LETTER

Non-differentiable large-deviation functionals in boundary-driven diffusive systems

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Abstract. We study the probability of arbitrary density profiles in conserving diffusive fields which are driven by the boundaries. We demonstrate the existence of singularities in the large-deviation functional, the direct analog of the free-energy in non-equilibrium systems. These singularities are unique to non-equilibrium systems and are a direct consequence of the breaking of time-reversal symmetry. The singular behavior is shown in an exactly solvable model and also in numerical simulations on a boundary-driven Ising model. We argue that this is expected to occur in models where the compressibility has a deep minimum. The mechanism is explained using a simple model.

Keywords: fluctuations (theory), stochastic processes (theory), large deviations in non-equilibrium systems

Consider a field \( \rho(x) \) with diffusive dynamics which are conserving in the bulk. Here, \( \rho(x) \) could describe the density of a gas in a capillary connecting two reservoirs. When the reservoirs are of equal density \( \bar{\rho} \), the system is in equilibrium. Then the steady-state probability \( P[\rho_f] \) of an arbitrary density profile \( \rho_f(x) \) is given by \( P[\rho_f] \sim e^{-F[\rho_f]/k_B T} \), where \( F \) is the free-energy. Generically, for a system with short-range interactions, \( F \) is a local functional of \( \rho_f(x) \) and, in the disordered phase, it is a smooth functional. For instance, when the particles in the capillary interact only through hard-core exclusion, \( F[\rho_f] = N \int_0^1 dx \{ \rho_f(x) \log(\rho_f(x)/\bar{\rho}) + (1 - \rho_f(x)) \log((1 - \rho_f(x))/(1 - \bar{\rho})) \} \), where \( N \) is the...
length of the capillary and the density is normalized such that $\rho = 1$ corresponds to a filled capillary.

A fundamental goal of non-equilibrium statistical mechanics is to evaluate and understand the general structure of $P[\rho_f]$ when the densities of the two reservoirs are different, such that a current flows through the system. For diffusive systems there has been great progress in recent years, and by now several important properties of $P[\rho_f]$ have been established. For example, it is known that $P[\rho_f] \sim e^{-N^d\phi[\rho_f]}$, where $N^d$ is the volume of the system and $\phi[\rho_f]$ is a non-equilibrium analog of the free-energy, called the large-deviation functional (LDF). It attains a minimum at the most probable density profile and, when the system is driven out of equilibrium, it becomes a non-local functional of $\rho_f(x)$. This has important consequences, manifested for example through long-range correlations which are present even when the interactions are strictly local [1, 2]. In contrast to equilibrium the LDF depends on the dynamics, and not only on the Boltzmann weights.

Recently, a framework within which $\phi[\rho_f]$ can be calculated has been laid out, building on standard tools from the theory of large deviations [3]–[8]. The key observation is that, in these systems, the probability of an atypical event is dominated by a single history leading up to it, and starting from the most probable profile; other histories are exponentially less likely in the system size. While calculating the LDF within the framework is in general extremely difficult, it has allowed one to establish exact solutions in a number of simple models [4, 9] (in some cases building on solutions obtained by other methods [10]). In addition, the framework has led to efficient numerical algorithms for evaluating $\phi[\rho_f]$ for arbitrary diffusive models [11], as well as LDFs of global quantities such as the current [12]–[15].

Despite the successes, a general understanding of the properties of $\phi[\rho_f]$ out of equilibrium is lacking. For example, for boundary-driven currents induced by reservoirs held at different densities, $\phi[\rho_f]$ is a smooth functional of $\rho_f$ for particles diffusing with hard-core exclusion, just as in equilibrium [10]. In stark contrast, in the presence of a strong bulk drive (for example, when a constant force acts on the particles in the capillary) $\phi[\rho_f]$ becomes non-differentiable [16]. This is reminiscent of singular behavior in the Fokker–Planck equation in the presence of small noise [21]–[26], which has been studied extensively in the past. The phenomenon stems from the existence of multiple histories leading to the same $\rho_f$ with comparable weight, and immediately imply singular behavior in the LDF of other more global quantities. These singularities are more subtle than those observed for the current or particle number [17]–[20], which can arise even if the LDF on the phase space is smooth, hence even in equilibrium models (in contrast to the present phenomenon, see below). It is far from clear which models exhibit such singular behavior, and how to characterize these singularities. It is also natural to ask whether these singularities can appear when no bulk drive is present.

In this letter we show for the first time that $\phi$ can be non-differentiable in boundary-driven diffusive systems. We first provide an example of an exactly solvable model which exhibits such a singularity. We then use numerics to demonstrate that this phenomenon occurs in a broad class of models (for example, in a boundary-driven Ising model). We elucidate the mechanism by which the singularities arise, by introducing a simple model which contains the essential ingredients leading to the singularity. This model shows the close connection between the singularities and spontaneous symmetry breaking. We provide guidelines to systems which can be expected to show this singular behavior.
Background theory. We study large deviations of bulk-conserving diffusive systems which are driven out of equilibrium by the boundaries. For simplicity we consider one dimension, with $0 \leq x \leq 1$. The conserved density $\rho(x,t)$ is related to the current $J(x,t)$ by $\partial_t \rho + \partial_x J = 0$, where the current is given by

$$J = -D(\rho(x,t)) \partial_x \rho(x,t) + \sqrt{\sigma(\rho(x,t))}\eta(x,t).$$

$D(\rho(x,t))$ is a density-dependent diffusivity function, while $\sigma(\rho(x,t))$ controls the amplitude of the white noise $\eta(x,t)$, which satisfies $\langle \eta(x,t) \rangle = 0$ and $\langle \eta(x,t) \eta(x',t') \rangle = N^{-1}\delta(x-x')\delta(t-t')$. The prefactor $N^{-1}$ in the noise variance results from the fact that we have scaled distances by the system size $N$, and time by $N^2$. After this rescaling the noise is small due to the system size, as a consequence of the coarse graining and rescaling. Equation (1) describes a broad range of transport phenomena, including electronic systems, ionic conductors, and heat conduction [5, 27, 28]. $D(\rho)$ and $\sigma(\rho)$ are related via a fluctuation-dissipation relation, which for particle systems reads $\sigma(\rho) = 2k_B T \rho^2 \kappa(\rho) D(\rho)$, where $\kappa(\rho)$ is the compressibility [2]. For example, for diffusing hard-core particles, $D = 1$ and $\sigma = 2\rho(1-\rho)$ [2]. The system is attached to reservoirs which fix the densities at the boundaries of the segment, $\rho(0) = \rho_L$ and $\rho(1) = \rho_R$. If $\rho_L \neq \rho_R$ a current is induced through the system, driving it out of equilibrium.

The average, or most probable density profile for the system $\bar{\rho}$, is obtained by solving $\partial_t [D(\bar{\rho}) \partial_x \bar{\rho}] = 0$, with $\bar{\rho}(0) = \rho_L$ and $\bar{\rho}(1) = \rho_R$ at the boundaries. In equilibrium (i.e. when $\rho_L = \rho_R$), the steady-state probability of any other density profile $\rho_f(x)$ is easy to obtain—the LDF $\phi[\bar{\rho}]$ is then given by the free-energy which is local in $\rho_f$, $\phi[\bar{\rho}] = \int f(\rho,\bar{\rho}) \, dx$, where

$$f(\rho,r) \equiv \int_r^\rho \int_r^{\rho_1} \int_r^{\rho_2} \frac{2D(\rho_2)}{\sigma(\rho_2)} \, d\rho_1 \, d\rho_2.$$  

(2)

Note that in this case $\bar{\rho}$ is constant, $\bar{\rho} = \rho_L = \rho_R$. By contrast, the steady-state probability distribution away from equilibrium is notoriously hard to compute, and very different from the naive guess $\phi[\rho] = \int f(\rho,\bar{\rho}) \, dx$, now with space-dependent $\bar{\rho}(x)$. In fact, as stated above, $\phi[\rho]$ is non-local.

To compute the large deviation for the model described above, one uses the fact that the probability of a history $\{\rho(x,t),J(x,t)\}$ during time $-\infty \leq t \leq 0$ is $P \sim \exp(-NS)$, where the action $S$ is given by [3]–[7]

$$S = \int_{-\infty}^0 dt \int_0^1 dx \frac{|J(x,t)|^2 + D(\rho(x,t)) \partial_x \rho(x,t)^2}{2\sigma(\rho(x,t))}.$$  

(3)

For large $N$, the probability $P \sim \exp(-N\phi[\rho])$ is given by $\phi[\rho_f] = \inf_{\rho,J} S$, where the infimum is over histories satisfying $\partial_t \rho + \partial_x J = 0$, with initial and final conditions $\rho(x,t \rightarrow -\infty) = \bar{\rho}(x)$, $\rho(x,t = 0) = \rho_f(x)$, and boundary conditions $\rho(x = 0,t) = \rho_L$ and $\rho(x = 1,t) = \rho_R$.

In many cases, including in equilibrium and in previously studied exactly solvable non-equilibrium models [9, 10], the action $S$ in equation (3) has a single local minimum and $\phi[\rho_f]$ is then a smooth functional. However, as we show below this need not be the case. The action can, in general, have more than one local minimum in the space of histories $\{\rho(x,t),J(x,t)\}$. In regions of the space of final states where $\rho_f$ has more than one minimal history leading to it, the global minimum might switch between two locally

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minimizing solutions. This is analogous to a first-order phase transition in equilibrium statistical mechanics, where the system switches between two metastable states, which are both local minima of the free-energy. The transition between local minima is accompanied by a jump in the functional derivative of the large deviation \(\delta \phi/\delta \rho\). We will therefore refer to it as a large-deviation singularity (LDS). This phenomenon, first studied by Graham and Tél [21], is unique to non-equilibrium: in equilibrium \(\phi\) is smooth whenever the dynamical model (Langevin equation) contains only smooth functions. Note that while LDSs are expected to be generic in models where the zero-noise dynamics feature a number of basins, or unstable fixed points, here the only fixed point is \(\bar{\rho}(x)\), and therefore the existence of the singularity is not guaranteed, and indeed is not present in the previously studied models [4, 9, 10]. We also note that this phenomenon requires non-linearities in the saddle-point equations. It is therefore not captured by the linear approximation to the dynamics [1], which gives a quadratic LDF (Gaussian probability distribution). In other words, multiple minimizing solutions exist only for a large enough fluctuation.

We first study this phenomenon in an exactly solvable model, and then consider its generalizations.

**Exactly solvable model.** Consider the model in equation (1) with \(D = 1\) and a quadratic \(\sigma(\rho) = 1 + \rho^2\), a parabola clear above the axis\(^1\). In [9] it was shown that the LDF is given by \(\phi[\rho_f] = \min \phi_{\text{ext}}\), where \(\phi_{\text{ext}}\) are extremal values of the action given by

\[
\phi_{\text{ext}} = \int_0^1 dx \left\{ f(\rho_f(x), g(x)) - \ln \frac{g'(x)}{\bar{\rho}'(x)} \right\}
\]

where \(f(\rho, g)\) is defined in equation (2) and \(g(x)\) is an auxiliary function satisfying the differential equation

\[
0 = \frac{g(x) - \rho_f(x)}{\sigma(g(x))} - \frac{g''(x)}{[g'(x)]^2}
\]

with boundary conditions \(g(0) = \rho_L\), and \(g(1) = \rho_R\). Note that as \(D = 1\), the most probable configuration \(\bar{\rho}(x)\) is linear, with \(\bar{\rho}(0) = \rho_L\) and \(\bar{\rho}(1) = \rho_R\). As we now show, solutions to the differential equation equation (4) with initial and final boundary conditions may be non-unique, i.e. there exist profiles \(\rho_f(x)\) for which more than one solution \(g(x)\) exists. Equation (4) is solved via a numerical shooting method [29]: here we treat the problem as an initial value problem with initial conditions \(g(0) = \rho_L\), and \(g'(0) = \alpha\), and scan systematically over values of \(\alpha\) to find solutions with \(g(1) = \rho_R\). This type of exhaustive search ensures that all extremal states are discovered.

To illustrate the existence of multiple solutions, we consider profiles of the form \(\rho_f(x) = \bar{\rho}(x) + \alpha_1 \cos (\pi x/2) + \alpha_2 \sin (\pi x)\), varying \(\alpha_1\) and \(\alpha_2\), and with boundary conditions \(\rho_L = -3, \rho_R = 3\). Figure 1(a) shows an example of a profile \(\rho_f\) for which only one solution \(g(x)\) to equation (4) exists. In contrast, in figure 1(b) a profile \(\rho_f(x)\), which is close to that of figure 1(a), see figures 1(b) and (c), but with three solutions \(g(x)\) is shown. Two of them correspond to local minima, and one to a saddle point. Figure 1(c) shows the region in which there are multiple solutions. Here we have chosen to parametrize the profiles \(\rho_f(x)\) in terms of \(\rho_f(1/3)\) and \(\rho_f(2/3)\), which are simply related to \(\alpha_1\) and \(\alpha_2\). Note the marked caustics, indicating the boundaries between regions with one and three extremal solutions, and the switching line, on which the two locally minimal solutions have the same

\[^1\] It is easy to show that the phenomenon exists in all models with \(D = 1\) and \(\sigma(\rho) = a + b\rho^2\) with \(a, b\) positive.
value for the action. On the switching line the gradient of the LDF is discontinuous [21]; the occurrence of this LDS is the focus of the paper. In addition, the history preceding a rare event is expected to be different on both sides of the switching line, as the history minimizing the action changes. The cusp is found for profiles \( \rho_f \) relatively ‘far’ from \( \bar{\rho} \), and for \( \rho_f(1/3) \) positive and \( \rho_f(2/3) \) negative (here \( \rho_L < \rho_R \)). Interestingly, this type of profile \( \rho(x) \) is similar to the profiles for which the singularity was observed for a model with bulk bias [16]. It is interesting to see whether there is a relation between the detailed mechanism causing the singularity in the present case (as is discussed below) and the case with bulk bias. However, this is beyond the scope of the current Letter. More generally, the phase space of profiles \( \rho_f \) is infinite dimensional, and the caustics and switching line become manifolds. The picture shown in figure 1(c) is a particular two-dimensional cross section. We find similar behavior for \( \rho_f \) of similar shapes which are not of the exact form described above.

In fact, for this model one can prove that: (a) for any non-equilibrium boundary conditions \( \rho_L \neq \rho_R \) there exist profiles \( \rho_f \) for which there is more than one solution to equation (4), and (b) profiles which have two locally minimizing histories with the same value of \( \phi_{\text{ext}} \) always exist. A proof of these facts will be given elsewhere [30].

The existence of multiple minimizers of \( S \) can be intuitively understood as follows: looking at equation (3), we see that the contribution to the action is smaller wherever \( \sigma(\rho) \) is high. If the variation in \( \sigma(\rho) \) is large enough, trajectories passing through regions of high \( \sigma(\rho) \) may be favored. If there are two such regions, as around a minimum of \( \sigma(\rho) \), there might be different paths of the action utilizing the different favorable \( \sigma(\rho) \) regimes. When \( D(\rho) \) also varies, we expect the same logic to apply to regions with high and low \( \sigma(\rho)/D(\rho) \). This argument suggests that the phenomenon is robust, and will occur in other modes with similar features, i.e. when \( \sigma(\rho)/D(\rho) \) has a pronounced minimum. Below we make this argument precise, but first we demonstrate the generality of the phenomenon by studying it on a different model which admits a concrete microscopic realization.

**Boundary-driven Ising model.** We turn to study a boundary-driven Ising model, a lattice gas with on-site exclusion and nearest-neighbor interaction. (This is a variant of the Katz–Lebowitz–Spohn model [31], but with no bulk bias.) Each site \( i = 1, \ldots, N \)
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Figure 2. The boundary-driven Ising model. (a) $D(\rho)$ and $\sigma(\rho)$. (b) Evolution of two locally minimizing histories leading to the same $\rho_f$. (c) The coexistence region, showing the caustics (solid black line), switching line (dashed line), and cross-sections of two histories (gray line). Dashed arrows depict paths of the final state in the numerics, which yield different local minimizers. (d) Contours of equal LDF (solid lines) and the switching line (dashed).

of a one-dimensional lattice can be either occupied (‘1’) or empty (‘0’). The jump rate from site $i$ to site $i + 1$ depends on the occupation at sites $i - 1$ to $i + 2$ according to the following rules: $0100 \xrightarrow{\delta} 0010$, $1011 \xrightarrow{\delta} 1010$, $1010 \xrightarrow{\epsilon} 1100$, and their spatially inverted counterparts with identical rates. The parameter $\epsilon$, with $0 < \epsilon < 1$, corresponds to repulsive interactions between the particles; $\delta$ controls the density dependence of the mobility. As shown in [32, 33], for each parameter set $(\epsilon, \delta)$ one can write implicit analytic equations for $D(\rho)$, which can then be inverted numerically. Then $\sigma(\rho)$ is obtained via the fluctuation-dissipation relation. Figure 2(a) shows $D(\rho)$ and $\sigma(\rho)$ for $(\epsilon, \delta) = (0.05, 0.995)$. For equilibrium boundary conditions this model admits an Ising measure.

We now solve for local minimizers of the action $S$, using the numerical technique described in [11]. The numerical solutions are obtained by gradually changing the end profile $\rho_f$, while continuously maintaining a locally minimizing history $\rho(x, t)$. Different locally minimizing solutions are obtained by changing the final profile to enter the bistable region from different directions, see figure 2(c). As for the exactly solvable model, we again find configurations $\rho_f(x)$ for which multiple local minimizers of $S$ exist. In figure 2(b), we show two different histories $\rho(x, t)$ leading to the same profile $\rho_f$, which is chosen again to be of the form: $\rho_f = \bar{\rho}(x) + \alpha_1 \sin \pi x + \alpha_2 \sin 2\pi x$. In figure 2(c) we plot the numerically-obtained region in the $\rho_f(1/3), \rho_f(2/3)$ plane with multiple minimizers, together with the ‘switching line’ and the caustics. In addition, for a particular $\rho_f(x)$ we depict the two histories leading to it by showing the time-dependent values of $\rho(x = 2/3, t)$ against $\rho(x = 1/3, t)$. Lines of equal LDF are plotted in figure 2(d) to show the jump in its gradient.

Note that in this model and for the chosen parameters, $\sigma(\rho)/D(\rho)$ has a double-hump structure with a deep minimum, so an LDS is expected from the simple considerations discussed above. Indeed, we have experimented with various forms of $D(\rho)$ and $\sigma(\rho)$, and conclude that an LDS occurs when $\sigma(\rho)/D(\rho)$ has a minimum which is deep enough.
Recall that by fluctuation-dissipation $\sigma(\rho)/D(\rho)$ is related to the compressibility via $\sigma(\rho)/D(\rho) = 2k_B T \rho^2 \kappa(\rho)$. These features have to be large enough for this to happen; small changes to a model which does not feature an LDS are generally not enough. When LDSs do appear, we have always found them in regions of phase space where the profiles $\rho_f(x)$ have a similar shape to that shown in figures 1(b) and 2(b). The exact conditions and profiles for an LDS to appear must be studied on a model-specific basis. Below we provide a simple model which illustrates the mechanism leading to LDSs in models with these features.

**Mechanism.** Finally we elucidate the connection between a deep minimum in $\sigma(\rho)$ and the LDS in the configurations discussed above. To do so we introduce a simple sweater sleeve model. We consider a model with $D = 1$ and $\sigma(\rho)$ of order one everywhere except for a narrow range of density values, where it is small:

$$
\sigma(\rho) = \begin{cases} 
\sigma_0 \varepsilon^2 & \rho \in [-\varepsilon/2, \varepsilon/2] \\
\sigma_1(\rho) & \text{otherwise.}
\end{cases}
$$

Here $\varepsilon$ is a small parameter, and $\sigma_1(\rho)$ is some function independent of $\varepsilon$. Then $\sigma(\rho)$ has a deep minimum around $\rho = 0$. (The fact that $\sigma(\rho)$ is discontinuous is not essential—a smoothed version can also be used.)

The key to estimating the action is noting that pushing a mass through the density region $\rho \in [-\varepsilon/2, \varepsilon/2]$ may be costly in terms of the action, creating a ‘noise barrier’. Consider the cost of passing a small mass element $m$ from $\rho = -\varepsilon/2$ to $\rho = \varepsilon/2$ or vice versa, by applying a current $J$, see figure 3(a). This process is done over a time $\Delta t$. The region of space where $\rho \in [-\varepsilon/2, \varepsilon/2]$ is of length $\Delta x$. As $J = m/\Delta t, \partial_x \rho = \varepsilon/\Delta x$, the
action \( S = \int dx \, dt \left( J + \partial_x \rho \right)^2 / (2\sigma) \) is, to order \( O(\varepsilon^{-1}) \),
\[
S = \frac{\Delta x \Delta t}{2\sigma_0 \varepsilon^2} \left( \frac{m}{\Delta t} + \frac{\varepsilon}{\Delta x} \right)^2 = \frac{1}{2\sigma_0 \varepsilon^2} \left( m^2 v + \frac{\varepsilon^2}{v} + 2m \varepsilon \right)
\]
where \( v \equiv \Delta x / \Delta t \). We distinguish between two cases: \( J \) can either be ‘uphill’, (against Fick’s law, requiring a strong noise), with \( \text{sgn}(J) = \text{sgn}(\partial_x \rho) \), or ‘downhill’ with \( \text{sgn}(J) = -\text{sgn}(\partial_x \rho) \). In the ‘uphill’ case, \( m/\Delta t \) and \( \varepsilon/\Delta x \) have the same sign, and minimizing \( S \) over \( v \) we obtain \( v = \varepsilon/m \) or \( S \geq 2m/\sigma_0 \varepsilon + O(\varepsilon^0) \). To push a macroscopic mass \( M \), the bound will then read \( S \geq 2M/\sigma_0 \varepsilon + O(\varepsilon^0) \). In contrast, if \( J \) is ‘downhill’, \( J + \partial_x \rho \) can be made small, with no bound of order \( O(\varepsilon^{-1}) \).

We now argue that for some profiles \( \rho_f \), such as the one depicted in figures 3(c) and (d), the global minimizing history is not unique. We consider boundary conditions \( \rho_L < -\varepsilon/2 \) and \( \varepsilon/2 < \rho_R \), so that \( \rho_f \) crosses the noise barrier three times. To highlight the symmetry-breaking aspect of the phenomenon, we focus on a model with a \( \mathbb{Z}_2 \) symmetry where \( \sigma_1(\rho) = \sigma_1(-\rho) \), \( \rho_L = -\rho_R \), and on a final profile satisfying \( \rho_f(x) = -\rho_f(1-x) \). Then \( \sigma(\rho) \), \( D \) and \( \rho_f \) have a symmetry under the combined operation of \( \rho \to -\rho \) and \( x \to 1-x \). Let \( \rho_{\text{sym}}(x,t) \) be the minimizer of \( S \) subject to this symmetry. In the space of histories it is extremal, but not necessarily the minimizing history. Indeed, we show that other solutions have lower action, spontaneously breaking the symmetry. Referring to figure 3(c) we note that in a symmetric history \( \rho(x,t) \) we have \( \rho(x = 1/2, t) = 0 \), and masses in the region shaded in gray, 2\( M \), must be pushed ‘uphill’ through the \( \rho = 0 \) line, hence \( S \geq 4M/\sigma_0 \varepsilon \). However, in the symmetry-breaking history shown in figure 3(d) only a mass \( M \) has to be pushed ‘uphill’, so that \( S = 2M/\sigma_0 \varepsilon + O(\varepsilon^0) \). Therefore, for a small enough \( \varepsilon \) this solution is favorable. By symmetry, the solution with \( \rho \to -\rho \) and \( x \to 1-x \) has the same action. Therefore the symmetry is spontaneously broken, leading to the LDS with a switching line on profiles obeying this symmetry. Note that some profiles \( \rho_f \), such as those in figure 3(b), can be generated by a history which only pushes mass ‘downhill’ through \( \rho \in [-\varepsilon/2, \varepsilon/2] \), and the above argument for multiple histories will not hold. While the above argument emphasizes the breaking of a symmetry, the phenomenon exists even in the absence of an explicit symmetry in either the final configuration or the model, as was demonstrated in the first two models considered.

The non-differentiability shown in figures 1(c), 2(c) and (d) is perhaps the simplest possible singularity \([26]\). Due to the high dimensionality of the phase space of profiles, more elaborate structures are possible. Understanding and classifying the possible singularities in these models is an exciting direction for future research.

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