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Nonequilibrium grand-canonical ensemble built from a physical particle reservoir

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We introduce a nonequilibrium grand-canonical ensemble defined by considering the stationary state of a driven system of particles put in contact with a particle reservoir. When an additivity assumption holds for the large deviation function of density, a chemical potential of the reservoir can be defined. The grand-canonical distribution then takes a form similar to the equilibrium one. At variance with equilibrium, though, the probability weight is "renormalized" by a contribution coming from the contact, with respect to the canonical probability weight of the isolated system. A formal grand-canonical potential can be introduced in terms of a scaled cumulant generating function, defined as the Legendre-Fenchel transform of the large deviation function of density. The role of the formal Legendre parameter can be played, physically, by the chemical potential of the reservoir when the latter can be defined, or by a potential energy difference applied between the system and the reservoir. Static fluctuation-response relations naturally follow from the large deviation structure. Some of the results are illustrated on two different explicit examples, a gas of noninteracting active particles and a lattice model of interacting particles.

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I. INTRODUCTION

Ensemble equivalence plays a key role in equilibrium statistical physics [1–3], and knowing whether such a concept can be extended to nonequilibrium situations is an important issue in view of building a nonequilibrium thermodynamics for steady states [4–10]. For instance, it would be valuable to know whether a driven stationary system behaves in the same way when its number of particles is fixed, or when it is allowed to exchange particles with a reservoir, corresponding, respectively, to the nonequilibrium extensions of the canonical and grand-canonical ensembles. When attempting to build a grand-canonical ensemble for driven steady-state systems, a first issue may be the ability to define a nonequilibrium chemical potential in a thermodynamically consistent way [6,11,12]. In particular, the study of phase separation in steady-state driven systems has shown that equilibrium concepts need to be generalized [13–18]. Contrary to the case of temperature, for which the lack of energy conservation out of equilibrium hinders a thermodynamically consistent definition in nonequilibrium steady states [19–23], a notion of nonequilibrium chemical potential based on the conservation of the number of particles has been proposed some time ago [24,25], and tested in numerical simulations of stochastic lattice gases [26,27]. This approach relies on the assumption that the large deviation function of particle density is additive when the system is split into subsystems [24,25,28]. Recently, the validity conditions of this assumption have been clarified, by a careful analysis of the coarse-grained dynamics describing the contact between subsystems [29,30]. It has been found in particular that if the coarse-grained contact dynamics satisfies both a factorization property and a macroscopic detailed balance property, the large deviation function is additive and a chemical potential can be defined. However, this chemical potential does not satisfy in general an equation of state [29–31], meaning that it does not depend only on bulk quantities like the density, but also on the contact dynamics itself, at odds with equilibrium situations. A similar lack of an equation of state has also generically been reported for the mechanical pressure in gases of active particles, unless specific symmetries are satisfied [14,32–39].

Having introduced a proper nonequilibrium framework to define chemical potentials for systems in contact, one can try to define a nonequilibrium grand-canonical ensemble and study whether its properties are equivalent to that of the nonequilibrium canonical (fixed particle number) system. The theoretical framework allowing for the definition of nonequilibrium chemical potentials consists in considering two systems in contact, in the weak exchange rate limit [28–30,40] (however, note that interesting phenomena also appear for nonvanishing exchange rates [41]). While the two systems have previously been assumed to have comparable sizes, it is of interest to discuss the case when one of the systems is much larger than the other and plays the role of a particle reservoir. A natural and important question is then to know whether the standard equilibrium thermodynamic structure of the grand canonical ensemble remains essentially valid, or if a different structure emerges in this case. This is the question we explore in this paper. We point out that, contrary to equilibrium where the reservoir of particle is naturally itself at equilibrium, both equilibrium and out-of-equilibrium conditions for the reservoir can be considered here. We also argue that it is important to derive the grand-canonical ensemble by

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explicitly connecting the system to a reservoir, because the contact dynamics between the system and the reservoir plays a role in the statistics of configurations that cannot be neglected in a nonequilibrium context.

The paper is organized as follows. In Sec. II, we briefly review the framework introduced in [29,30,40] to describe the steady state of driven systems in contact exchanging particles at a vanishing rate. Then in Sec. III, we build the nonequilibrium grand-canonical distribution obtained by putting a driven system in contact with a particle reservoir, which may itself be in a nonequilibrium steady state. Under the assumption of additivity (see Sec. II below and [29,30]), the grand-canonical distribution takes a form similar to the equilibrium one, with in particular the usual exponential factor in the chemical potential of the reservoir times the number of particles. However, at odds with equilibrium, the probability weight multiplying this exponential factor is "renormalized" by a contribution coming from the nonequilibrium character of the contact between the system and the reservoir, as compared to the probability weight of the isolated system. The chemical potential of the reservoir plays, as in equilibrium, the role of a Legendre parameter generating all the cumulants by derivation of the associated scaled cumulant generating function. We show that this formal grand-canonical structure is also present even in the absence of additivity, when a chemical potential of the reservoir cannot be defined. This structure leads to a generic static fluctuation-response relation, which can be realized in practice (under some assumptions on the contact dynamics) by using a potential energy difference as a control parameter. Finally we discuss in Sec. IV two different explicit examples of grand-canonical ensembles, a gas of noninteracting active particles, and a lattice model of interacting particles.

II. GENERAL FRAMEWORK FOR SYSTEMS IN CONTACT

Before dealing with the grand-canonical situation of a system in contact with a reservoir, we first review in this section the general case of two systems in (weak) contact. The grand-canonical ensemble will be derived in Sec. III as a limit case, where one of the systems becomes much larger than the other and acts as a reservoir.

A. Two systems in weak contact

In line with our previous works [29–31,40], we consider the following general framework of two systems in contact in the weak exchange rate limit, that we call for short weak contact. Our general setup consists in two driven particle systems A and B with stochastic dynamics, as sketched on Fig. 1. Particles are assumed to interact between themselves and can be subject to external forces. Also, both systems are assumed to be in contact with the same heat bath at a uniform temperature. The systems exchange particles at a low rate as compared to the characteristic frequency of the internal dynamics of each system. For instance, two lattice gases [30] are in a weak contact if the rates to exchange one particle between them stay very small compared to the rates to move the particles within the bulks of each system.

Both systems are subject to driving forces f_A and f_B, respectively, that break global microscopic detailed balance. The number of particles, volume, and density of system $k = A, B$ are, respectively, denoted as N_k, V_k, and $\rho_k = N_k/V_k$. The total number of particles $N_T = N_A + N_B$ is fixed. The microscopic contact dynamics between the two systems is assumed to be orthogonal to the driving force, in the sense of the classification of contacts proposed by Sasa and Tasaki [6]. As a consequence, the contact dynamics does not depend on the driving forces [26,27,29,30] and satisfies the local detailed balance with respect to the equilibrium distribution. However, the contact dynamics does not satisfy in general the microscopic detailed balance relation with respect to the steady-state distributions at nonzero drives. We emphasize that the driving forces are not considered to be small: the systems can be arbitrarily away from equilibrium as long as the weak contact limit holds.

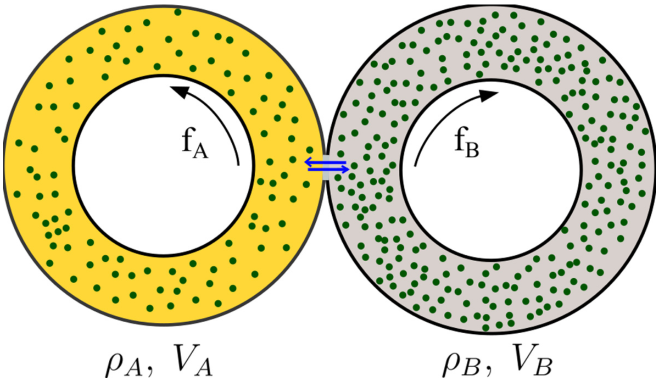

FIG. 1. Systems A and B in weak contact. The *global* densities are ρ_A and ρ_B and the volumes are V_A and V_B. The driving forces are f_A and f_B, respectively.

B. Large deviations of particle densities and definition of chemical potentials

We are specifically interested in determining the joint distribution $P(\rho_A, \rho_B)$ of the global particle densities ρ_A and ρ_B within each system. We note that this does not preclude the local density field to not be perfectly uniform, especially close to the edges of the systems, as for equilibrium systems in the absence of drive. It has been argued in [29,30,40] that in the weak exchange rate limit, the contact dynamics can be conveniently encoded into a coarse-grained exchange rate $\varphi(\Delta N_A; \rho_A, \rho_B)$ with $\Delta N_A = N'_A - N_A$ the number of exchanged particles during a single transition, and ρ_A and ρ_B the densities in each system. These coarse-grained rates rule in particular the time evolution of the joint probability $P_t(\rho_A, \rho_B)$ through a coarse-grained master equation [29,30,40]. Roughly speaking, the coarse-grained exchange rates (evaluated as functions of the global particle densities ρ_A, ρ_B within each system) are obtained by averaging out the microscopic exchange rates with respect to the joint probability distributions of configurations conditioned on the densities ρ_A, ρ_B. In a general setting, one expects the systems to be highly coupled. However, if the microscopic exchange rates between the systems are very weak compared to the bulk dynamics of each system, both systems have the time to relax into their own stationary states between exchanges of particles. The coarse-grained exchange rate $\varphi(\Delta N_A;\rho_A,\rho_B)$ thus becomes an average of the microscopic rates with respect to the product of the stationary probability distributions at densities ρ_A and ρ_B, respectively. In the thermodynamic limit, the joint stationary distribution $P(\rho_A,\rho_B)$ of the number of particles in systems A and B takes the large deviation form

$$P(\rho_A,\rho_B)\asymp e^{-V_\mathrm{T}\,I(\rho_A,\rho_B)}\,, \tag{1}$$

where the symbol $\asymp$ denotes logarithmic equivalence, $V_\mathrm{T}=V_A+V_B$. From the master equation obeyed by the joint distribution $P(\rho_A,\rho_B)$ and the large deviation scaling (1), one can show [29,30] that the large deviation function $I(\rho_A,\rho_B)$ obeys the so-called Hamilton-Jacobi equation

$$\sum_{\Delta N_A\neq 0}\left[\varphi(\Delta N_A;\rho_A,\rho_B)\,e^{I'(\rho_A,\rho_B)\Delta N_A}-\varphi(-\Delta N_A;\rho_A,\rho_B)\right]=0. \tag{2}$$

The derivative I' in Eq. (2) is defined as

$$I'\equiv\frac{1}{\gamma}\frac{d}{d\rho_A}I(\rho_A,\rho_B(\rho_A))=\frac{1}{\gamma}\frac{\partial I}{\partial\rho_A}-\frac{1}{1-\gamma}\frac{\partial I}{\partial\rho_B} \tag{3}$$

having taken into account the conservation law

$$\gamma\rho_A+(1-\gamma)\rho_B=\overline{\rho}\equiv N_\mathrm{T}/V_\mathrm{T}\,, \tag{4}$$

where $\gamma=V_A/V_\mathrm{T}$ is a geometric factor. A situation of specific interest is when the large deviation function is additive, namely,

$$I(\rho_A,\rho_B)=\gamma I_A(\rho_A)+(1-\gamma)I_B(\rho_B)\,. \tag{5}$$

In terms of the derivative I', the additivity condition takes the simple form

$$I'(\rho_A,\rho_B)=I_A'(\rho_A)-I_B'(\rho_B)\,, \tag{6}$$

which allows for the definition of a dimensionless nonequilibrium chemical potential for the systems in contact [24–31]:

$$\mu_k^{\mathrm{cont}}(\rho_k)=I_k'(\rho_k)\qquad(k=A,B)\,. \tag{7}$$

The superscript "cont" aims at highlighting that these chemical potentials are defined for the systems in contact and do not necessarily coincide with chemical potentials that could be defined locally in the bulk of both systems, or even from equilibrium statistical mechanics, if one of the systems is at equilibrium. In the present framework, the validity or not of the additivity condition is a consequence of the contact dynamics, and it is determined by solving Eq. (2). The latter equation can easily be solved in the particular case when the coarse-grained contact dynamics satisfies the macroscopic detailed balance property defined as

$$\varphi(\Delta N_A;\rho_A,\rho_B)\,e^{I'(\rho_A,\rho_B)\Delta N_A}-\varphi(-\Delta N_A;\rho_A,\rho_B)=0\,, \tag{8}$$

for all ΔN_A. Macroscopic detailed balance is obeyed for instance when the stochastic exchange dynamics at contact allows only for single particle exchange. Macroscopic detailed balance is a formal property of the coarse-grained master equation, defined by analogy with the usual microscopic detailed balance property that may be satisfied by the microscopic master equation. However, let us emphasize that the nonequilibrium character of the system is defined by the breaking of *microscopic* detailed balance. While microscopic detailed balance implies its macroscopic counterpart, the reverse is not true, and macroscopic detailed balance may hold in nonequilibrium systems in contact [29,30,40].

It follows from Eq. (8) that

$$I'(\rho_A,\rho_B)=\frac{1}{\Delta N_A}\ln\frac{\varphi(-\Delta N_A;\rho_A,\rho_B)}{\varphi(\Delta N_A;\rho_A,\rho_B)}\,, \tag{9}$$

the resulting expression being independent of ΔN_A. This explicit expression of I' allows for a simple characterization of the additivity property of the large deviation function, as defined by Eq. (6). In this framework, additivity is satisfied when the contact dynamics is factorized between the two systems [29,30]. When the contact dynamics is not factorized, or when the macroscopic detailed balance relation (8) is not obeyed, the large deviation function $I(\rho_A,\rho_B)$ is generically nonadditive [40]. Let us briefly explain why the additivity property is lost. We consider a reference situation for which macroscopic detailed balance is obeyed, characterized by a factorized coarse-grained rate φ_0. The corresponding large deviation function $I_0(\rho_A,\rho_B)$ is thus additive. Let us now introduce a small perturbation of the contact dynamics such that the coarse-grained transition rate reads

$$\varphi=\varphi_0+\varepsilon\varphi_1, \tag{10}$$

with $\varepsilon\ll 1$. We assume that with this perturbed coarse-grained rate, the solution I' of the coarse-grained master Eq. (2) no longer obeys macroscopic detailed balance, thereby making I' more difficult to evaluate. However, it is possible to determine I' perturbatively, to first order in ε, as $I'=I_0'+\varepsilon I_1'+\mathcal{O}(\varepsilon^2)$.

While the leading contribution I_0' is additive if φ_0 takes a factorized form, the subleading contribution I_1' breaks the additivity property, as seen from its expression [40]

$$I_1'=\frac{\sum_{\Delta N_A\neq 0}\varphi_1(\Delta N_A;\rho_A,\rho_B)\big(e^{I_0'(\rho_A,\rho_B)\Delta N_A}-1\big)}{\sum_{\Delta N_A\neq 0}\Delta N_A\varphi_0(\Delta N_A;\rho_A,\rho_B)}. \tag{11}$$

Having determined the large deviation function $I(\rho_A,\rho_B)$, a multiscale analysis in the slow exchange limit shows that the joint distribution of configurations $P(\mathcal{C}_A,\mathcal{C}_B)$ is given to leading order in the small exchange rate by [40]

$$P(\mathcal{C}_A,\mathcal{C}_B)\propto P_A(\mathcal{C}_A|\rho_A V_A)\,P_B(\mathcal{C}_B|\rho_B V_B)\,e^{-V_\mathrm{T}I(\rho_A,\rho_B)} \tag{12}$$

where $P_A(\mathcal{C}_A|N_A)$ and $P_B(\mathcal{C}_B|N_B)$ are the steady-state configuration distributions in systems A and B when isolated, with fixed particle numbers $N_A=\rho_A V_A$ and $N_B=\rho_B V_B$. The densities ρ_A and ρ_B are related by the conservation law $\gamma\rho_A+(1-\gamma)\rho_B=\bar{\rho}$.

We now briefly comment on the interpretation of the parameter ε. As mentioned above, this parameter characterizes the deviation from the macroscopic detailed balance property. As a result, it is *not* a measure of the distance to equilibrium. Indeed, ε can vanish even far from equilibrium, if macroscopic detailed balance is satisfied. On specific examples, like the lattice gas model of Sec. IV B below, ε can be expressed in terms of the drives in both systems. Note that since the first nonzero out-of-equilibrium term of the stationary probability distributions of driven systems is quadratic in the forcing [42–44], the parameter ε is a combination of the square of the driving forces.

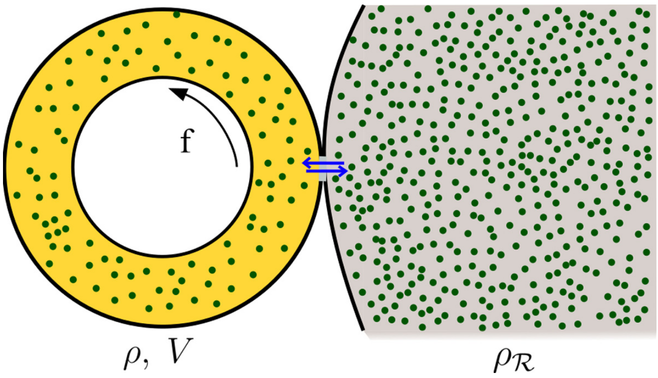

FIG. 2. System of interest in (weak) contact with a reservoir $\mathcal{R}$. The *global* densities are ρ and $\rho_{\mathcal{R}}$, respectively.

III. BUILDING A GRAND-CANONICAL ENSEMBLE

A. System in contact with a reservoir

In the following, we consider system A as the system of interest, and system B is a reservoir the degrees of freedom of which are integrated over, as illustrated in Fig. 2. To emphasize these different roles of the two systems, we slightly change notations and drop the subindex A for quantities characterizing system A (which we simply call "the system" in what follows), while we use from now on the subindex $\mathcal{R}$ for the reservoir (system B). The reservoir being by definition much larger than the system of interest, we take the limit $V_{\mathcal{R}} \to \infty$ keeping fixed the volume V of the system of interest, which implies that the ratio $\gamma = V/(V + V_{\mathcal{R}}) \to 0$. Note that we choose here to consider a general reservoir that is not necessarily at equilibrium and can be driven by nonconservative forces.

The case when the reservoir is an equilibrium system is certainly natural and will be considered henceforth as a particular case. However, note that the case when the reservoir is driven is also a natural situation, for instance when considering self-propelled colloids which are very far from equilibrium (e.g., with a high Péclet number) [45–50].

Generally speaking, the notion of grand-canonical ensemble we consider for the system of interest is the same as in equilibrium, namely, the probability distribution of the microstates of the system of interest when the latter is allowed to exchange particles with a reservoir. In order to derive such a grand-canonical distribution, the joint distribution $P(\mathcal{C}, \mathcal{C}_{\mathcal{R}})$ given in Eq. (12) can be integrated over $\mathcal{C}_{\mathcal{R}}$ to give the distribution of configurations $P(\mathcal{C})$ of the system:

$$P(\mathcal{C}) \propto P(\mathcal{C}|\rho V)\, e^{-V_{\mathrm{T}} I_\gamma(\rho,\rho_{\mathcal{R}})} \tag{13}$$

with $\rho_{\mathcal{R}} = (\bar{\rho} - \gamma\rho)/(1-\gamma)$, and where we have emphasized the γ dependence of the large deviation function $I_\gamma(\rho, \rho_{\mathcal{R}})$ [see Eq. (5) for the explicit dependence of $I_\gamma(\rho, \rho_{\mathcal{R}})$ on γ in the additive case]. The proportionality symbol $\propto$ in Eq. (13) indicates that the normalization factor is not included explicitly.

B. General form of the grand-canonical distribution

To derive the general form of the grand-canonical distribution, we now consider the limit where the reservoir is infinitely larger than the system of interest. We thus take the limit $V_{\mathcal{R}} \to \infty$ at fixed V, implying $\gamma = V/(V + V_{\mathcal{R}}) \to 0$ in Eq. (13). The distribution $P(\mathcal{C}|\rho V)$ of the system considered as isolated does not depend on γ, since we have fixed the volume V. In contrast, the large deviation $I_\gamma(\rho, \rho_{\mathcal{R}})$ generically depends on γ. Introducing the most probable value ρ^* such that $I_\gamma(\rho^*, \rho_{\mathcal{R}}(\rho^*)) = 0$ [we assume here that $I_\gamma(\rho, \rho_{\mathcal{R}})$ is a convex function of ρ, so that ρ^* is unique], we can write, using the definition of I' given in Eq. (3),

$$V_{\mathrm{T}} I_\gamma(\rho, \rho_{\mathcal{R}}) = V \int_{\rho^*}^{\rho} I'\!\left(\rho_1, \frac{\bar{\rho} - \gamma\rho_1}{1-\gamma}\right) d\rho_1 \,. \tag{14}$$

It is important to note at this stage that the derivative $I'(\rho, \rho_{\mathcal{R}})$ of the large deviation function does not depend on γ when considered as a function of two independent arguments ρ and $\rho_{\mathcal{R}}$, because the Hamilton-Jacobi Eq. (2) does not depend explicitly on γ. This is why we write $I'(\rho, \rho_{\mathcal{R}})$ instead of $I'_\gamma(\rho, \rho_{\mathcal{R}})$. It is only when explicitly considering the conservation law $\rho_{\mathcal{R}} = (\bar{\rho} - \gamma\rho)/(1-\gamma)$ that γ comes into play. Taking the limit $\gamma \to 0$ in Eq. (14), we have that $\rho_{\mathcal{R}} \to \bar{\rho}$. Since in this limit $\bar{\rho}$ is also the average density of the reservoir, we will use in the following the notation $\bar{\rho}_{\mathcal{R}}$ instead of $\bar{\rho}$, to avoid possible confusion with the average density of the system. We thus obtain

$$V_{\mathrm{T}} I_\gamma(\rho, \rho_{\mathcal{R}}) \xrightarrow[\gamma\to 0]{} V J(\rho|\bar{\rho}_{\mathcal{R}}) \,, \tag{15}$$

where we have defined

$$J(\rho|\bar{\rho}_{\mathcal{R}}) = \int_{\rho^*}^{\rho} I'(\rho_1, \bar{\rho}_{\mathcal{R}}) d\rho_1 \,. \tag{16}$$

Note that for finite γ, ρ^* may depend on γ if γ is varied while keeping $\bar{\rho}_{\mathcal{R}}$ fixed. In what follows, we assume that the limit $\gamma \to 0$ has been taken, and we define ρ^* by the relation

$$J'(\rho^*|\bar{\rho}_{\mathcal{R}}) = I'(\rho^*, \bar{\rho}_{\mathcal{R}}) = 0, \tag{17}$$

where $J'(\rho|\bar{\rho}_{\mathcal{R}})$ is the derivative of $J(\rho|\bar{\rho}_{\mathcal{R}})$ with respect to ρ. We can now rewrite Eq. (13) more explicitly in the limit $\gamma \to 0$ as

$$P_{\mathrm{GC}}(\mathcal{C}) \propto P(\mathcal{C}|\rho V)\, e^{-VJ(\rho|\bar{\rho}_{\mathcal{R}})} \,. \tag{18}$$

As indicated by the symbol $\propto$, Eq. (18) is not a strict equality as it does not include subleading order terms $\Omega_V(\rho|\bar{\rho}_{\mathcal{R}})$ [such that $\lim_{V\to\infty} V^{-1} \ln \Omega_V(\rho|\bar{\rho}_{\mathcal{R}}) = 0$] that would contribute to the density distribution for small finite volume V [$P_V(\rho) = \Omega_V(\rho|\bar{\rho}_{\mathcal{R}}) e^{-VJ(\rho|\bar{\rho}_{\mathcal{R}})}$]. The expression Eq. (18) is only relevant in the thermodynamic limit.

Equation (18) is the most general form of the grand-canonical distribution in the present nonequilibrium setting. By grand-canonical distribution, we mean the distribution $P_{\mathrm{GC}}(\mathcal{C})$ of the configurations $\mathcal{C}$ of the system, in the limit $\gamma \to 0$ where the reservoir is infinitely larger than the system of interest. To better understand the interpretation of the large deviation function $J(\rho|\bar{\rho}_{\mathcal{R}})$, let us mention its expression in the equilibrium case, anticipating the results below:

$$J_{\mathrm{eq}}(\rho|\bar{\rho}_{\mathcal{R}}) = \beta[f(\rho) - f(\rho^*) - \mu_{\mathrm{eq}}(\bar{\rho}_{\mathcal{R}})(\rho - \rho^*)] \tag{19}$$

where $f(\rho) = \lim_{V\to\infty} -\beta V^{-1} \ln Z_{\mathrm{can}}(\rho V)$ is the equilibrium free energy defined from the canonical partition function $Z_{\mathrm{can}}(\rho V)$ and $\mu_{\mathrm{eq}}(\bar{\rho}_{\mathcal{R}})$ is the equilibrium (dimensionful) chemical potential characterizing the reservoir. At equilibrium, $P(\mathcal{C}|\rho V)$ is the canonical distribution:

$$P(\mathcal{C}|\rho V) = \frac{F(\mathcal{C})}{Z_{\mathrm{can}}(\rho V)} \propto F(\mathcal{C})\, e^{V\beta f(\rho)} \tag{20}$$

where $F(\mathcal{C})$ is the probability weight, simply given at equilibrium by the Boltzmann weight $e^{-\beta E(\mathcal{C})}$, with $E(\mathcal{C})$ the energy of configuration $\mathcal{C}$. It follows that the equilibrium grand-canonical distribution can be reexpressed in its standard form, using Eq. (18), as

$$P_{\mathrm{GC}}(\mathcal{C}) \propto \frac{F(\mathcal{C})}{Z_{\mathrm{GC}}}\, e^{\mu_{\mathrm{eq}}(\bar{\rho}_{\mathcal{R}})V\rho}, \tag{21}$$

where $Z_{\mathrm{GC}} \propto e^{-V[\beta f(\rho^*)+\mu_{\mathrm{eq}}(\bar{\rho}_{\mathcal{R}})\rho^*]}$ is the grand-canonical partition function. We try below to follow a similar path in order to express the nonequilibrium grand-canonical distribution in a form analogous to the equilibrium one, when this is possible.

C. Formal grand-canonical structure

The existence of the grand-canonical large deviation function $J(\rho|\bar{\rho}_{\mathcal{R}})$ guarantees a formal grand-canonical thermodynamic structure, analogous to the equilibrium one. We briefly discuss in this subsection three main features of this grand-canonical thermodynamic structure, namely, ensemble equivalence, the existence of a generalized partition function, and a fluctuation-response relation.

First of all, the large deviation form of the density distribution in Eq. (18) ensures macrostate equivalence [3] in the absence of phase transition. The average of any generic observable $O(\mathcal{C})$ with respect to the grand-canonical ensemble converges to the same limit as the canonical average at density ρ^* in the limit $V \to \infty$, namely,

$$\lim_{V\to\infty} \langle O(\mathcal{C})\rangle_{\mathrm{GC}} = \lim_{V\to\infty} \langle O(\mathcal{C})\rangle_{\rho^*} \tag{22}$$

with $\langle\cdot\rangle_\rho$ the canonical average for a fixed density ρ. We recall that the density ρ^* is the one at which $J'(\rho^*|\bar{\rho}_{\mathcal{R}})$ [or equivalently $I'(\rho^*, \bar{\rho}_{\mathcal{R}})$] vanishes.

Another main feature of a thermodynamic ensemble is the existence of a scaled cumulant generating function, played at equilibrium by the (intensive) Helmholtz free energy for the canonical ensemble or the (intensive) grand potential for the grand-canonical ensemble. A grand potential for the nonequilibrium grand-canonical ensemble defined by Eq. (18) is thus naturally provided by the scaled cumulant generating function of Eq. (18), noted $G(\theta) \equiv \lim_{V\to\infty} V^{-1} \ln\langle e^{V\theta\rho}\rangle$, which reads as

$$G(\theta) = \sup_\rho\, [\theta\rho - J(\rho|\bar{\rho}_{\mathcal{R}})]. \tag{23}$$

This function $G(\theta)$ is the Legendre-Fenchel transform of $J(\rho|\bar{\rho}_{\mathcal{R}})$, with θ the conjugated variable to the density. In the absence of metastability and phase transition (see the Gärtner-Ellis theorem [2]), $J(\rho|\bar{\rho}_{\mathcal{R}})$ is convex. Hence, there exists a single density $\rho^*(\theta)$ that realizes the supremum in Eq. (23):

$$G(\theta) = \theta\rho^*(\theta) - J(\rho^*(\theta)|\bar{\rho}_{\mathcal{R}}), \tag{24}$$

with

$$J'(\rho^*(\theta)|\bar{\rho}_{\mathcal{R}}) = \theta\,. \tag{25}$$

One recovers naturally that $G'(0) \equiv \langle\rho\rangle = \rho^*$ with $J'(\rho^*|\bar{\rho}_{\mathcal{R}}) = 0$. The Legendre-Fenchel transform $G(\theta)$ allows one, by definition, to compute all the cumulants of the density in the thermodynamic limit. This is the case in particular for the second cumulant:

$$G''(0) \equiv \lim_{V\to\infty} \frac{1}{V}\, \mathrm{Var}_V(N)\,, \tag{26}$$

where $\mathrm{Var}_V(N)$ is the variance of the total number of particles $N = V\rho$ at a fixed volume V. Using Eqs. (24) and (25), we have $G'(\theta) = \rho^*(\theta)$. By differentiating again with respect to θ we get, using also Eq. (26),

$$G''(0) = \left.\frac{d\rho^*(\theta)}{d\theta}\right|_{\theta=0} = \lim_{V\to\infty} \frac{1}{V}\, \mathrm{Var}_V(N)\,. \tag{27}$$

The generalized fluctuation-response relation Eq. (27) is thus a consequence of the Legendre-Fenchel duality between $J(\rho|\bar{\rho}_{\mathcal{R}})$ and $G(\theta)$. More generally, one has for any θ that $G''(\theta) = 1/J''(\rho^*(\theta)|\bar{\rho}_{\mathcal{R}}) = d\rho^*/d\theta$.

The existence of such a grand-canonical structure remains nevertheless purely theoretical, as long as the conjugated variable θ does not gain any physical significance. Can the latter be played by a nonequilibrium chemical potential attached to a reservoir, as in equilibrium? Or, if such a nonequilibrium chemical potential does not exist, could an external potential play this role? We aim at answering these issues in the next section.

D. Nonequilibrium grand-canonical chemical potential under the additivity condition

Within the weak contact limit, we have seen in Sec. II B that the large deviation function $I(\rho_A, \rho_B)$ defined in Eq. (1) for two systems brought into contact could be additive under certain circumstances. Sufficient conditions for additivity are the existence of a macroscopic detailed balance relation for the density dynamics (9) as well as a factorization property of the contact that itself relies on the very definition of the microscopic dynamics at contact. We refer the reader to [29,30,40] for a more detailed analysis on this point.

In the limit $\gamma \to 0$ where one of the systems plays the role of a reservoir, the additivity condition (6) leads to

$$J(\rho|\bar{\rho}_{\mathcal{R}}) = I(\rho) - I(\rho^*) - \mu_{\mathcal{R}}(\rho - \rho^*)\,, \tag{28}$$

with, from Eq. (7),

$$I(\rho) = \int^\rho \mu^{\mathrm{cont}}(\rho_1) d\rho_1\,, \tag{29}$$

and $\mu_{\mathcal{R}} \equiv \mu_{\mathcal{R}}(\bar{\rho}_{\mathcal{R}})$. Note that $I(\rho^*) \neq 0$ *a priori*. As seen by comparing the expressions (19) and (28) of the large deviation function $J(\rho|\bar{\rho}_{\mathcal{R}})$, the function $I(\rho)$ appears as a natural generalization of the dimensionless equilibrium free energy $\beta f(\rho)$. By analogy with the equilibrium form of (20) of $P(\mathcal{C}|\rho V)$, it is natural to define

$$F_{\mathrm{cont}}(\mathcal{C}) \propto P(\mathcal{C}|\rho V) e^{-VI(\rho)} \tag{30}$$

as a canonical weight of a configuration $\mathcal{C}$, such that Eq. (18) can be recast into

$$P_{\mathrm{GC}}(\mathcal{C}) \propto \frac{F_{\mathrm{cont}}(\mathcal{C})}{\mathcal{Z}_{\mathrm{GC}}(\mu_{\mathcal{R}})} e^{\mu_{\mathcal{R}} V\rho}\,, \tag{31}$$

with $\mathcal{Z}_{\rm GC}$ a nonequilibrium grand-canonical partition function. Note that Eq. (31) precisely mirrors the equilibrium form (21) of the grand-canonical distribution. However, as discussed in more detail below, the probability weight $F_{\rm cont}(\mathcal{C})$ is now "dressed" with a contribution coming from the nonequilibrium character of the contact dynamics, at odds with the equilibrium case.

Here again, Eqs. (30) and (31) become valid in the thermodynamic limit only: some subleading order terms in V are missing [see comment after Eq. (18)]. This last equation Eq. (31) shares the exact same structure as the equilibrium grand-canonical distribution. In particular, the chemical potential of the reservoir $\mu_{\mathcal{R}}$ also appears as the conjugated variable to ρ, in the very same way as θ in Sec. III C. This connection can be made even more specific by considering the nonequilibrium grand-canonical potential

$$G_{\rm GC}(\mu_{\mathcal{R}}) \equiv \lim_{V\to\infty} \frac{1}{V} \ln \mathcal{Z}_{\rm GC}(\mu_{\mathcal{R}}) \,. \tag{32}$$

This nonequilibrium grand potential is the Legendre-Fenchel transform of $I(\rho)$ (with $\mu_{\mathcal{R}}$ as the conjugated variable) (23) and corresponds to its associated scaled cumulant generating function. Note that Eqs. (23) and (32) contain the exact same information: when the additivity assumption holds, $G(\theta) = G_{\rm GC}(\theta + \mu_{\mathcal{R}}) + C$, with C a constant. It follows that θ and $\mu_{\mathcal{R}}$ play a similar role, in the sense that differentiating $G(\theta)$ or $G_{\rm GC}(\mu_{\mathcal{R}})$ yields the successive cumulants of the density. In particular, the fluctuation-response relation (27) reads

$$G''_{\rm GC}(\mu_{\mathcal{R}}) = \frac{d\rho^*}{d\mu_{\mathcal{R}}} = \lim_{V\to\infty} \frac{1}{V}\, {\rm Var}_V(N)\,, \tag{33}$$

which clearly shows that the role of the formal parameter θ is played here by the chemical potential $\mu_{\mathcal{R}}$ of the reservoir, as in equilibrium.

E. Influence of the contact dynamics on the probability weight $F_{\rm cont}(\mathcal{C})$

Although the nonequilibrium grand-canonical ensemble Eq. (18) shares the very same structure as its equilibrium analog when additivity of the joint large deviation function (6) holds, an important difference remains: as the subscript "cont" in $F_{\rm cont}$ suggests, all the quantities coming from $J(\rho, \bar{\rho}_{\mathcal{R}})$, namely, the chemical potential of the reservoir $\mu_{\mathcal{R}}$ and the large deviation function $I(\rho)$, are in general affected by the microscopic dynamics at contact.

We leave aside the contact dependence of $\mu_{\mathcal{R}}$: although the dynamics at contact may not be completely controlled by the experimenter, we assume that the latter can easily vary the bulk density of the reservoir to set any value of $\mu_{\mathcal{R}}$. In addition, the dependence of the chemical potential of the reservoir on the contact dynamics vanishes when the reservoir is at equilibrium, which is a specific case of interest. However, even if one may disregard for practical purposes the effect of the contact dynamics on the reservoir, the contact still has an essential influence on the system itself. We would like in this subsection to discuss the effect of the contact dynamics on the canonical weight $F_{\rm cont}(\mathcal{C})$ introduced in Eq. (30).

From nonequilibrium exactly solvable models (see, e.g., [51–55]) and general perturbative formula of nonequilibrium steady states [42,44], the canonical probability distribution of the system of interest is indeed found to break down into

$$P(\mathcal{C}|\rho V) = \frac{F_{\rm iso}(\mathcal{C})}{Z(\rho, V)}\,, \tag{34}$$

for a configuration $\mathcal{C}$ with $N = \rho V$ particles, and $P(\mathcal{C}|\rho V) = 0$ otherwise. The quantity $F_{\rm iso}(\mathcal{C})$ is the probability weight of configuration $\mathcal{C}$; at equilibrium it would correspond to the Boltzmann-Gibbs factor $e^{-\beta E(\mathcal{C})}$. $Z(\rho, V)$ is a normalization constant, similar to the partition function at equilibrium, with

$$\psi(\rho) = -\lim_{V\to\infty} \frac{1}{V} \ln Z(\rho, V) \tag{35}$$

its associated rate function that can be thought of as an effective, dimensionless nonequilibrium free energy density [at equilibrium, we would have $\psi(\rho) = \beta f(\rho)$, with $f(\rho)$ the free energy].

As further discussed below, it is important to distinguish the probability weight $F_{\rm iso}(\mathcal{C})$ defined in Eq. (34) for an isolated system, from the probability weight $F_{\rm cont}(\mathcal{C})$ introduced in Eq. (30) for a system in contact with a reservoir. While both weights would be identical at equilibrium, nonequilibrium effects resulting from the contact dynamics yield a distinction between these two probability weights. Accordingly, the nonequilibrium effective free energy $\psi(\rho)$ of the isolated system also differs from its counterpart $I(\rho)$ defined in Eq. (29) for a system in contact with a reservoir.

Following [24,25], one can associate with $\psi(\rho)$ a nonequilibrium chemical potential $\mu_{\rm iso}(\rho)$ such that

$$\psi(\rho) = \int^{\rho} \mu^{\rm iso}(\rho_1) d\rho_1 \,. \tag{36}$$

This nonequilibrium chemical potential $\mu^{\rm iso}$ of the isolated system is the direct analog of an equilibrium chemical potential. First, it would appear naturally as the chemical potential ruling the exchange of mass with a reservoir if the microscopic exchange dynamics would obey a microscopic local detailed balance relation with respect to the weight $F_{\rm iso}(\mathcal{C})$ (see Appendix B for more detail on this point). On the other hand, for the nonequilibrium systems considered in [51–55], the chemical potential $\mu^{\rm iso}$ thus defined becomes uniform at stationarity when the isolated systems are partitioned into virtual subsystems.

Injecting (34) into (30) yields

$$F_{\rm cont}(\mathcal{C}) \propto F_{\rm iso}(\mathcal{C}) e^{-V\zeta(\rho)} \tag{37}$$

with

$$\zeta(\rho) = I(\rho) - \psi(\rho) = \int^{\rho} [\mu^{\rm cont}(\rho_1) - \mu^{\rm iso}(\rho_1)] d\rho_1. \tag{38}$$

In Eq. (37), ρ is the density associated with configuration $\mathcal{C}$. Here again, the subleading terms in V have been dropped out. In this expression, the canonical weight $F_{\rm iso}$ thus appears to be *renormalized* [in the grand-canonical distribution (31)] at the leading order in V by a term $e^{-V\zeta(\rho)}$ that generally depends on ρ, as long as $\mu^{\rm iso} \neq \mu^{\rm cont}$. The ρ dependence of the factor $e^{-V\zeta(\rho)}$ precisely encodes the effect resulting from the nonequilibrium character of the contact.

The reason for this discrepancy between $\mu^{\rm iso}$ and $\mu^{\rm cont}$ is the breaking of the microscopic detailed balance at contact, as discussed in detail in [29,30]. Indeed, the exchange rates at contact are not perturbed by the driving forces (driving forces being orthogonal to the contact) and are thus the same as in equilibrium. Nonetheless, the density fluctuations at contact are generally different from that in equilibrium: the microscopic local detailed balance at contact, which holds with respect to the equilibrium distributions, is generally broken in the presence of drives in the systems which modify the steady-state distributions of configurations. A new *macroscopic* balance between thermodynamic forces then settles at contact. The chemical potentials μ^{cont} are precisely the chemical potentials associated with these thermodynamic forces, whereas the chemical potentials of the isolated systems $\mu^{\text{iso}}(\rho)$ are those attached to the thermodynamic forces that would be observed if the microscopic exchange rates at contact would satisfy a local detailed balance with respect to the weight $F_{\text{iso}}(\mathcal{C})$ of the nonequilibrium systems (see Appendix B).

In addition to this generic nonequilibrium effect, the stationary density fluctuations at the contact region may differ from the bulk if the contact is located near a wall for instance, so that the density field is not uniform. This position-dependent effect may add to the generic discrepancy due to the breaking of microscopic detailed balance at contact. Note that at equilibrium, such boundary effects on the density field may also be present, but they have no consequence on the chemical potentials because of the global microscopic detailed balance.

Finally, let us emphasize that the existence of μ^{cont} (or even μ^{iso}) is not granted for nonequilibrium systems. In particular, the definition of μ^{cont} relies on properties of the contact dynamics (like a factorization property of the coarse-grained exchange rates [29,30]) that may not be met for certain systems. A more detailed discussion can be found in [40]. We discuss this issue further below in the context of the nonequilibrium grand-canonical ensemble.

F. Lack of additivity and effect of an external potential

In the above subsections, we have assumed that the additivity condition is satisfied, allowing for the definition of nonequilibrium chemical potentials for systems in contact. When the additivity property does not hold, the chemical potential of the reservoir cannot be defined. In this case, the grand-canonical distribution can be written, using (18) and (34), as

$$P_{\text{GC}}(\mathcal{C}) = \frac{F_{\text{iso}}(\mathcal{C})}{\mathcal{N}_{\text{GC}}(V)}\, e^{V\lambda(\rho)} \tag{39}$$

with

$$\lambda(\rho) = \psi(\rho) - \psi(\rho^*) - J(\rho|\bar{\rho}_{\mathcal{R}}). \tag{40}$$

In Eq. (39), $\mathcal{N}_{\text{GC}}(V)$ is a normalization constant. An explicit example is provided in Sec. IV B 2. Note that the function $\lambda(\rho)$ introduced in Eq. (39) is not uniquely defined and can be shifted by a constant, depending on $\mathcal{N}_{\text{GC}}(V)$. We have chosen for convenience to define $\lambda(\rho)$ such that $\lambda(\rho^*) = 0$. Using the definition (16) of $J(\rho|\bar{\rho}_{\mathcal{R}})$ and the expression (36) of $\psi(\rho)$, the function $\lambda(\rho)$ may be rewritten as

$$\lambda(\rho) = \int_{\rho^*}^{\rho} [\mu^{\text{iso}}(\rho_1) - I'(\rho_1, \bar{\rho}_{\mathcal{R}})]\, d\rho_1 . \tag{41}$$

The major drawback of the absence of a chemical potential for the reservoir is that a fluctuation-response relation as the one given in Eq. (33) is *a priori* lacking. However, we may look for another instantiation of the formal Legendre parameter θ introduced in Sec. III C. We assume here that macroscopic detailed balance holds, but that the factorization property of the coarse-grained dynamics is not met (see Sec. II B), implying that the additivity property does not hold. In this case, one can introduce another control parameter, which plays a role similar to the chemical potential, by applying a potential difference U between the system and the reservoir, through a modification of the contact dynamics. By an appropriate choice of the contact dynamics, the thermodynamic force can be made linear in the applied potential difference U [40]. Then, the large deviation function $J(\rho|\bar{\rho}_{\mathcal{R}})$ is biased by a linear term as

$$J_U(\rho|\bar{\rho}_{\mathcal{R}}) = J(\rho|\bar{\rho}_{\mathcal{R}}) + \beta U \rho , \tag{42}$$

where β is the inverse temperature of the heat bath. Therefore, in a situation where no chemical potentials can be defined, an external potential difference between the system and the reservoir may appear as the conjugated variable to the density, leading to a grand-canonical structure with $\theta = -\beta U$ (see Sec. III C). In particular, the fluctuation-response relation (27) can be conveniently reformulated in terms of the potential difference U, as

$$\left.\frac{d\rho_U^*}{dU}\right|_{U=0} = -\lim_{V\to\infty} \frac{\beta}{V}\, \text{Var}_V(N) , \tag{43}$$

where ρ_U^* is the average stationary density of the system once a potential difference U is applied between the system and the reservoir.

IV. APPLICATION TO SPECIFIC MODELS

A. Gas of noninteracting active particles

We start by the simple example of a gas of noninteracting active particles, considering either active Brownian particles (ABPs) or run-and-tumble particles (RTPs) [56]. ABPs model experiments on active colloids [45–50] or self-propelled grains [57–59], while RTPs describe the behavior of some type of bacteria [60,61]. As discussed in [31] and sketched in Fig. 3, such a gas can be split into two compartments in contact through a potential energy barrier $U(\mathbf{r})$ (see also [62,63]). We take the second compartment to be much larger than the first one, so that it plays the role of a reservoir $\mathcal{R}$ of particles.

In two dimensions, the overdamped dynamics of the position $\mathbf{r} = (x, y)$ of an active particle reads

$$\dot{\mathbf{r}} = v_0\, \mathbf{e}(\theta) - \kappa \nabla U \tag{44}$$

where v_0 is the self-propulsion speed, θ is the polarity angle of the particle along which the self-propulsion force is applied, and κ is a mobility coefficient. Different models for the dynamics of the angle can be used, for instance a diffusive dynamics $\dot{\theta} = \xi(t)$ for ABPs, with $\xi(t)$ a white noise of diffusion coefficient D_{r}, or a jump dynamics with rate α for RTPs [56]. It is convenient to assume that the potential $U(\mathbf{r})$ depends only on x, and is invariant along the y direction. We set the origin

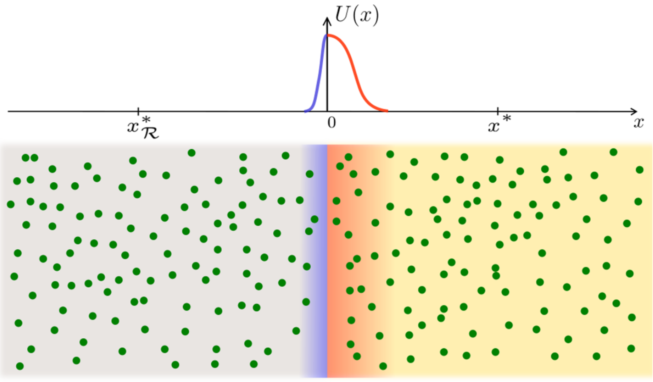

FIG. 3. Two gases of noninteracting active particles in contact. The barrier potential $U(x)$ that forms the contact is typically asymmetric.

$x = 0$ at the maximum of the potential barrier, and define the system of interest to be on the positive part of the x axis, while the reservoir is on the negative part of the axis. Taking the limit of a fast angular dynamics, corresponding to a large $D_{\rm r}$ or a large α, one finds that the large deviation function $I(\rho_A, \rho_B)$ is additive, and one can compute the chemical potential of the two systems in contact. One obtains (see [31] and Appendix A)

$$\mu^{\rm cont}(\rho) = \mu^{\rm iso}(\rho) + \eta\,, \quad \mu^{\rm cont}_{\mathcal{R}}(\bar\rho_{\mathcal{R}}) = \mu^{\rm iso}(\bar\rho_{\mathcal{R}}) + \eta_{\mathcal{R}} \tag{45}$$

where $\mu^{\rm iso}(\rho) = \ln\rho$ is the chemical potential of the isolated ideal gas, whereas η and $\eta_{\mathcal{R}}$ are the nonequilibrium corrections due to the contact. To leading order in $1/D_{\rm r}$ or $1/\alpha$, the correction η_k is given by

$$\eta = -\eta_0 \int_{x^*}^{0} dx\,[U'(x)]^3\,, \quad \eta_{\mathcal{R}} = -\eta_0 \int_{x^*_{\mathcal{R}}}^{0} dx\,[U'(x)]^3\,, \tag{46}$$

with $\eta_0 = \frac{7}{2}\kappa^3 D_{\rm r}/v_0^4$ for active Brownian particles and $\eta_0 = 2\kappa^3\alpha/v_0^4$ for run-and-tumble particles [31] (note that the large $D_{\rm r}$ or large α limit is taken by fixing the effective positional diffusion coefficient $D_{\rm ABP} = v_0^2/2D_{\rm r}$ or $D_{\rm RTP} = v_0^2/2\alpha$). Here, $x^* > 0$ and $x^*_{\mathcal{R}} < 0$ are arbitrary points in the bulk of the system and of the reservoir, respectively, where the smooth potential $U(x)$ vanishes. The parameters η and $\eta_{\mathcal{R}}$ depend explicitly on the shape of the barrier, that is, on the details of the contact dynamics. However, η and $\eta_{\mathcal{R}}$ do not depend on density, which results from the assumption of noninteracting particles.

For the gas of N noninteracting particles, the canonical probability weight $F_{\rm iso}(\mathcal{C})$ is a constant and can be chosen equal to $F_{\rm iso}(\mathcal{C}) = 1$, having defined the configuration $\mathcal{C} = (\mathbf{r}_1, \theta_1, \dots, \mathbf{r}_N, \theta_N)$ as the list of positions and orientations of all particles. Following the results of Sec. III D, this probability weight is renormalized by the contact into

$$F_{\rm cont}(\mathcal{C}) = e^{-V\zeta(\rho)} \tag{47}$$

with here

$$\zeta(\rho) = \int^{\rho} [\mu^{\rm cont}(\rho') - \mu^{\rm iso}(\rho')]\, d\rho' = \eta\rho\,. \tag{48}$$

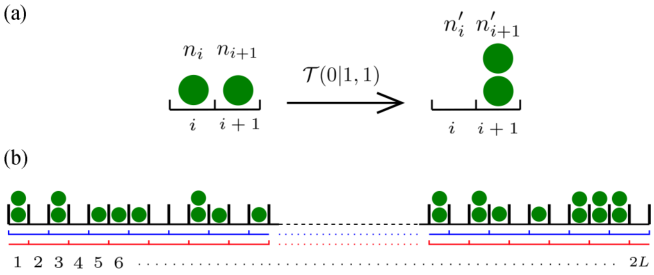

FIG. 4. One-dimensional model of interacting particles in a lattice. (a) Sketch of the update rule on a link $(i, i+1)$ through the transition rate $\mathcal{T}$ [Eq. (50)]. (b) Sketch of the full one-dimensional system. Partition $\mathcal{P}_1$ is denoted in blue and partition $\mathcal{P}_2$ is denoted in red.

The grand-canonical distribution thus reads

$$P_{\rm GC}(\mathcal{C}) \propto e^{(\mu^{\rm cont}_{\mathcal{R}} - \eta)\rho} \tag{49}$$

and the contribution η coming from the contact simply shifts, in practice, the chemical potential of the reservoir. We will see below other examples where the nonequilibrium character of the contact has a more drastic effect.

B. Lattice model of interacting particles

We now turn to an example of a model where the large deviation function is not additive, which can be realized for instance in interacting lattice particle models. Such lattice models are useful benchmarks to test nonequilibrium concepts, as exemplified by the paradigmatic Katz-Lebowitz-Spohn model [64]. Following [30,40], we consider here the particle version of the continuous mass lattice model introduced in [55] as a driven generalization of the model originally defined in [65]. The present model has the advantage that the probability distribution of configurations depends on the drive, at variance with more standard particle [54] or mass transport models [52,66].

The model is defined as follows (see Fig. 4 for a graphical illustration): on each site $i = 1, \dots, 2L$ of a one-dimensional lattice with periodic boundary conditions $(2L + 1 \equiv 1)$, a number $n_i \geqslant 0$ of particles is defined. The dynamics of n_i proceeds by sublattice parallel updates, where one of the two partitions of links $\mathcal{P}_1 = \{(2k-1, 2k), k = 1, \dots, L\}$ and $\mathcal{P}_2 = \{(2k, 2k+1), k = 1, \dots, L\}$ is randomly chosen at each step with equal probabilities. Having chosen a partition $\mathcal{P}_a$ ($a = 1$ or 2), numbers of particles (n_i, n_{i+1}) on each link $(i, i+1)$ of the chosen partition are updated in parallel to (n'_i, n'_{i+1}) according to the probability

$$\mathcal{T}(n'_i|n_i, n_{i+1}) = \frac{e^{-[\bar\varepsilon(n_i)+\bar\varepsilon(n_{i+1})]+\frac{1}{2}f(n'_{i+1}-n'_i)}}{Q(n_i+n_{i+1})} \tag{50}$$

where $n'_{i+1} = n_i + n_{i+1} - n'_i$ is given by the local conservation of the number of particles. The function $\bar\varepsilon(n)$ can be interpreted as the effective energy of n interacting particles, whereas the parameter f is a driving force breaking microscopic detailed balance. The function Q ensures a normalization condition $\sum_{n'_i} \mathcal{T}(n'_i|n_i, n_{i+1}) = 1$ (important to make the model exactly soluble). We focus here on the simplest yet nontrivial model in this class, for which $\bar\varepsilon(n) = +\infty$

for $n > 2$, which amounts to having a maximal number of particles $n_{\max} = 2$ on each site.

We now consider two copies of the model, possibly with different parameters (e.g., different drives), and put them in weak contact by allowing them to exchange particles at a slow rate. The second model is assumed to be much larger than the first one, and plays the role of a reservoir of particles. Quantities characterizing the reservoir are labeled by an index $\mathcal{R}$. We discuss separately the additive and nonadditive cases.

1. Additive case

To illustrate the additive case, we choose the so-called Sasa-Tasaki (ST) contact dynamics [6], which depends only on the configuration of the system from which particles are transferred. The transition rate at contact is defined as [30]

$$\mathcal{T}_{\mathrm{c}}(n_i', n_{j_\mathcal{R}}' | n_i, n_{j_\mathcal{R}}) \propto \begin{cases} \exp\{-[\bar\varepsilon(n_i') - \bar\varepsilon(n_i)]\} & \text{if } n_i' < n_i \\ \exp\{-[\bar\varepsilon_\mathcal{R}(n_{j_\mathcal{R}}') - \bar\varepsilon_\mathcal{R}(n_{j_\mathcal{R}})]\} & \text{if } n_i' > n_i \end{cases} \tag{51}$$

where i and j are two sites of the contact, with i belonging to the system and j to the reservoir. The ST contact dynamics ensures that additivity applies, and hence that a chemical potential $\mu_\mathcal{R}$ of the reservoir can be defined. In this model, the probability weight $F_{\mathrm{iso}}(\mathcal{C})$ of the isolated system associated with a configuration $\mathcal{C} = (n_1, \dots, n_{2L})$ is given by [30,55]

$$F_{\mathrm{iso}}(\mathcal{C}) = \exp\left(\sum_{i=1}^{2L} \bar\varepsilon(n_i)\right) \cosh\left(\sum_{i=1}^{2L} (-1)^i f n_i\right). \tag{52}$$

Once connected to a reservoir with the ST contact dynamics, this probability weight is renormalized into

$$F_{\mathrm{cont}}(\mathcal{C}) = F_{\mathrm{iso}}(\mathcal{C})\, e^{-2L\zeta(\rho)} \tag{53}$$

with $\zeta(\rho) = \int^\rho \eta(\rho')\, d\rho'$ and $\eta(\rho) = \mu^{\mathrm{cont}}(\rho) - \mu^{\mathrm{iso}}(\rho)$. The grand-canonical distribution is then given by

$$P_{\mathrm{GC}}(\mathcal{C}) \propto F_{\mathrm{cont}}(\mathcal{C})\, e^{2L\mu_\mathcal{R}\rho}. \tag{54}$$

The function $\eta(\rho)$ is known in terms of a function $\eta(\mu)$, where $\mu = \mu^{\mathrm{iso}}$ is the chemical potential of the isolated system, which is known only implicitly through a function $\rho(\mu)$. The simplest case is the linear one according to which $\bar\varepsilon(n) = \bar\varepsilon_1 n$ for $n \leqslant 2$ [we recall that $\bar\varepsilon(n) = +\infty$ for $n > 2$]. One can absorb the energy parameter $\bar\varepsilon_1$ into a redefinition of μ^{iso}, introducing $\mu = \mu^{\mathrm{iso}} - \bar\varepsilon_1$. The explicit expressions of the functions $\eta(\mu)$ and $\rho(\mu)$ then read

$$\eta(\mu) = \ln \frac{c_1 + (1 + c_2)e^\mu + 2c_1 e^{2\mu} + e^{3\mu}}{1 + 2c_1 e^\mu + (1 + c_2)e^{2\mu} + c_1 e^{3\mu}}, \tag{55}$$

$$\rho(\mu) = \frac{c_1 e^\mu + (1 + 2c_2)e^{2\mu} + 3c_1 e^{3\mu} + 2e^{4\mu}}{1 + 2c_1 e^\mu + (1 + 2c_2)e^{2\mu} + 2c_1 e^{3\mu} + e^{4\mu}} \tag{56}$$

with $c_1 = \cosh f$ and $c_2 = \cosh(2f)$. For $f = 0$, $\eta(\mu) = 0$ as expected, since at equilibrium $\mu^{\mathrm{cont}} = \mu^{\mathrm{iso}}$. Equations (55) and (56) provide a parametric representation of the function $\eta(\rho)$, which can then be plotted easily (see Fig. 5). The observed symmetry $\eta(\rho) = -\eta(2 - \rho)$ is a consequence of the microscopic particle-hole symmetry $[(\{n_i\}, f, \rho) \leftrightarrow (\{\tau_i\}, -f, 2 - \rho)$, with $\tau_i = 2 - n_i$ the number of holes at site $i]$ that exists for the present version of our model. This holds for μ^{iso} and μ^{cont} as well. The function $\zeta(\rho)$ may then be readily obtained by numerical integration of $\eta(\rho)$ (not shown).

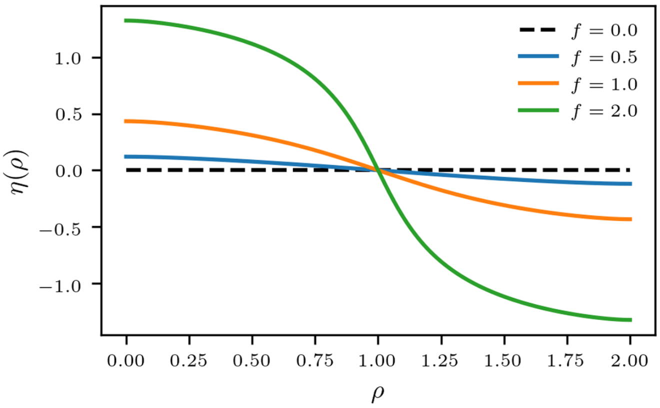

FIG. 5. Plot of the function $\eta(\rho)$ from Eqs. (55) and (56) for the lattice particle model. Parameters are $f = 0, 0.5, 1, 2$. The model settings are $n_{\max} = 2$ and $\bar\varepsilon(n) = \bar\varepsilon_1 n$.

Note that in the small f limit, the expression of $\eta(\mu)$ simplifies to

$$\eta(\mu) = \frac{1 + 2e^\mu - 2e^{2\mu} + e^{3\mu}}{1 + 2e^\mu + 2e^{2\mu} + e^{3\mu}} \frac{f^2}{2} + O(f^4). \tag{57}$$

However, the expression of the function $\rho(\mu)$ remains complicated even in this limit.

2. Nonadditive case

This model can also be used to illustrate the case when the additivity property of the large deviation function $I'(\rho, \bar\rho_{\mathrm{R}})$ breaks down, so that no chemical potential can be defined for the reservoir. This is achieved for instance by using a different contact dynamics, like the Kawasaki dynamics which does not factorize [30,40]. In this case, the grand-canonical distribution takes the general form given in Eq. (39):

$$P_{\mathrm{GC}}(\mathcal{C}) = \frac{F_{\mathrm{iso}}(\mathcal{C})}{\mathcal{N}_{\mathrm{GC}}}\, e^{2L\lambda(\rho)} \tag{58}$$

with $\lambda(\rho)$ defined in Eq. (41). Note that even if no chemical potential of the reservoir appears in the expression of the grand-canonical distribution (58), it is convenient for practical purposes to use the chemical potential $\mu = \mu^{\mathrm{iso}}$ of the isolated system to determine parametrically the ρ dependence of quantities of interest, as done in Sec. IV B 1. Let us emphasize again that the chemical potential μ^{iso} does not play the role of a genuine chemical potential for the systems in contact. It only acts as a useful parameter here.

To evaluate $\lambda(\rho)$, one can take advantage of the fact that the expression of $I'(\rho, \bar\rho_{\mathrm{R}})$ is known explicitly in terms of μ, $I'(\rho, \bar\rho_{\mathrm{R}}) = \tilde I'(\mu, \bar\rho_{\mathrm{R}})$ [30]. One then gets for λ, in terms of the variable μ,

$$\lambda(\mu) = \int^\mu [\mu_1 - \tilde I'(\mu_1, \bar\rho_{\mathrm{R}})] \frac{d\rho}{d\mu}(\mu_1)\, d\mu_1\,. \tag{59}$$

From this relation it is possible to obtain $\lambda(\rho)$ parametrically in terms of μ, using the expression of $\rho(\mu)$ given in Eq. (56).

V. CONCLUSION

As in equilibrium, the grand-canonical setting is expected to be an important physical situation in nonequilibrium thermodynamics. Here, we have followed a physically motivated path by explicitly considering that the system of interest is in contact with a much larger system playing the role of a reservoir, which may be either at or out of equilibrium. Our approach is based on the formalism developed in [29,30] to define nonequilibrium chemical potentials for systems in contact, through the study of the large deviation function of the densities of the two systems when this large deviation function satisfies an additivity property. When additivity holds, a chemical potential of the reservoir can be defined, and the grand-canonical distribution takes a form similar to the equilibrium one, but with a probability weight renormalized by a nonequilibrium contribution of the contact as compared to the probability weight of the isolated system. A formal Legendre grand-canonical structure follows from the large deviation form of the density distribution. The role of the corresponding formal Legendre parameter is played, at a physical level, either by the chemical potential of the reservoir or by a potential energy difference imposed between the system and the reservoir, resulting in several equivalent forms of a static fluctuation-response relation. We have also given the generic expression of the grand-canonical distribution when additivity does not hold, implying that no chemical potential can be defined for the reservoir. Explicit models have also been considered to illustrate on concrete examples the form taken by the grand-canonical distribution. These results provide a firm ground for the definition of a grand-canonical ensemble, and constitute a significant step forward in the quest for a nonequilibrium thermodynamics. They also show that equilibrium thermodynamic notions may not generalize far from equilibrium, since some simplifications resulting from the microscopic detailed balance property are lost, leading to a possibly strong influence of the microscopic contact dynamics.

Regarding future work, it would be of interest to investigate how this grand-canonical ensemble construction is modified when considering a nonvanishing exchange rate with the reservoir. The problem of a finite exchange rate is certainly difficult to address in a general framework, but the further assumption that one system is an ideal reservoir with fast relaxation might bring some simplifications to the problem.

ACKNOWLEDGMENTS

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APPENDIX A: CHEMICAL POTENTIAL OF A GAS OF NONINTERACTING ACTIVE PARTICLES

For the sake of completeness, we provide in this Appendix some more details about the calculations that lead to Eq. (45). A detailed and complete discussion of this system is available in [31].

We focus here for definiteness on the case of ABPs in a potential, but calculations are very similar for other models of active particles like RTPs [31]. The dynamics of the position $\mathbf{r} = (x, y)$ of an ABP in a potential $U(\mathbf{r})$ is defined by

$$\dot{\mathbf{r}} = v_0\, \mathbf{e}(\theta) - \kappa \nabla U\,, \tag{A1}$$

where the angle θ diffuses with angular diffusion coefficient D_r; κ is a mobility coefficient. The probability density $P(\boldsymbol{r}, \theta)$ is governed by the Fokker-Planck equation

$$\frac{\partial P}{\partial t}(\boldsymbol{r}, \theta) = -\nabla \cdot \{[v_0 \boldsymbol{e}(\theta) - \kappa \nabla U(\boldsymbol{r})] P(\boldsymbol{r}, \theta)\} + D_\mathrm{r} \frac{\partial^2 P}{\partial \theta^2}. \tag{A2}$$

Expanding this equation onto angular Fourier modes

$$f_k(\mathbf{r}, t) = \int_0^{2\pi} d\theta\, P(\mathbf{r}, \theta, t)\, e^{ik\theta}, \tag{A3}$$

we get

$$\begin{aligned}\frac{\partial f_k}{\partial t} &= -\frac{v_0}{2}[(\partial_x + i\partial_y) f_{k-1} + (\partial_x - i\partial_y) f_{k+1}] \\ &\quad + \kappa \nabla \cdot (f_k \nabla U) - k^2 D_\mathrm{r} f_k.\end{aligned} \tag{A4}$$

The mode $k = 0$ corresponds to the density field $\rho(\mathbf{r}, t)$, which is a conserved quantity and thus a slow mode. We aim at determining a closed equation on this density field. With this goal in mind, we consider a diffusive limit by taking $D_\mathrm{r} \to \infty$, keeping the effective spatial diffusion coefficient $D = v_0^2/2D_\mathrm{r}$ fixed. In this limit, modes f_k for $k \geqslant 1$ have a fast relaxation, and their time derivative can be neglected, which allows one to reexpress these modes in terms of the density field ρ (see [31] for details). One eventually finds the following closed equation on the density field $\rho(\boldsymbol{r})$:

$$\begin{aligned}\frac{\partial \rho}{\partial t} &= D \nabla \cdot \left[\nabla\left(\rho + \frac{D}{8D_\mathrm{r}} \Delta \rho\right)\right] \\ &\quad + \frac{\kappa D}{D_\mathrm{r}} \nabla \cdot [\Delta \rho \nabla U + (\nabla \rho \cdot \nabla) \nabla U] \\ &\quad + \kappa \nabla \cdot (\rho \nabla U).\end{aligned} \tag{A5}$$

We look for a steady-state solution of Eq. (A5) in the form

$$\rho(\boldsymbol{r}) \propto \exp\left(-\phi_0(\boldsymbol{r}) - \frac{1}{D_\mathrm{r}} \phi_1(\boldsymbol{r})\right). \tag{A6}$$

Under the assumption that the potential energy depends only on the coordinate x, $U(\boldsymbol{r}) = U(x)$, one obtains

$$\phi_0(x) = \frac{\kappa}{D} U(x), \tag{A7a}$$

$$\phi_1(x) = -\frac{\kappa}{8} U''(x) - \frac{13\kappa^2}{16D} U'(x)^2 + \frac{7\kappa^3}{8D^2} \int_0^x dz\, U'(z)^3. \tag{A7b}$$

We interpret the potential $U(x)$ as forming a potential energy barrier separating two compartments A and B, centered at $x = 0$. The smooth potential $U(x)$ vanishes in the bulk of each compartment. By continuity of the density profile $\rho(x)$ at $x = 0$, the steady-state densities ρ_A and ρ_B are related by

$$\rho_A^* e^{\phi(x_A^*)} = \rho_B^* e^{\phi(x_B^*)}\,, \tag{A8}$$

where x_k^* ($k = A, B$) are two points in the bulks of compartments A and B, far from the potential barrier. Since the potential $U(x)$ and its derivatives vanish in the bulk of each compartment, one has

$$\phi(x_k^*) = \frac{7\kappa^3}{8D^2 D_r} \int_0^x dz\, U'(z)^3. \tag{A9}$$

The densities ρ_A and ρ_B are thus found to depend on the shape of the potential barrier.

Given that particles are noninteracting, a simple combinatorial reasoning [31] shows that the large deviation function $I(\rho_A, \rho_B)$ defined in Eq. (1) is given by

$$I(\rho_A, \rho_B) = \gamma_A \rho_A \ln \frac{\rho_A}{\rho_A^*} + \gamma_B \rho_B \ln \frac{\rho_B}{\rho_B^*} \tag{A10}$$

with the constraint $\gamma_A \rho_A + \gamma_B \rho_B = \bar{\rho}$, where $\bar{\rho}$ is the total average density, and γ_k $(k = A, B)$ is the relative volume of compartment k. The large deviation function given in Eq. (A10) is thus additive. Given that the transfer dynamics between compartments proceeds by single particle exchange, macroscopic detailed balance holds. Transition rates have the Sasa-Tasaki form [6], and thus obey a factorization property between systems and reservoir [29,30]. One can thus define a nonequilibrium chemical potential [31]. After rearranging the terms in the expression of the derivative of the large deviation function, and using Eqs. (A8) and (A9), one obtains the expression of the chemical potential in compartment k:

$$\mu_k^{\text{cont}}(\rho) = \ln \rho + \eta_k \quad (k = A, B), \tag{A11}$$

where $\ln \rho$ describes the ideal gas contribution, while η_k is the nonequilibrium correction resulting from the contact. To leading order in $1/D_r$, the nonequilibrium correction η_k reads

$$\eta_k = -\eta_0 \int_{x_k^*}^0 dx\, [U'(x)]^3 \quad (k = A, B), \tag{A12}$$

with $\eta_0 = \frac{7}{2}\kappa^2 D_r / v_0^4$ for ABPs. Similar results hold for related models like RTPs, or ABPs in an external field [31].

APPENDIX B: EQUILIBRIUMLIKE CONTACT DYNAMICS AND ISOLATED CHEMICAL POTENTIAL μ^{iso}

In this Appendix, we briefly sketch the proof that an ideal, equilibriumlike, contact dynamics which obeys a microscopic detailed balance relation with respect to the weights $F_{\text{iso}}(\mathcal{C})$ [as defined in (34)] would lead to $\mu^{\text{cont}} = \mu^{\text{iso}}$, in a similar way as equilibrium.

To do so, one must come back to the generic situation of the contact between two systems A and B in the weak exchange limit, as described in Sec. II (we refer to [29,30,40] for a more detailed analysis). The microscopic dynamics of exchange is defined by the microscopic transition rates $T_c(\mathcal{C}_{A,B} \to \mathcal{C}'_{A,B})$ with $\mathcal{C}_{A,B} = (\mathcal{C}_A, \mathcal{C}_B)$ the pair of the microscopic configurations of systems A and B, respectively. The joint configuration $\mathcal{C}'_{A,B}$ is obtained from $\mathcal{C}_{A,B}$ by a transfer of particles between A and B. We assume the following microscopic local detailed balance relation to hold, for all configurations:

$$\frac{T_c(\mathcal{C}_{A,B} \to \mathcal{C}'_{A,B})}{T_c(\mathcal{C}'_{A,B} \to \mathcal{C}_{A,B})} = \frac{F_A(\mathcal{C}'_A)}{F_A(\mathcal{C}_A)} \frac{F_B(\mathcal{C}'_B)}{F_B(\mathcal{C}_B)}, \tag{B1}$$

with $F_{A,B}$ the respective weights [in the sense of the probability weights $F_{\text{iso}}(\mathcal{C})$ defined for the isolated systems A and B] associated with the canonical distributions (34). Note that such a microscopic dynamics might remain purely theoretical, contrary to the physically motivated dynamics described in Sec. II A which could in principle be implemented experimentally. From the microscopic detailed balance (B1), one directly shows that the coarse-grained transition rates $\varphi(\Delta N_A; \rho_A, \rho_B)$ defined as

$$\varphi(\Delta N_A; \rho_A, \rho_B) = \sum_{\mathcal{C}'_{A,B}} \sum_{\mathcal{C}_{A,B}} T_c(\mathcal{C}_{A,B} \to \mathcal{C}'_{A,B}) P_A(\mathcal{C}_A|\rho_A) P_B(\mathcal{C}_B|\rho_B) \tag{B2}$$

satisfy the macroscopic detailed balance relation (9), that yields

$$I'(\rho_A, \rho_B) = \mu_A^{\text{iso}}(\rho_A) - \mu_B^{\text{iso}}(\rho_B), \tag{B3}$$

with $\mu_{A,B}^{\text{iso}}$ associated with the nonequilibrium free energy (35), for any transition rate T_c obeying the microscopic detailed balance relation (B1). Equation (B3) naturally yields $\mu_A^{\text{iso}}(\rho_A^*) = \mu_B^{\text{iso}}(\rho_B^*)$ at stationarity.

[1] H. Touchette, R. S. Ellis, and B. Turkington, Physica A **340**, 138 (2004).

[2] H. Touchette, Phys. Rep. **478**, 1 (2009).

[3] H. Touchette, J. Stat. Phys. **159**, 987 (2015).

[4] Y. Oono and M. Paniconi, Prog. Theor. Phys. Supp. **130**, 29 (1998).

[5] K. Hayashi and S.-i. Sasa, Phys. Rev. E **68**, 035104(R) (2003).

[6] S.-i. Sasa and H. Tasaki, J. Stat. Phys. **125**, 125 (2006).

[7] T. S. Komatsu, N. Nakagawa, S.-i. Sasa, H. Tasaki, and N. Ito, Prog. Theor. Phys. Supp. **184**, 329 (2010).

[8] T. S. Komatsu, N. Nakagawa, S.-i. Sasa, and H. Tasaki, Phys. Rev. Lett. **100**, 230602 (2008).

[9] T. S. Komatsu, N. Nakagawa, S.-i. Sasa, and H. Tasaki, J. Stat. Phys. **142**, 127 (2011).

[10] T. S. Komatsu, N. Nakagawa, S.-i. Sasa, and H. Tasaki, J. Stat. Phys. **159**, 1237 (2015).

[11] R. Dickman and R. Motai, Phys. Rev. E **89**, 032134 (2014).

[12] R. Dickman, Phys. Rev. E **90**, 062123 (2014).

[13] R. Dickman, New J. Phys. **18**, 043034 (2016).

[14] T. Speck, EPL **114**, 30006 (2016).

[15] V. Prymidis, S. Paliwal, M. Dijkstra, and L. Filion, J. Chem. Phys. **145**, 124904 (2016).

[16] A. P. Solon, J. Stenhammar, M. E. Cates, Y. Kafri, and J. Tailleur, Phys. Rev. E **97**, 020602(R) (2018).

[17] A. P. Solon, J. Stenhammar, M. E. Cates, Y. Kafri, and J. Tailleur, New J. Phys. **20**, 075001 (2018).

[18] S. Paliwal, J. Rodenburg, R. van Roij, and M. Dijkstra, New J. Phys. **20**, 015003 (2018).

[19] J. Casas-Vázquez and D. Jou, Rep. Prog. Phys. **66**, 1937 (2003).
[20] L. F. Cugliandolo, J. Phys. A: Math. Gen. **44**, 483001 (2011).
[21] Y. Shokef, G. Shulkind, and D. Levine, Phys. Rev. E **76**, 030101(R) (2007).
[22] E. Bertin, O. Dauchot, and M. Droz, Phys. Rev. Lett. **93**, 230601 (2004).
[23] K. Martens, E. Bertin, and M. Droz, Phys. Rev. Lett. **103**, 260602 (2009).
[24] E. Bertin, O. Dauchot, and M. Droz, Phys. Rev. Lett. **96**, 120601 (2006).
[25] E. Bertin, K. Martens, O. Dauchot, and M. Droz, Phys. Rev. E **75**, 031120 (2007).
[26] P. Pradhan, C. P. Amann, and U. Seifert, Phys. Rev. Lett. **105**, 150601 (2010).
[27] P. Pradhan, R. Ramsperger, and U. Seifert, Phys. Rev. E **84**, 041104 (2011).
[28] S. Chatterjee, P. Pradhan, and P. K. Mohanty, Phys. Rev. E **91**, 062136 (2015).
[29] J. Guioth and E. Bertin, Europhys. Lett. **123**, 10002 (2018).
[30] J. Guioth and E. Bertin, Phys. Rev. E **100**, 052125 (2019).
[31] J. Guioth and E. Bertin, J. Chem. Phys. **150**, 094108 (2019).
[32] A. P. Solon, Y. Fily, A. Baskaran, M. E. Cates, Y. Kafri, M. Kardar, and J. Tailleur, Nat. Phys. **11**, 673 (2015).
[33] A. P. Solon, J. Stenhammar, R. Wittkowski, M. Kardar, Y. Kafri, M. E. Cates, and J. Tailleur, Phys. Rev. Lett. **114**, 198301 (2015).
[34] R. G. Winkler, A. Wysocki, and G. Gompper, Soft Matter **11**, 6680 (2015).
[35] S. C. Takatori, W. Yan, and J. F. Brady, Phys. Rev. Lett. **113**, 028103 (2014).
[36] S. C. Takatori and J. F. Brady, Phys. Rev. E **91**, 032117 (2015).
[37] T. Speck and R. L. Jack, Phys. Rev. E **93**, 062605 (2016).
[38] M. Joyeux and E. Bertin, Phys. Rev. E **93**, 032605 (2016).
[39] Y. Fily, Y. Kafri, A. P. Solon, J. Tailleur, and A. Turner, J. Phys. A: Math. Theor. **51**, 044003 (2018).
[40] J. Guioth and E. Bertin, J. Stat. Mech. (2020) 063209.
[41] E. N. M. Cirillo, M. Colangeli, and R. Dickman, J. Stat. Mech. (2019) 073203.
[42] C. Maes and K. Netočný, J. Math. Phys. **51**, 015219 (2010).
[43] M. Colangeli, C. Maes, and B. Wynants, J. Phys. A: Math. Theor. **44**, 095001 (2011).
[44] J. A. McLennan Jr., Phys. Rev. **115**, 1405 (1959).
[45] J. Palacci, C. Cottin-Bizonne, C. Ybert, and L. Bocquet, Phys. Rev. Lett. **105**, 088304 (2010).
[46] I. Theurkauff, C. Cottin-Bizonne, J. Palacci, C. Ybert, and L. Bocquet, Phys. Rev. Lett. **108**, 268303 (2012).
[47] I. Buttinoni, J. Bialké, F. Kümmel, H. Löwen, C. Bechinger, and T. Speck, Phys. Rev. Lett. **110**, 238301 (2013).
[48] J. Palacci, S. Sacanna, A. P. Steinberg, D. J. Pine, and P. M. Chaikin, Science **339**, 936 (2013).
[49] A. Bricard, J.-B. Caussin, N. Desreumaux, O. Dauchot, and D. Bartolo, Nature (London) **503**, 95 (2013).
[50] J. R. Gomez-Solano, A. Blokhuis, and C. Bechinger, Phys. Rev. Lett. **116**, 138301 (2016).
[51] B. Derrida, Phys. Rep. **301**, 65 (1998).
[52] M. R. Evans, S. N. Majumdar, and R. K. P. Zia, J. Phys. A: Math. Gen. **37**, L275 (2004).
[53] M. R. Evans, S. N. Majumdar, and R. K. P. Zia, J. Phys. A: Math. Gen. **39**, 4859 (2006).
[54] M. R. Evans and T. Hanney, J. Phys. A: Math. Gen. **38**, R195 (2005).
[55] J. Guioth and E. Bertin, J. Stat. Mech.: Theor. Exp. (2017) 063201.
[56] M. E. Cates and J. Tailleur, Europhysics Letters (EPL) **101**, 20010 (2013).
[57] J. Deseigne, O. Dauchot, and H. Chaté, Phys. Rev. Lett. **105**, 098001 (2010).
[58] V. Narayan, N. Menon, and S. Ramaswamy, J. Stat. Mech. (2006) P01005.
[59] A. Kudrolli, G. Lumay, D. Volfson, and L. S. Tsimring, Phys. Rev. Lett. **100**, 058001 (2008).
[60] F. Peruani, J. Starruss, V. Jakovljevic, L. Søgaard-Andersen, A. Deutsch, and M. Bär, Phys. Rev. Lett. **108**, 098102 (2012).
[61] S. Zhou, A. Sokolov, O. D. Lavrentovich, and I. S. Aranson, Proc. Natl. Acad. Sci. USA **111**, 1265 (2014).
[62] J. Rodenburg, S. Paliwal, M. de Jager, B. G. Bolhuis, M. Dijkstra, and R. van Roij, J. Chem. Phys. **149**, 174910 (2018).
[63] E. Woillez, Y. Kafri, and V. Lecomte, J. Stat. Mech.: Theor. Exp. (2020) 063204.
[64] S. Katz, J. L. Lebowitz, and H. Spohn, J. Stat. Phys. **34**, 497 (1984).
[65] E. Bertin, J.-P. Bouchaud, and F. Lequeux, Phys. Rev. Lett. **95**, 015702 (2005).
[66] M. R. Evans, S. N. Majumdar, and R. K. P. Zia, J. Stat. Phys. **123**, 357 (2006).