# Large Fluctuations and Irreversibility in Nonequilibrium Systems

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Large rare fluctuations in a nonequilibrium system are investigated theoretically and by analogue electronic experiment. It is emphasized that the *optimal paths* calculated via the eikonal approximation of the Fokker-Planck equation can be identified with the locus of the ridges of the *prehistory probability distributions* which can be calculated and measured experimentally for paths terminating at a given final point in configuration space. The pattern of optimal paths and its singularities, such as caustics, cusps and switching lines have been calculated and measured experimentally for a periodically driven overdamped oscillator, yielding results that are shown to be in good agreement with each other.

**Key words:** nonequilibrium system, fluctuation, Fokker-Planck equation

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## 1 Introduction

A fluctuating system typically spends most of its time in the close vicinity of a stable state. Just occasionally, however, it will undergo a much larger departure before returning again or perhaps, in some cases, making a transition to the vicinity of a different stable state. Despite their rarity, these large fluctuations are of great importance in diverse contexts including, for example, nucleation at phase transitions, chemical reactions, mutations in DNA sequences, protein transport in biological cells, and failures of electronic devices. In many cases of practical interest, the fluctuating system is far from thermal equilibrium. Examples include lasers [1], pattern-forming systems [2], trapped electrons which display bistability and switching in a strong periodic field [3], and Brownian ratchets [4] which can support a unidirectional current under nonequilibrium conditions. In general, the analysis of the behaviour of nonequilibrium systems is difficult, there being no general relations from which the stationary distribution or the probability of fluctuations can be obtained.

The most promising approach to the analysis of large fluctuations is through the concept of the *optimal path* [5]-[12]. This is the path that the system is predicted to follow with overwhelming probability during the course of qthe fluctuation. For many years it remained unclear how the optimal path - calculated as a trajectory of an auxiliary Hamiltonian system (see below) - is related to the behaviour of real fluctuating systems. Recently, however, through the introduction and use of the prehistory probability distribution [13] (see also [14]), it has been demonstrated that optimal paths are physical observables that can be measured experimentally for both equilibrium [13] and nonequilib-

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rium [15] systems. In what follows we review briefly what has been achieved and point out the opportunities that have now appeared for making rapid scientific progress in this burgeoning research field.

## 2 Theory

Consider an overdamped system driven by a periodic force $K(q;\phi)$ and white noise $\xi(t)$, with equation of motion

$$\begin{aligned} &\dot{q} = K(q;\phi) + \xi(t), \quad K(q;\phi+2\pi), \\ &\phi \equiv \phi(t) = \omega t + \phi_0; \langle \xi(t)\xi(t')\rangle = D\delta(t-t'). \end{aligned} \tag{1}$$

The familiar *overdamped* bistable oscillator driven by a periodic force provides a simple example of the kind of system we have in mind:

$$\begin{aligned} &\dot{q} = -U'(q) + A\cos\omega t + \xi(t), \\ &U(q) = \frac{1}{2}q^2 + \frac{1}{4}q^4. \end{aligned} \tag{2}$$

We consider a situation that is both *nonadiabatic* and *nonlinear*: neither $\omega$ nor $A$ need be small; only the noise intensity $D$ will be assumed small. We shall investigate rare fluctuations to a remote point $(q_f, \phi_f)$ coming from the metastable state within whose domain of attraction $(q_f, \phi_f)$ is located. The position of the stable state $q^0(t)$ is itself a periodic function of time,

$$\begin{aligned} &\dot{q}^{(0)} = K(q^{(0)};\phi), \\ &q^{(0)}(t + 2\pi\omega^{-1}) = q^{(0)}(t). \end{aligned} \tag{3}$$

The equations for optimal paths can be found using the eikonal approximation to solve the corresponding Fokker-Plank equation, or by using a path integral formulation and evaluating the path integral over the fluctuational paths in the steepest descent approximation (for details and discussion see [5, 6, 7, 16, 17]). The optimal path of a periodically driven system corresponds to the locus traced out by the maximum in the prehistory probability density, $p_h(q, \phi | q_f, \phi_f)$ [13, 15]. This is the probability density that a system arriving at the point $(q_f, \phi_f)$ at the instant $t_j(\phi(t_j) = \phi_j)$ had passed through the point $q, \phi$ at the instant $t(t < t_j)$. A particular advantage of this formulation is that $p_h$ is a physical quantity that can be measured experimentally. The approach can be extended to include the analysis of singular points in the pattern of optimal paths.

Using the path-integral expression for the transition probability density [16], one can write $p_h$. in the form [13]

$$\begin{aligned} p_h(q,\phi|q_f,\phi_f) = C\int_{q(t_i)\approx q^{(0)}(t_i)}^{q(t_f)=q_f} Dq(t')\times \\ \times\delta(q(t)-q)\exp\left[-\frac{S[q(t)]}{D} - \right. \\ \left. -\frac{1}{2}\int_{t_i}^{t_f} dt' \frac{\partial K}{\partial q}\right], \ t_i \to -\infty, \\ \phi \equiv \phi(t), \quad \phi_f \equiv \phi(t_f). \end{aligned} \tag{4}$$

Here, $C$ is a normalization constant determined by the condition

$$\int dq p_h(q,\phi|q_f,\phi_f) = 1.$$

S[q(t)] has the form of an action functional for an auxiliary dynamical system with time-dependent Lagrangian $L(\dot{q}, q; \phi)$ :

$$\begin{aligned} &S[q(t)] = \int_{t_i}^{t_f} dt\ L(\dot{q}, q; \phi_f), \\ &L(\dot{q}, q; \phi) = \frac{1}{2}[\dot{q} - K(q;\phi)]^2. \end{aligned} \tag{5}$$

In the range of small noise intensities $D$ the optimal path $q_{opt}(t|q_f, \phi_f)$ to the point $(q_f, \phi_f)$ is given by the condition that the action $S$ be minimal. The variational problem for $S$ to be extremal gives Hamiltonian equations of motion for the coordinate $q$ and momentum $p$ of the auxiliary system

$$\begin{aligned} &\frac{dq}{dt} = \frac{\partial H}{\partial p}, \quad \frac{dp}{dt} = -\frac{\partial H}{\partial q}, \quad \frac{dS}{dt} = \frac{1}{2}p^2 \\ &H \equiv H(q,p;\phi) = \frac{1}{2}p^2 + pK(q;\phi), \\ &H(q,p;\phi) = H(q,p;\phi+2\pi) \end{aligned} \tag{6}$$

The boundary conditions for the extreme paths (6) follow from (4) and (5)

$$\begin{aligned} &q(t_f) = q)_f; \\ &q(t_i) \to q^{(0)}(t_i), \ p(t_i) \to 0, \ S(t_i) \to 0 \\ &\text{for} \ \ t_i \to -\infty. \end{aligned} \tag{7}$$

Since the Hamiltonian $H(q,p;\phi)$ is periodic in $\phi$, the set of paths $\{q(t),p(t)\}$ is also periodic: the paths that arrive at a point $(q_f,\phi_f+2\pi)$ are the same as the paths that arrive at the point $(q_f,\phi_f)$, but shifted in time by the period $2\pi/\omega$. The action $S(q_f,\phi_f)$ evaluated along the extreme paths is also periodic as a function of the phase $\phi_f$ of the final point $(q_f,\phi_f)$ The function $S(q,\phi)$ satisfies the Hamilton-Jacobi equation

$$\begin{aligned} &\omega\frac{\partial S}{\partial\phi} = -H\left(q,\frac{\partial S}{\partial q};\phi\right), p\equiv\frac{\partial S}{\partial q},\\ &S(q,\phi)=S(q,\phi+2\pi). \end{aligned} \tag{8}$$

It is straightforward to see that the extreme paths obtained by solving (6) form a one-parameter set $\{q(t;\triangle),p(t;\triangle)\}$. The parameter $\triangle$ can be chosen as the distance from the extreme path to the attractor $q^{(0)}(t)$ for a certain large negative value of the initial phase $\phi(t_i)$ (we note that to identify the path we have to specify the value of $\phi(t_i)$ for which $\triangle$ has been chosen). To find the value of the momentum $p$ and of the action $S$ for a given $\triangle$ we note from (7) that, for $t\to-\infty$, the extreme paths are confined to the immediate neighbourhood of the attractor. Therefore Eqs. (6) can be linearized about $q^{(0)}(t)$.If for a certain large negative $t_i$, the difference between $q(t_i)$ and $q^{(0)}(t_i)$ is $\triangle$, then the momentum $p(t_i)$ is also proportional to $\triangle$:

$$\begin{aligned} &q(t_i)\equiv q(t_i;\triangle)=q^{(0)}(t_i)+\triangle,\\ &p(t_i)\equiv p(t_i;\triangle)=a(\phi(t_i))\triangle,\\ &S(t_i;\triangle)\equiv S(q(t_i;\triangle),\phi(t_i)=\frac{1}{2}a(\phi(t_i))\triangle^2. \end{aligned} \tag{9}$$

The function $a(\phi)$ can be found from the linearized Hamilton-Jacobi equation (6), noting that the periodicity of $S(q,\phi)$ also implies the periodicity of $a(\phi)$:

$$\begin{aligned} &a(\phi)=\omega(\Upsilon-1)\left[\int_0^{2\pi}d\varphi\Upsilon(\phi,\varphi)\right]^{-1},\\ &\Upsilon(\phi,\varphi)=\exp\left[-2\omega^{-1}\int_\phi^{\phi+\varphi}d\phi'\left(\frac{\partial K}{\partial q}\right)^{(0)}\right],\\ &\Upsilon=\Upsilon(\phi,\phi+2\pi), a(\phi)\equiv a(\phi+2\pi) \end{aligned} \tag{10}$$

the derivative $(\partial K/\partial q)^{(0)}$ is evaluated for $q=q^{(0)}(t'),t'\equiv\omega^{-1}(\phi'-\phi_0))$. This formulation of the problem of fluctuational paths in terms of the Hamiltonian dynamics of an auxiliary system makes it possible to apply to the problem of large fluctuations powerful analytical methods developed earlier in the theory of Hamiltonian systems.

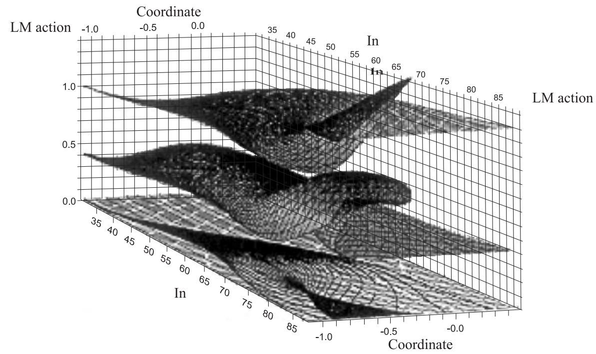


FIG. 1. From top to bottom: action surface, Lagrangian manifold (LM) and extreme paths calculated for the system (2) using equations (6) with initial conditions (9). Parameters for the system were: *A=0.264, w=1.2.* To clarify interrelations between singularities in the optimal paths pattern, action surface, and LM surface, they are shown in one figure: the action surface has been shifted up by one unit, and the LM scaled by a factor 1/2 and shifted up by 0.4.

It is known from the theory of dynamical systems (see for instance [18]) that trajectories emanating from a stationary state lie on a Lagrangian manifold (LM) in phase space $(q,\phi,p=\partial S/\partial q)$ (the unstable manifold of the corresponding state) and form a one-parameter set. The action *S(q, t)* is a smooth single-valued function of position on the LM. It is a Lyapunov function: it is nondecreasing along the trajectories of the initial system in the absence of noise $\dot q=K(q;\phi)$. Therefore *S(q, t)* may be viewed as a generalised nonequilibrium thermodynamic potential for a fluctuating dynamical system [16]. The projections of trajectories in phase space onto configuration space form the extreme paths. Optimal paths are the extreme paths that give the minimal action to a given point in the configuration space. These are the optimal paths that can be visualised in an experiment via measurements of the prehistory probability distribution.

The pattern of extreme paths, LM, and action surfaces for an overdamped periodically driven oscillator (2) are shown in Fig. 1. The figure illustrates generic topological features of the pattern in question. It can be seen from Fig.1 that, although there is only one path to a point $(q, \phi, p)$ in phase space, several different extreme paths may come from the stationary periodic state to the corresponding point $(q, \phi)$ in configuration space. These paths cross each other. This is a consequence of the folding of the Lagrangian manifold.

A generic feature related to folding of LMs is the occurrence of *caustics* in the pattern of extreme paths. Caustics are projections of the folds of an LM. They start at cusp points. It is clear from Fig.1 that an LM structure with two folds merging at the cusp must give rise to a local swallowtail singularity in the action surface. The spinode edges of the action surface correspond to the caustics. A *switching line* emanates from the cusp point at which two caustics meet. This is the projection of the line in phase space along which the two lowest sheets of the action surface intersect. The switching line separates regions which are reached along different *optimal* paths, and the optimal paths intersect on the switching line. The intersection occurs *prior* to a caustic being encountered by the optimal path. The formation of the singularities, avoidance of caustics, and formation of switching lines were analyzed numerically by Jauslin [8], and a complete theory was given by Dykman et al [10]. Until very recently, the generic topological features of the pattern of optimal paths had not been observed in any experiment. We now describe briefly the experimental technique [15] that enables the pattern of optimal paths and its singularities to be observed, and we present and discuss some of our initial results.

The experiments are based on analogue electronic circuits that are designed and built [20] to model the systems of interest, and are then driven by appropriate external forces. Their response is measured and analysed digitally to create the statistical quantity of interest which, in the present case, was usually a prehistory probability distribution [13, 15]. Note that such experiments provide a valid test of the -theory, which should be universally applicable to *any* system described by (1), including natural systems, technological ones, or the electronic models studied here. Some experiments on a model of (2) are now described and discussed as an illustrative example of what can already be achieved.

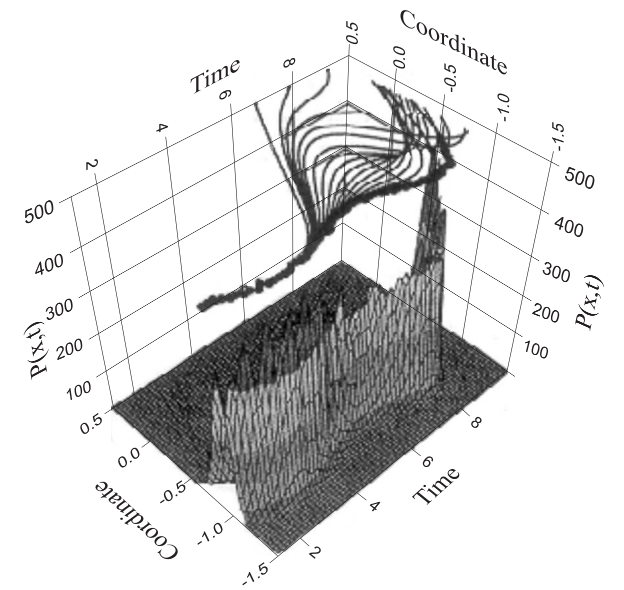


FIG. 2. Distribution of fluctuational paths which terminate inside the rectangle $-0.7 \leq q_f \leq -0.65$, $9.0 \leq t_f \leq 9.2$ for the system (2) with A = 0.264, w = 1.2. The maxima of the distribution represent the optimal paths. The theoretical extreme paths and the contour plot of the distribution are shown at the top.

The model was driven continuously by external quasi-white noise from a noise generator and by a periodic force from a frequency synthesiser. The fluctuating voltage representing *q(t)* .was digitized and analysed in discrete blocks of 32768 samples using a Nicolet NIC-1180 data-processor. The input sweeps were triggered by the frequency synthesiser so that information about the phase of the periodic force could be retained. Whenever *q(t)* entered a designated square centred on a particular $(q_f, \phi_f))$ value, the immediately preceding part of the trajectory was collected and stored; in cases where relaxation trajectories were also of interest, the immediately following part of the trajectory was preserved too. The trajectories that had arrived in any chosen square could subsequently be ensemble-averaged together to create the prehistory probability distribution $p_h(q, \phi|q_f, \phi_f)$ corresponding to the chosen $(q_f, \phi_f)$, with or without the relaxational tail back towards the stable state.

Because the fluctuations of interest were - by definition - rare, it was usually necessary to continue the data acquisition process for several weeks in order to build up acceptably smooth distributions. For this reason, the analysis algorithm was designed to enable trajectories to several termination squares (not just one) to be sought in parallel: an 8 X 8 matrix of 64 adjacent termination squares, each centred on a different $(q_f, \phi_f)$ was scanned.

Experimentally measured prehistory probabilities for arrivals at two different points in configuration space are shown in Figs. 2 and 3. It is immediately evident: (i) that the prehistory distributions are sharp and have a well defined ridges; (ii) that the shapes of the ridges are very different for different final points; and (iii) that the ridges follow very closely the theoretical trajectories obtained by solving numerically the equations of motion for the optimal paths, shown by the full curves on the topplanes. It is also of interest to compare the fluctuational path bringing the system to $(q_f, \phi_f)$ with the relaxational path back towards the stable state. Fig. 4 plots the ridges of a distribution recorded for the special situation that arises when the termination point lies on the switching line [15]. The data are compared with the theoretically predicted fluctuational (thin full curve) and relaxational(thin dashed curve) optimal paths calculated by the methods described above. It can be seen: that there are *two* distinct paths via which the system can arrive at $(q_f, \phi_f)$ but only *one* relaxational path taking it back to the stable state; and that unlike the behaviour expected and seen [21] in equilibrium systems, neither of the fluctuational paths is a time-reversed image of the relaxational one.

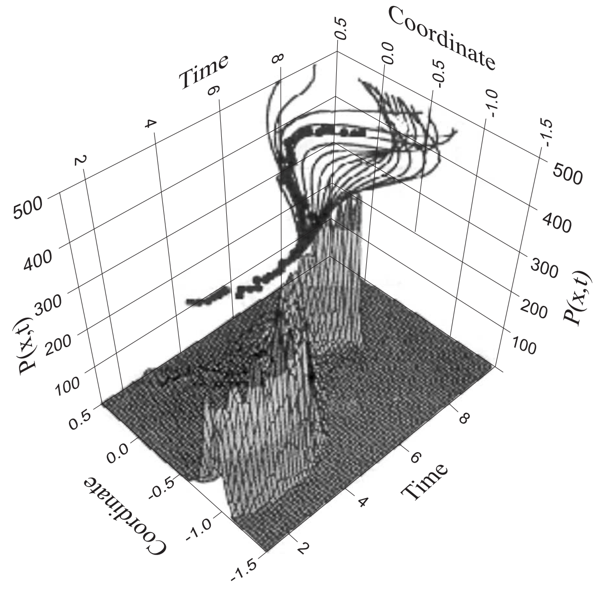


FIG. 3. Distribution of fluctuational paths which terminate inside the rectangle $-0.4 \leq q_f \leq -0.35$, $7.8 \leq t_f \leq 8.1$ for the system (2) with A = 0.264, w = 1.2. The maxima of the distribution represent the optimal paths. The theoretical extreme paths and the contour plot of the distribution are shown at the top.

## 3 Discussion

It is evident from Figs. 2-4 that theory and experiment are in good agreement, provided that the ridges of the experimental ($D \neq 0$) distributions are identified with the optimal paths calculated from the ($D \rightarrow 0$) theory. Results like those of Fig. 4. verifying the existence of a switching line, also demonstrate the nondifferentiability of the generalised nonequilibrium potential. The fact that the fluctuational paths (e.g. those in Fig 4) are observed not to be time-reversed relaxational ones can be understood in the following terms. The theory envisages, even under equilibrium conditions, that the fluctuational and relaxational trajectories belong to two different manifolds of the system (6), with $p \neq 0$ and $p = 0$ respectively. In equilibrium, their projections onto configuration space are time-reversed images of each other; but this degeneracy is immediately lifted by the presence of an external field in the non-equilibrium case, e.g. the periodic force in (2), which breaks time-reversal symmetry.

## 4 Conclusion

The above results, and those of [15], show that our analogue electronic technique makes it possible to test fundamental tenets of fluctuation theory, and

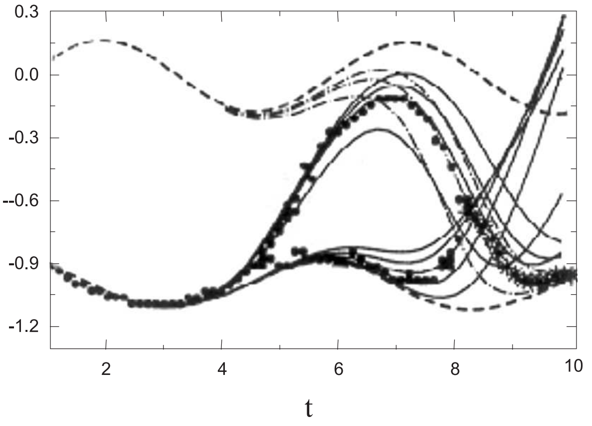


FIG. 4. Measured and calculated fluctuational behaviour for the system (2). The fluctuational and relaxational paths (filled circles and asterisks respectively) to/from the remote state $x_j = -0.63, t = 0.83$that lies on the switching line were determined by tracing the ridges of a measured $P_j(x,t)$ distribution. The time-dependent stable and unstable states near x = - 1 and x = 0 are shown by the dashed lines. The fluctuational and relaxational paths calculated from (6) with initial conditions (9) are shown as full and dash-dotted lines respectively. It can be seen that there are two possible fluctuational paths, forming a corral [15], but only one relaxational path back to the stable state.

thus to provide an experimental basis on which the theory can advance. In particular, we can investigate the pattern of optimal paths for thermally nonequilibrium systems and reveal its singularities including, in particular, switching lines and strong (nonanalytic in the noise intensity) smearing of the prehistory probability distribution near cusp points. The particular system we have investigated has the least number of degrees of freedom necessary to observe these singularities, and therefore it is most appropriate for analysis in these initial investigations. Detailed experimental data related to the connection between microscopic and macroscopic irreversibility, singularities in the pattern of optimal paths, and the theory of self-similar Lagrangian manifolds, will be presented elsewhere. The approach that we have described is in principle applicable to any nonequilibrium system, and we believe it will be found useful in a wide range of applications.


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