. A **209**, 21 (1995); D. R. Chialvo and M. M. Millonas, Phys. Lett. A **209**, 26 (1995); Phys. Rev. E **53**, 2239 (1996).
[4] A. I. Larkin and Yu. N. Ovchinnikov, J. Low Temp. Phys. **63**, 317 (1986); B. I. Ivlev and V. I. Mel'nikov, Phys. Lett. A **116**, 427 (1986); S. Linkwitz and H. Grabert, Phys. Rev. B **44**, 11 888 (1991); **44**, 11 901 (1991).
[5] M. I. Freidlin and A. D. Wentzell, *Random Perturbations in Dynamical Systems* (Springer Verlag, New York, 1984); M. I. Dykman and M. A. Krivoglaz, Sov. Phys. JETP **50**, 30 (1979); A. J. Bray and A. J. McKane, Phys. Rev. Lett. **62**, 493 (1989); M. I. Dykman, Phys. Rev. A **42**, 2020 (1990).
[6] R. Graham, in *Noise in Nonlinear Dynamical Systems*, edited by F. Moss and P. V. E. McClintock (Cambridge University Press, Cambridge, 1989), Vol. 1, p. 225.
[7] R. S. Maier and D. L. Stein, Phys. Rev. Lett. **71**, 1783 (1993); **77**, 4860 (1996).
[8] M. S. Sherwin, in *Quantum Chaos*, edited by G. Casati and B. Chirikov (Cambridge University Press, Cambridge, 1995), p. 209.
[9] J. S. Langer, Ann. Phys. **41**, 108 (1967); S. Coleman, Phys. Rev. D **15**, 2929 (1977).
[10] M. I. Dykman, M. A. Krivoglaz, and S. M. Soskin, in Ref. [6], Vol. 2, p. 347; see M. Marder, Phys. Rev. Lett. **74**, 4547 (1995), and references therein, for the discussion of optimal fluctuational paths in thermal equilibrium systems.
[11] M. I. Dykman, H. Rabitz, V. N. Smelyanskiy, and B. E. Vugmeister, Report No. cond-mat/970436.
[12] S. M. Soskin, Physica (Amsterdam) **180A**, 386 (1992).