

Fluctuations, Escape, and Nucleation in Driven Systems: Logarithmic Susceptibility

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We analyze the probabilities of large infrequent fluctuations in *nonadiabatically* driven systems. In a broad range of the driving field magnitudes, the logarithm of the fluctuation probability is linear in the field, and the response can be characterized by a logarithmic susceptibility (LS). We evaluate the activation energies for nucleation, with account taken of the field-induced lifting of time and spatial degeneracy of instantonlike nucleation trajectories. LS for nucleation in systems with nonconserved order parameter is shown to be a nonmonotonic function of ω and \mathbf{k} . [S0031-9007(97)04328-7]

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As first pointed out by Debye [1], systems with coexisting metastable states may strongly respond to the driving field through the effect of the field on the probabilities of transitions between the states. For classical systems, the transition probability W is described by the activation law, $W \propto \exp(-R/kT)$. Even a relatively weak field h , for which the change of the activation energy $\Delta R \propto h$ is much less than R , can strongly affect W provided $|\Delta R|/kT$ is not small. This effect has been investigated for various systems and has attracted much attention recently in the context of stochastic resonance [2]. Similarly, in extended systems driving fields can exponentially strongly affect nucleation probabilities. So far most results on modulation of transition probabilities have been obtained for adiabatically slow driving, where the change of the field over the relaxation time of the system is small and transitions occur “instantaneously,” for a given value of the field and for the corresponding value of the activation barrier.

The physical picture of transitions is different for *nonadiabatic driving* where, during the transition, the field changes in time and space, and the transition rate is not determined by the instantaneous activation barrier. In this Letter we provide an analytic solution of the long-standing problem of large fluctuations in nonadiabatically driven systems. We show that, over a broad range of field strengths, the field-induced change of the *logarithm* of the fluctuation probability is linear in the field amplitude. However, it is no longer determined by the instantaneous value of the field. The change of the activation energy ΔR can be described in terms of an *observable* characteristic, the *logarithmic susceptibility* (LS). We use LS to analyze nucleation and escape in nonadiabatically driven systems—the problem of a broad physical interest.

The notion of LS and the way to evaluate it are based on the idea of the optimal fluctuational path which goes back to [3]. This is the path along which the system moves, with overwhelming probability, when it fluctuates to a given state or escapes from a metastable state.

Optimal paths in dynamical systems driven by Gaussian noise have attracted much theoretical interest [4] and were recently observed in experiments [5]. The notion of an

optimal path applies also to spatially extended systems [6,7]. Optimal paths provide a solution to the variational problem of finding the most probable realization of the noise which brings the system to a given state. This variational problem describes dynamics of an auxiliary noise-free system, and the minimum of the variational functional determines the logarithm of the probability of the fluctuation.

When the initial fluctuating system is driven by an external field, the variational functional acquires a correction which is linear in the field, for not too strong fields. This correction, and thus the LS can be evaluated along the unperturbed trajectory of the auxiliary system. However, the problem becomes nontrivial when nucleation or escape are analyzed, because of time degeneracy of the unperturbed trajectories.

We will consider optimal paths and logarithmic susceptibility for systems with a nonconserved order parameter. Examples include Ising antiferromagnets and alloys which undergo order-disorder transition [8(a)]. In these systems, fluctuations are described by the Langevin equation

$$\frac{\partial \eta(\mathbf{x}, t)}{\partial t} = -\frac{\delta F}{\delta \eta(\mathbf{x}, t)} + \xi(\mathbf{x}, t), \quad (1)$$

$$F[\eta] = \int d\mathbf{x} \left[\frac{1}{2} (\nabla \eta)^2 + V(\eta) - h(\mathbf{x}, t)\eta \right],$$

where $V(\eta)$ is the biased Landau potential, $h(\mathbf{x}, t)$ is the ac driving field, and $\xi(\mathbf{x}, t)$ is delta-correlated noise of intensity $2kT$. The model (1) describes also fluctuations of an overdamped Brownian particle with a coordinate η (in this case η is independent of \mathbf{x}).

Away from the critical region, the probability density for the system to fluctuate to a state $\eta_f \equiv \eta_f(\mathbf{x})$ at a time t_f is described by the activation law, $W[\eta_f; t_f] \propto \exp(-R[\eta_f; t_f]/kT)$, with the activation energy $R (\gg kT)$ given by the solution of the variational problem [4,6]

$$R[\eta_f; t_f] = \min \frac{1}{4} \int_{-\infty}^{t_f} dt \int d\mathbf{x} \left[\frac{\partial \eta}{\partial t} + \frac{\delta F}{\delta \eta} \right]^2. \quad (2)$$

Here, the minimum is taken with respect to the paths $\eta(\mathbf{x}, t)$ that emanate from the stable state $\eta_{st}(\mathbf{x}, t)$ at

$t \rightarrow -\infty$, and arrive at the final state $\eta_f(\mathbf{x})$ for $t = t_f$. For a periodic field h , the state η_{st} is also periodic.

Equation (2) has the form of the action for an auxiliary Hamiltonian system. The equations of motion for the coordinate $\eta(\mathbf{x}, t)$ and momentum $\pi(\mathbf{x}, t)$ of this system are of the form

$$\dot{\eta} = 2\pi - \frac{\delta F}{\delta \eta}, \quad \dot{\pi} = \hat{M}\pi, \quad \pi(\mathbf{x}, t) = \frac{\delta R[\eta; t]}{\delta \eta(\mathbf{x})}, \quad (3)$$

$$\hat{M}\pi(\mathbf{x}, t) \equiv \int d\mathbf{x}' \frac{\delta^2 F}{\delta \eta(\mathbf{x}) \delta \eta(\mathbf{x}')} \pi(\mathbf{x}', t).$$

The extreme paths $\eta(\mathbf{x}, t)$ that minimize R are optimal fluctuational paths of the original system (1).

In the absence of driving ($h = 0$) the system (1) is in thermal equilibrium, the activation energy $R^{(0)}$ is given by the Gibbs distribution, and one can see from (3) that the optimal paths $\eta^{(0)}(\mathbf{x}, t)$ are the time-reversed paths of (1) in the neglect of noise [9],

$$\dot{\eta}^{(0)} = \delta F^{(0)} / \delta \eta, \quad R^{(0)}[\eta_f] = F^{(0)}[\eta_f] - F^{(0)}[\eta_{st}^{(0)}] \quad (4)$$

(the superscript 0 refers to the case $h = 0$).

To the first order in h , the field-induced change of R is given by the term $\propto h$ in the integrand in (2) evaluated along the unperturbed optimal path $\eta^{(0)}$:

$$R^{(1)}[\eta_f; t_f] = \int_{-\infty}^{t_f} dt \int d\mathbf{x} \chi(\mathbf{x}, t_f - t) h(\mathbf{x}, t), \quad (5)$$

$$\chi(\mathbf{x}, t_f - t) = -\dot{\eta}^{(0)}(\mathbf{x}, t), \quad \eta^{(0)}(\mathbf{x}, t_f) = \eta_f(\mathbf{x}). \quad (6)$$

The quantity χ describes the change $\Delta \ln W \approx -R^{(1)}/kT \propto h$ of the logarithm of the probability density to reach the state η_f . This change may be *large*, and χ may be reasonably called the logarithmic susceptibility. Like susceptibility in linear response theory, LS has a causal form: the probability to reach a given state at a time t_f is affected by the values of the field at $t < t_f$. We note that Eq. (5) suggests how to *measure* LS for various states $\eta_f(\mathbf{x})$.

Of special interest are effects of the field on the probability of escape from a metastable state of the system. For $h = 0$, the critical nucleus $\eta_{cr}^{(0)}(\mathbf{x} - \mathbf{x}_c)$ is the unstable stationary solution of the equation $\dot{\eta} = -\delta F^{(0)} / \delta \eta$ (the saddle point in the functional space), and the position of the center of the nucleus \mathbf{x}_c is arbitrary.

The optimal nucleation path $\eta^{(0)}(\mathbf{x} - \mathbf{x}_c, t - t_c)$ for $h = 0$ is the solution of Eq. (4) which approaches the state $\eta_{cr}^{(0)}(\mathbf{x} - \mathbf{x}_c)$ as $t \rightarrow \infty$. It is a real-time instanton (cf. Fig. 1) and, as in the case of conventional instantons [10], the velocity $|\dot{\eta}^{(0)}|$ is large only within a time interval $|t - t_c| \lesssim t_{rel}$ centered at an arbitrary instant t_c , where t_{rel} is the relaxation time of the system.

Translational and time degeneracy of the optimal nucleation paths are *lifted* when the system is driven by

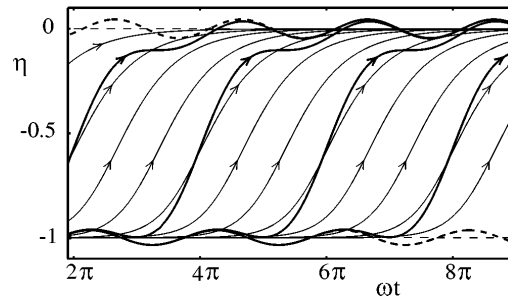


FIG. 1. Optimal escape paths (bold solid lines) of a periodically driven Brownian particle, $\dot{\eta} = \eta - \eta^3 + A \cos \omega t + \xi(t)$, for $A = 0.1$, $\omega = 2$. The paths go from the stable to the unstable periodic states shown by bold dashed lines (by thin dashed lines, in the absence of driving). Thin solid lines show optimal paths in the absence of driving $\eta^{(0)}(t - t_c) = -\{1 + \exp[2(t - t_c)]\}^{-1/2}$, with different t_c . The driving lifts the degeneracy with respect to t_c . The paths $\eta^{(0)}(t - t_c)$ with the “right” t_c [as given by (14)] are the ones around which the exact paths are oscillating. For given A, ω , the linear theory gives the decrease of the activation barrier to an accuracy 12%.

an external field. This is the major qualitative feature of the problem of escape or nucleation in a driven system. The driven system has only *one* optimal nucleation path (one path per period of the field, for a periodic driving), and this path is close to only *one* unperturbed path $\eta^{(0)}(\mathbf{x} - \mathbf{x}_c, t - t_c)$, with certain t_c, \mathbf{x}_c [11]. The difference from the corresponding unperturbed path is $\sim h$, as shown in Fig. 1 for escape paths of a Brownian particle, in which case η is a function of time only.

We obtain the correct values of t_c, \mathbf{x}_c and the activation energy of nucleation by modifying the Mel’nikov method in the theory of perturbed Hamiltonian systems [12]. It follows from Eqs. (4)–(6) that the first order corrections to the unperturbed nucleation trajectory $\eta^{(0)}, \pi^{(0)} = \dot{\eta}^{(0)}$ are of the form

$$\dot{\eta}^{(1)} = \hat{M}^{(0)}\eta^{(1)} + h + 2 \frac{\delta R^{(1)}}{\delta \eta},$$

$$\pi^{(1)} = \hat{M}^{(0)}\eta^{(1)} + \frac{\delta R^{(1)}}{\delta \eta}, \quad R^{(1)} \equiv R^{(1)}[\eta^{(0)}(\mathbf{x}, t); t], \quad (7)$$

$$\eta^{(1)}(\mathbf{x}, t) \rightarrow \eta_{cr}^{(1)}(\mathbf{x}, t), \quad \pi^{(1)}(\mathbf{x}, t) \rightarrow 0, \quad \text{for } t \rightarrow \infty. \quad (8)$$

Here, $\hat{M}^{(0)}$ is the operator \hat{M} taken for $h = 0$ and the path $\eta^{(0)}$; for the problem (1) $\hat{M}^{(0)} = -\nabla^2 + V''(\eta^{(0)})$. In (8), $\eta_{cr}^{(1)}$ is the linear field-induced correction in η_{cr} , and we took into account that the momentum $\pi \rightarrow 0$ as the system approaches the critical nucleus [cf. (3)]. In other terms, we are looking for a heteroclinic orbit of the Hamiltonian system (3). We note, however, that our method makes it possible to find the whole manifold formed by the orbits (3) near the critical nucleus, where the standard perturbation theory diverges.

The solution of (7) near the critical nucleus can be expanded in the eigenfunctions $\psi_n(\mathbf{x} - \mathbf{x}_c)$ of the operator $\hat{M}_{\text{cr}}^{(0)} \equiv -\nabla^2 + V''(\eta_{\text{cr}}^{(0)})$. The corresponding eigenvalue problem coincides with that for the dynamical equation (1) in the absence of noise, but the eigenvalues have opposite signs. The operator $\hat{M}_{\text{cr}}^{(0)}$ has one nondegenerate negative eigenvalue $\lambda_0 < 0$, with the eigenfunction

$$\psi_0(\mathbf{x} - \mathbf{x}_c) = C \lim_{t \rightarrow \infty} e^{-\lambda_0(t-t_c)} \dot{\eta}^{(0)}(\mathbf{x} - \mathbf{x}_c, t - t_c). \quad (9)$$

$\hat{M}_{\text{cr}}^{(0)}$ also has a degenerate zero eigenvalue $\lambda_1 = 0$, with the eigenfunctions $\psi_{1\alpha} \propto \partial \eta_{\text{cr}}^{(0)} / \partial x_\alpha$ ($\alpha = 1, 2, 3$); and positive eigenvalues $\lambda_n > 0$, $n > 1$ [10,13].

We now expand in Eq. (7) $\delta R^{(1)} / \delta \eta$ in the functions ψ_n . It follows from (4)–(6) that the variation $\delta R^{(1)}[\eta; t]$ due to the variation of $\eta(\mathbf{x})$ is of the form

$$\delta R^{(1)} = - \int_{t'}^t dt' \int d\mathbf{x}' \delta \eta(\mathbf{x}', t') \hat{M}^{(0)} h(\mathbf{x}', t'), \quad (10)$$

where $\delta \eta(\mathbf{x}', t')$ is the deviation from the optimal nucleation path $\eta^{(0)}(\mathbf{x}' - \mathbf{x}_c, t' - t_c)$ due to the variation $\delta \eta(\mathbf{x})$ of the state $\eta(\mathbf{x})$ reached by this path at the instant t . For $\eta(\mathbf{x})$ close to the critical nucleus $\eta_{\text{cr}}(\mathbf{x} - \mathbf{x}_c)$, and for not too large $t - t'$ the solution of the linearized equation (4) for $\delta \eta(\mathbf{x}', t')$ can be expressed in terms of the expansion coefficients δ_n of $\delta \eta(\mathbf{x}) = \sum_n \delta_n \psi_n(\mathbf{x} - \mathbf{x}_c)$ as

$$\delta \eta(\mathbf{x}', t') \approx \sum_n \delta_n \psi_n(\mathbf{x}' - \mathbf{x}_c) e^{\lambda_n(t'-t)}. \quad (11)$$

The terms with $n > 1$ in this expression decay with the increasing $t - t'$. This makes it possible to use the local approximation (11) when evaluating the coefficients c_n of the expansion of $\delta R^{(1)} / \delta \eta$ in the eigenfunctions $\psi_{n>1}$:

$$c_n(t) = -\lambda_n \int_{t'}^t dt' \int d\mathbf{x}' \exp[\lambda_n(t' - t)] \times \psi_n(\mathbf{x}' - \mathbf{x}_c) h(\mathbf{x}', t'), \quad n > 1. \quad (12)$$

In contrast to $c_{n>1}$, the expansion coefficients $c_0, c_{1\alpha}$ may not be obtained from the local approximation (11). However, they can be found directly from the explicit form of the eigenfunctions $\psi_0, \psi_{1\alpha}$ and the fact that $R^{(1)} = R^{(1)}[\eta^{(0)}(\mathbf{x} - \mathbf{x}_c, t - t_c); t]$ is a function of the parameters t_c, \mathbf{x}_c of the optimal nucleation path. Therefore

$$c_0 = -C e^{-\lambda_0(t-t_c)} \frac{\partial R^{(1)}}{\partial t_c}, \quad c_{1\alpha} \propto \frac{\partial R^{(1)}}{\partial x_{c\alpha}}. \quad (13)$$

The values of t_c, \mathbf{x}_c can be found from the boundary conditions (8) for the corrections $\eta^{(1)}, \pi^{(1)}$ to the unperturbed nucleation trajectory. It is seen from (7) that, since $\lambda_1 = 0$, the correction $\pi^{(1)} \rightarrow 0$ for $t \rightarrow 0$ provided only the expansion coefficients $c_{1\alpha} = 0$. The coefficient c_0 in (13) contains a growing exponential for $t \rightarrow \infty$ and, according to (7), it should also be equal to zero in order for $\eta^{(1)}(\mathbf{x}, t)$ to satisfy (8). Therefore, for

the optimal nucleation path $c_0 = c_{1\alpha} = 0$. Then from (7) $\eta^{(1)}(t) = -\sum_{n=2}^{\infty} \lambda_n^{-1} c_n(t) \psi_n(\mathbf{x})$ for $t \rightarrow \infty$. It can be seen from (12) and (1) that this expression coincides with the correction to the critical nucleus $\eta_{\text{cr}}^{(1)}(t)$.

The condition that $c_0, c_{1\alpha}$ should vanish is a consequence of the unperturbed system being “soft” in the functional-space directions ψ_0 and $\partial \eta_{\text{cr}}^{(0)} / \partial x_\alpha$ which correspond to the shifts of the nucleation path along t and \mathbf{x} . Taking account of Eqs. (5) and (13), this condition can be written in the form

$$\frac{\partial R_\infty^{(1)}(\mathbf{x}_c, t_c)}{\partial \mathbf{x}_c} = 0, \quad \frac{\partial R_\infty^{(1)}(\mathbf{x}_c, t_c)}{\partial t_c} = 0,$$

$$R_\infty^{(1)} \equiv \int_{-\infty}^{\infty} dt \int d\mathbf{x} \chi_\infty(\mathbf{x} - \mathbf{x}_c, t - t_c) h(\mathbf{x}, t), \quad (14)$$

$$\chi_\infty(\mathbf{x} - \mathbf{x}_c, t - t_c) = -\dot{\eta}^{(0)}(\mathbf{x} - \mathbf{x}_c, t - t_c). \quad (15)$$

Equations (14) determine \mathbf{x}_c, t_c for the optimal nucleation path. They may have several roots which correspond to different heteroclinic orbits of the Hamiltonian system (3). For example, for a periodic driving $R_\infty^{(1)}$ is a periodic function, and it has at least one minimum and maximum per period (it may have more minima, for nonsinusoidal driving). The physically relevant root is the one that provides the absolute minimum to the nucleation barrier $R = R^{(0)}[\eta_{\text{cr}}^{(0)}] + \Delta R$, where ΔR is the field-induced correction. According to Eqs. (5) and (14),

$$\Delta R = \min_{\mathbf{x}_c, t_c} R_\infty^{(1)}(\mathbf{x}_c, t_c). \quad (16)$$

Equations (14)–(16) provide the nonadiabatic theory of the nucleation rate. They have a simple physical meaning: in the presence of a time- and coordinate-dependent field, the optimal fluctuation finds the “best” time t_c and place \mathbf{x}_c to occur. For thermal equilibrium systems, ΔR (16) is the maximal work that can be extracted from the field along the fluctuational path. The results apply to a broad class of extended dynamical models, and also to the problem of escape of a Brownian particle, in which case η is the coordinate of the particle (cf. Fig. 1).

One can see from (15) and (16) that, for a field of the form of a running or standing sinusoidal wave, $h = h_0 \cos(\mathbf{k} \cdot \mathbf{x} - \omega t)$ or $h = h_0 \cos(\mathbf{k} \cdot \mathbf{x}) \cos \omega t$, the correction to the activation energy is negative, and $\Delta R = -|\tilde{\chi}_\infty(\mathbf{k}, \omega)| h_0$ provided ω exceeds the nucleation rate [$\tilde{\chi}_\infty(\mathbf{k}, \omega)$ is the Fourier transform of $\chi_\infty(\mathbf{x}, t)$].

Since χ_∞ is large for $|t - t_c| \lesssim t_{\text{rel}}$, it follows from Eq. (16) that, for pulsed fields, ΔR is nonpositive as well. If the pulse effectively lowers the nucleation barrier, the optimal fluctuation occurs where the field is “on”; otherwise it is most likely to occur where there is no field.

Analytical results for the logarithmic susceptibility for nucleation χ_∞ (15) can be obtained in limiting cases, in particular, for a weakly asymmetric double-well potential $V(\eta) = \frac{1}{4} \eta^4 - \frac{1}{2} \eta^2 - H \eta$, $|H| \ll 1$. The critical

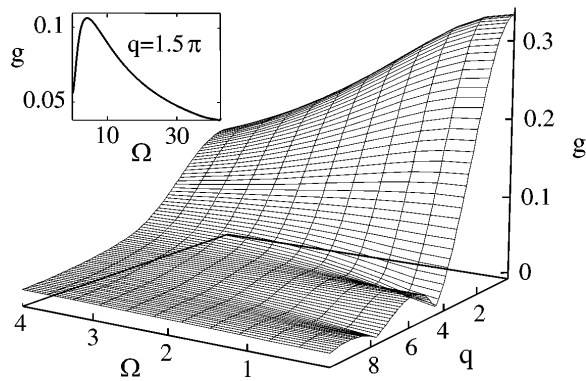


FIG. 2. Reduced absolute value of the logarithmic susceptibility for nucleation $g(q, \Omega)$ (17) in the case of a weakly asymmetric potential V . Inset: The peak in the frequency dependence of the nucleation barrier for $q \neq 0$.

nucleus in this case is a thin-wall droplet of a nucleating phase [10,14]. The optimal nucleation path corresponds to the increase of the radius of the droplet ρ until it reaches the critical value ρ_c , and is described by the time-reversed collapse [14] of the droplet in the absence of fluctuations. The resulting expression for the Fourier transform $\tilde{\chi}_\infty(\mathbf{k}, \omega)$ of the logarithmic susceptibility $\chi_\infty(\mathbf{x}, t)$ is of the form:

$$\tilde{\chi}_\infty(\mathbf{k}, \omega) \equiv \frac{6R^{(0)}}{|H|} g(\rho_c k, \rho_c^2 \omega), \quad (17)$$

$$g(q, \Omega) = \int_0^1 dz \frac{\sin qz}{q} e^{-i\Omega z/2} (1-z)^{-i\Omega/2}$$

(the free energy of the critical droplet $R^{(0)}$ and ρ_c are given in [10,14]).

The LS $|\tilde{\chi}_\infty|$ as given by (17) is shown in Fig. 2. For $\omega = 0$ the LS becomes zero for $k\rho_c = 2\pi n$: the effect of the static sinusoidal field is averaged to zero by a thin-wall critical nucleus with these radii. For $\omega \neq 0$, where the field varies in time while the critical nucleus is growing, LS is finite for all k , and its frequency dependence displays broad resonant peaks, as shown in Fig. 2.

In conclusion, we have provided the nonadiabatic theory of nucleation and escape rates in systems driven by time-dependent fields. The effect of the field on the probabilities of large fluctuations and the transition rates has been described in terms of the logarithmic susceptibility. LS has been analyzed for systems with a nonconserved order parameter. However, the approach is not limited to such systems. Similar to standard susceptibility in the linear response theory, LS is expressed in terms of the characteristics of a system (optimal fluctuation paths) in the absence of the driving field. Therefore it can be calculated for known models, in particular, the explicit form and scaling of LS in the nucleation problem can be obtained for different dynamic universality classes [8]. We note that,

as in the case of standard susceptibility, LS can be measured experimentally even if the underlying dynamics of the system is not known. The results can be used for optimal control, by ac fields, of activated processes such as diffusion in solids and nucleation [15].

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