Work fluctuations of self-propelled particles in the phase separated state

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We study the large deviations of the distribution \( P(W_\tau) \) of the work associated with the propulsion of individual active brownian particles in a time interval \( \tau \), in the region of the phase diagram where macroscopic phase separation takes place. \( P(W_\tau) \) is characterised by two peaks, associated to particles in the gaseous and in the clusterised phases, and two separate non-convex branches. Accordingly, the generating function of \( W_\tau \)’s cumulants displays a double singularity. We discuss the origin of such non-convex branches in terms of the peculiar dynamics of the system phases, and the relation between the observation time \( \tau \) and the typical persistence times of the particles in the two phases.

INTRODUCTION

The physical observables of equilibrium systems sit at the minima of thermodynamic potentials with small fluctuations regulated by Boltzmann-Einstein expressions [1]. A general framework for the description of larger fluctuations, holding also beyond equilibrium and static phenomena, is given by the theory of large deviations [2]. Consider a quantity \( W_\tau = \sum_{i=1}^{\tau} W_i \), namely the sum of a large number \( \tau \) of stochastic variables: if a large deviation principle (LDP) holds, the asymptotics of the probability distribution \( P(W_\tau) \) is characterized by a \( \tau \)-independent rate function \( I(w) \), \( w = W_\tau/\tau \) being the empirical average, such that

\[
- \lim_{\tau \to \infty} \frac{1}{\tau} \ln \{ P(W_\tau) \} = I(w). \tag{1}
\]

In equilibrium statistical mechanics, considering for instance \( W \) as the energy, and \( \tau \) the number of particles, Einstein’s theory of fluctuations states that the entropy is the rate function, while its Legendre-Fenchel transform is the free energy. When the former is convex, the latter is a regular function. Conversely, non-convexity implies a singular free energy and the lack of ensemble equivalence, occurring, for instance, in the presence of first order phase-transitions.

Via large deviation theory, one can define the analogue of entropy and free-energy functions for dynamical problems, where \( \tau \) is an observation time and \( W_\tau \) an additive functional of the system trajectory in \([0, \tau] \) (such as the entropy production or the work done by some force [3]). A trajectory-based description [4,5] has been adopted for both general considerations, such as proofs of the fluctuation theorem [6–8], and model-specific studies, in the context of sheared fluids [9] or in that of the glass transition [10–12]. The goal of this letter is to use this description to characterise fluctuations in systems of active particles undergoing motility-induced phase separation (MIPS).

In active matter systems, the basic units consume internal energy resources to establish a permanent non-equilibrium condition where work is done on the surrounding environment. This work, which will be later defined for our model of self propelled particles (SPP), is the observable \( W_\tau \) considered in this paper. The interest in systems of SPPs was fostered by the display of properties without any analogue in passive systems [13–16], including MIPS [17–28] and other relevant properties such as accumulation at the system boundaries [29–34], or the possibility to build bio-driven microgears [35,36]. Besides, the presence of an internal mechanism with its own time- and length-scales, as is self-propulsion, causes also fluctuations to display nontrivial features [37,43].

Understanding such properties is instrumental to the development of a stochastic thermodynamics of active suspensions, a topic which has attracted much attention in recent times [39–41].

In this context, we have examined in [40] the large deviations of the individual work \( W_\tau \) done by the self-propulsion force which pushes one active particle, both in the dilute limit and at small but finite density. Here we repeat the analysis for denser suspensions, for which MIPS occurs. We find a double-peaked probability distribution, so that the corresponding rate function of Eq. (1) presents two distinct non-convex branches—a feature not typical of the thermodynamic potentials of passive phase coexistence [2]. We show that different sectors of the rate function correspond to dynamical trajectories of individual particles with different qualitative features. This sheds a new light on the MIPS transition and reveals a rich dynamical behaviour of the cluster phase, complementing the results of [40] where the collective work done by all the active forces in the system was studied.
THE MODEL

We consider, as customary \cite{20}, a system of \( N \) self propelled disks of diameter \( \sigma_d \), with only soft excluded volume interaction, in a two-dimensional square box of side \( L \) with periodic boundary conditions. Each particle is propelled by an active force with modulus \( F_{\text{act}} \) and direction \( \mathbf{n}_i = (\cos \theta_i(t), \sin \theta_i(t)) \). The \( i \)-th particle position \( \mathbf{r}_i \) and orientation \( \theta_i \) obey

\[
\dot{\mathbf{r}}_i = -\gamma \dot{\mathbf{r}}_i + F_{\text{act}} \mathbf{n}_i - \nabla_i \sum_{j \neq i} U(r_{ij}) + \xi_i, \quad \dot{\theta}_i = \eta_i, \quad (2)
\]

where \( r_{ij} = |\mathbf{r}_i - \mathbf{r}_j| \) is the inter-particle distance and \( U(r) \) is a purely repulsive potential \( U(r) = 4\varepsilon (\sigma/r)^{64} - (\sigma/r)^{32} + \varepsilon \) if \( r < 2^{1/32}\sigma \) and 0 otherwise, \( \sigma_d = 2^{1/32}\sigma \) in order to have the potential truncated at its minimum, set equal to the disks diameter \( \sigma \) \cite{27,28}. \( \xi \) and \( \eta \) are zero-mean Gaussian noises satisfying \( \langle \xi_i(t) \xi_j(t') \rangle = 2\gamma k_B T \delta_{ij} \delta(t - t') \mathbf{1} \) and \( \langle \eta_i(t) \eta_j(t') \rangle = 2D_\theta \delta_{ij} \delta(t - t') \). The units of length, mass and energy are given by \( \sigma_d, m \) and \( \varepsilon \), respectively. The rotational diffusion coefficient is set to \( D_\theta = 3\gamma k_B T/\sigma_d^2 \) \cite{20}. The controlling parameters are the packing fraction \( \phi = \pi \sigma_d^2 N/(4L^2) \), which is tuned fixing \( N \) and varying \( L \), and the Péclet number \( Pe = F_{\text{act}} \sigma_d/(k_B T) \), which we change by varying \( F_{\text{act}} \) at \( \gamma = 10 \) and \( k_B T = 0.05 \). For this choice of parameters, inertial terms are typically negligible. The phase diagram of this system has been studied in detail \cite{28,51,52}. For \( Pe \geq Pe_c \approx 38 \), when \( \phi \) exceeds a Péclet dependent threshold, an initial homogeneous state separates into a dense and a gaseous phase.

A typical configuration of the system in the phase separated region \( Pe \geq Pe_c \) is shown in Fig. 1. One observes a large aggregate of particles coexisting with a gaseous phase: due to activity, small clusters roam the gaseous phase, while the dense phase is filled with holes \cite{53} and other defects \cite{54}.

ACTIVE WORK

We evaluate, for each disk \( i \), the active work

\[
W_\tau = F_{\text{act}} \int_0^\tau \mathbf{n}_i(t) \cdot \dot{\mathbf{r}}_i(t) \, dt, \quad (3)
\]

where \( t = 0 \) corresponds to some time after the system has reached a stationary state. \( W_\tau \) represents the steady-state, single-particle contribution to the entropy production \cite{27}, and, in general, it has been proposed as a relevant observable for the thermodynamical description of phase transitions in ABP systems \cite{55,56}. The probability distribution \( P(W_\tau) \) is the object of our investigation. The quantity

\[
I_\tau(w) = -\frac{1}{\tau} \ln P(W_\tau)|_{W_\tau = w_\tau}, \quad (4)
\]

yields the rate function in the large \( \tau \) limit, i.e. \( I(w) = \lim_{\tau \to \infty} I_\tau(w) \).

Fig. 2 shows \( I_\tau(w) \) at \( Pe = 100 \) and \( \phi = 0.5 \), as in Fig. 1, while, in the inset, \( \phi = 0.1 \) (for the same \( Pe \)), as in \cite{59}. Here \( \tau = 10 \), but the structure of \( I_\tau \) is preserved at least up to \( \tau \sim 500 \) \cite{57}, as we will discuss further below. In the homogeneous state (for \( \phi = 0.1 \), see inset), the rate function has a single minimum with a linear branch departing from its left, a fact that was interpreted as due to a condensation transition occurring at smaller-than-average \( w \)’s \cite{59}. In the MIPS region, instead, \( I_\tau(w) \) shows two minima at \( w = w_c \) and \( w = w_d > w_c \), corresponding to the typical values of \( w \) for particles belonging to the cluster (light blue in Fig. 1) and those in the gaseous phase (yellow in Fig. 1). Therefore, this structure is a natural manifestation of the two phases coexisting in the system—one where particles are normally propelled by the active forces and another, jammed, where those forces do a smaller work due to steric hindrances. Fig. 2 also elucidates the different character of our approach with respect to that of \cite{10}, where \( W_\tau \) is summed over all the SPPs. The differences between particles in the gaseous and dense phase are masked in the sum, together with the inhomogeneities in the system. However, the analysis of \cite{10} is more suited to detect some other features of the system, such as collective motion.

Besides an overall similarity of our \( I_\tau \) with usual thermodynamic potentials in first order phase transitions,
The large-\(\tau\) limit of the SCGF, \(G(\tau) = \lim_{\tau \to \infty} G_\tau(s)\), coincides with the Legendre-Fenchel transform (LFT) of \(I(w)\), \(G(s) = \sup_w \{sw - I(w)\}\). When convex, also \(I(w)\) is the LFT of \(G(s)\). The two functions, in fact, can be regarded as thermodynamic potentials associated to a microcanonical (fixed \(W_\tau\)) and canonical (\(W_\tau\) fixed on average by a bias \(s\)) ensembles of trajectories with a given \(W_\tau\), respectively. \(G_\tau(s)\) can be computed directly in our simulations and is shown in Fig. 2a) as a blue solid line; parameters are the same as in Fig. 2b). Despite the finiteness of \(\tau\), there is a good agreement between \(G_\tau(s)\) and the LFT of \(I_\tau(w)\), which is shown in the same figure as a black solid line. Let us then discuss how the different branches of \(I_\tau(w)\) and \(G_\tau(s)\) are mapped into each other, and the nature of the associated particle trajectories.

\(I_\tau(w)\) is well approximated by a Gaussian for \(w\) larger than a certain threshold value \(w^\dagger\). Comparing Fig. 2 with Fig. 1 one concludes that this range corresponds to the yellow particles in the gaseous phase, for which many body effects can be neglected. The corresponding branch of \(G_\tau\), for \(s > s_b \simeq 0.14\) (the slope of the convex envelope between the two minima of \(I_\tau(w)\), red line in Fig. 2b), is quadratic in \(s\), even though it might appear flat on the scales of Fig. 2b. Proceeding towards lower \(w^\dagger\)'s, the concave sector of \(I_\tau(w)\) between the two minima is mapped by the LFT into the single point \(s = s_b\), where the LFT displays a discontinuous first derivative. Typical trajectories contributing to this sector are those of the red particles in Fig. 1. Going further, the fluctuations described by \(I_\tau(w)\) around the minimum in \(w_c\) are also approximatively Gaussian, hence \(G_\tau(s)\) is again parabolic in the corresponding sector \(s_b < s < s_b\) (as for \(s > s_b\), on the scale of the figure this part looks rather flat). The particles which contribute to this branch are the light blue particles of Fig. 1 which are stuck inside the aggregate. For \(w\) smaller than a certain value \(w^\dagger\), \(I_\tau(w)\) is linear, thus its LFT diverges. \(G_\tau(s)\), instead, does not, due to the limited sampling. These fluctuations are originated by the dark blue particles in the cluster of Fig. 1.

Notice that all the information relative to the trajectories of the red particles, contained in the concave branch of \(I_\tau\), is lost when going from \(I_\tau(w)\) to \(G_\tau(s)\), due to the non-involutivity of the LFT of concave functions. This property is strictly connected to statistical ensemble inequivalence and phase transitions [58–63], which extends to the ensemble of trajectories considered here [64]. As a result, when a dynamical phase transition occurs, the transform of \(G_\tau\) does not yield back \(I_\tau\), but only its convex envelope (the red solid line in Fig. 2b).

### Linear branches

\(I_\tau(w)\) is endowed with two linear branches, one to the left of \(w^\dagger\) and the other for \(w < w^\dagger\). The first is produced by those particles in the gaseous phase which, by hitting other disks or small clusters, do a reduced work. This feature of \(I_\tau(w)\) is also manifest at low densities, where MIPS does not occur, as it has been reported in [50]. In-
FIG. 3. Snapshot of a portion of size $250 \sigma_x$ of a system with $N = 1024^2$ particles, at $Pe = 100$ and $\phi = 0.5$. a) Particles are colored according to the hexatic parameter related to the local orientation of the triangular lattice occupied by the particles \cite{65}. b) Same configuration as in a), but colored according to $W_\tau/\tau$, with $\tau = 10$, as in Fig. 4.

FIG. 4. Behavior of the same rate function of Fig. 2 (for $Pe = 50, 100, 200$ from left to right) as $\tau$ is changed. In the upper row $\tau^{-1} \ln P(W_\tau)$ is plotted against $w$, while in the lower row we plot $\tau^{-1} \ln P(W_\tau)$, with $\beta = 0.4, 0.3, 0.2$, respectively, for $Pe = 50, 100, 200$.

deed, this linear branch is better observed in the diluted system, as shown in the inset of Fig. 2, because in this case it extends to all values $w < w^{\dagger}$. In the presence of MIPS, instead, it has to end before the dense-phase minimum at $w_c$. In \cite{50} the linear branch was interpreted as due to a transition at the level of fluctuations.

Let us now discuss the other linear part of $I_\tau(w)$, namely that to the left of $w^{\dagger}$ which, instead, is originated inside the cluster and therefore is only present when MIPS occurs. As it can be seen in Fig. 3 the particles inside the cluster which are able to move, and hence to do some work, are concentrated along the boundaries of the
different hexatically ordered domains, which are coloured differently in Fig. [3]. Most of these particles are red in Fig. [3], meaning that they contribute (together with other red particles in the gaseous phase) to the region between the two minima of $I_\tau(w)$. Inside the cluster some of these particles move and push other disks against their propulsion force. This back-pushed trajectories give rise to the branch with $w < w^{\dag\dag}$. In conclusion, both the linear branches of $I_\tau(w)$ are originated by particles whose work is limited by the dragging due to the others.

On the other hand, a large negative work ($w < w^{\dag\dag}$) is realised by dense-phase particles pushed against their director, a peculiar feature due to activity.

The above scenario for the structure of $P(W_\tau)$ is found not only for different choices of the Péclet number but also for different packing fractions. The exponent $\beta$, however, turns out to be parameter dependent. Furthermore, an analogous structure of the work fluctuations is found for active particles different from colloids, such as the dumbbells shown in Fig.1-2 of the SI.

Finally, we observe that the residence time of particles in each of the two phases (condensed and gaseous) is large but finite. This is shown in Fig. [5] where the probabilities for a generic particle to spend a time $t$ in one of the system phases are plotted. One sees that such probabilities decay as exponentials, with typical times $\tau_{\text{res}} \simeq 300$ or $\tau_{\text{res}} \simeq 100$ for the condensed and gaseous phase, respectively. Hence, for $\tau \gg \tau_{\text{res}}$ each particle has moved between the phases several times. Then, considering values of $\tau$ much larger than those shown in Fig. [4] namely $\tau \gg \tau_{\text{res}}$, one expects both the double-peaked structure of $I_\tau(W_\tau)$ and its scaling properties to change. The smearing of the double-peaked form with $\tau$ can be observed in the upper row of Fig. [4] as well as in inset of Fig. [5].

FIG. 5. Distributions of times for particles persisting inside a cluster (dashed line) and outside the cluster (continuous line), for colloids at $\phi = 0.5$ and $Pe = 100$. A fit using $e^{-t/\tau_{\text{res}}}$ provides $\tau_{\text{res}} = 265$ and $\tau_{\text{res}} = 92$ for particles inside and outside the cluster, respectively. Inset: rate functions for the same case as main figure at $\tau = 500, 1000, 5000 > \tau_{\text{res}}$, showing mixing of peaks.

Scaling with the observation time $\tau$

The above description is entirely based on measurements at fixed $\tau$. We now investigate the $\tau$ dependence of $I_\tau(w)$. Fig. [4] shows the results for different values of $Pe$ and $\phi = 0.5$. As it is clear, the large deviation principle (1) does not hold, except for $w > w^{\dag}$. Due to the LFT duality discussed previously, the corresponding branch of $G(s)(s)$, for $s > s_b$, will also converge to a well defined limit $G(s)$ as $\tau$ grows large.

For $w < w^{\dag\dag}$ the suppression of large fluctuations upon increasing $\tau$ is much slower, resulting in $\lim_{\tau \to \infty} I_\tau(w) = 0$, as it can be seen in Fig. [4]. However, our data suggest that $\lim_{\tau \to \infty} \frac{1}{\tau} \ln \{\text{Prob.}(W_\tau = w)\}$ is finite for a proper choice of $\beta$. For instance, for $Pe = 100$, in the region $w < w^{\dag\dag}$ the best fit yields $\beta \simeq 0.3$. In a shallow region around the minimum at $w = w_c$, a somewhat larger value, compatible with $\beta = 0.5$, is found. These unequal values can be ascribed to the different role played by activity in these two sectors. Indeed, a vanishing work ($w = w_c$) is done by particles stacked into a jammed region where the role of activity is not particularly relevant.

CONCLUSIONS

To sum up, we have studied the large fluctuations of the work done by a tagged particle in a system of self-propelled disks, with parameters set so as to have motility-induced phase separation in steady state. With respect to approaches based on collective variables, such as in [40], we found our single-particle approach particularly suitable when two or more phases coexist in the
system: on the one hand, it reflects the presence of a dense and a gaseous phase via a bimodal structure of the probability distribution; on the other hand, it provides informations on additional features of such phases not observed in passive systems. In the homogeneous state, for instance, one learns about the importance of long-lived micro-clusters [50] for the dynamics. In the phase separated state, instead, fluctuations highlight the continuous rearrangement of the macroscopic cluster, relevant for phenomena such as cage-breaking and fluidisation of the active dense phase [69].

Interestingly, the double peaked probability distribution of the active work in the MIPS phase is endowed with two distinct non-convex branches. Correspondingly, the Legendre-Fenchel transform displays a double singularity, differently from what commonly found for instance in liquid-vapor first order transitions. For very large observation times \( \tau \) we find the bimodal structure to disappear. The physical reason, as discussed, is due to the finite residence time \( \tau_{res} \) in both phases. Thus, for \( \tau \gg \tau_{res} \) each SPP will have been in and out of the cluster many times. This leads to an homogeneisation of fluctuations, whose general structure does not reflect anymore the presence of different phases in the system.

This whole pattern of fluctuations, is shown to be rather general, being observed also in other models of active brownian particles, as in a system of dumbbells and can be considered as an hallmark of an active segregation phenomenon.

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