

Fluctuation relation and heterogeneous superdiffusion in glassy transport

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Current fluctuations and related steady-state fluctuation relation are investigated in simple coarse-grained lattice-gas analogs of a non-Newtonian fluid driven by a constant and uniform force field in two regimes of small entropy production. Non-Gaussian current fluctuations and deviations from fluctuation relation are observed and related to the existence of growing amorphous correlations and heterogeneous anomalous diffusion regimes.

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I. INTRODUCTION

Fluctuation theorem (FT) and nonequilibrium work relations are results of remarkable generality representing an important step toward the formulation of statistical mechanics far from equilibrium [1–3]. FT states that the ratio of probabilities of observing an entropy production W_τ over a long-time interval τ , to that of observing the opposite value, $-W_\tau$, is

$$\frac{\Pi_\tau(W_\tau)}{\Pi_\tau(-W_\tau)} = e^{W_\tau}. \quad (1)$$

Nevertheless, the class of systems obeying Eq. (1) is unknown—even for stochastic dynamics, where FT is most easily derived [4,5]—for it is not clear *a priori* when the asymptotic large-deviation/long-time regime understood in Eq. (1) is attainable and whether it does generally reflect most situations of physical interest. In fact, genuine deviations from Eq. (1) have been early observed [6] and are now well established in various contexts [7–13]. Their general characterization—if they are accidental in nature or rather bring relevant information about the stationary measure—however, remains a widely open problem.

In this paper, I show that deviations respecting the time-reversal invariance of Eq. (1), $W_\tau \rightarrow -W_\tau$, generally occur in a large class of stochastic dynamics and are a signature of glassy correlations. Their origin is traced back to the presence of heterogeneous anomalous diffusion regimes that extend possibly beyond any range of physically accessible values of τ . Two distinct small entropy production limits will be considered: (i) vanishing forcing, i.e., near equilibrium, and (ii) far from equilibrium when currents become small at increasing drive, i.e., a negative-resistance regime. Deviations in the former limit are induced by a transient subdiffusion and tend to decrease at long times. On the contrary, deviations in the latter limit increase with τ and are due to a long-lived superdiffusion regime. Against our naive expectation based on the behavior of relaxational glassy systems (where dynamics is typically subdiffusive in the aging regime), we find that mean-square current fluctuations grow superdiffusively at high density, even though the average current becomes vanishingly small at increasing field. This particularly surprising behavior is suggested to occur generally in driven systems dominated by steric hindrance and cage effect. Systems of experimental relevance include

shear-thickening/jamming fluids, where the glassy correlations are intimately related to dynamic heterogeneity. The existence of nonmonotonic responses is a peculiar feature of nonequilibrium steady state (NESS) and gives us the possibility of testing FT beyond the linear response and in situations which are of physical interest for driven soft matter.

II. MODELS

The models we study are emerging as a new paradigm for the interpretation of glassy phenomena [14]. When driven into a NESS, they show a nonmonotonic dependence of the relaxation time on the applied force, a feature which is qualitatively similar to the behavior of viscosity in sheared concentrated suspensions [15]. Two ingredients characterize the dynamics: (i) the cage effect—a universal feature of glassy systems, which is implemented on a coarse-grained scale through a local kinetic constraint and (ii) a nonconservative force which consists of a uniform and constant drive, allowing for nonzero net current in the NESS. For simplicity, we consider two-dimensional (2D) square lattice systems in which the force is applied along a lattice axes and the kinetic constraint takes the following form: a randomly chosen particle can move to a randomly chosen nearest-neighbor site if (i) the site is empty and (ii) the particle has at most two nearby particles before and after the move. Further, if the move of a mobile particle is attempted in direction opposite to the field, the probability is $e^{-\beta E}$, otherwise it is 1.

The interplay of the two above ingredients gives a rather rich dynamic behavior [15]. At small forces, the cage around a given particle is just slightly distorted allowing easily for particle flow. At increasing forces, however, particles become generally more caged by their neighbors, as moves against the field direction are much less probable. Local rearrangements are thus more difficult and transport more obstructed. As we shall see, this situation leads to non-Gaussian current fluctuations, growing heterogeneous spatiotemporal correlations and anomalous diffusion regimes.

For locally reversible and irreducible Markov chains, such as the one introduced above, the proof of FT is straightforward [5]. One considers the action

$$W_\tau(\{\sigma\}) = \sum_{t=0}^{\tau-1} \ln \frac{w(\sigma_t, \sigma_{t+1})}{w(\sigma_{t+1}, \sigma_t)}, \quad (2)$$

where $w(\sigma, \sigma') \geq 0$ are the transition probabilities for jumping from configuration σ to σ' . If the “border term” B

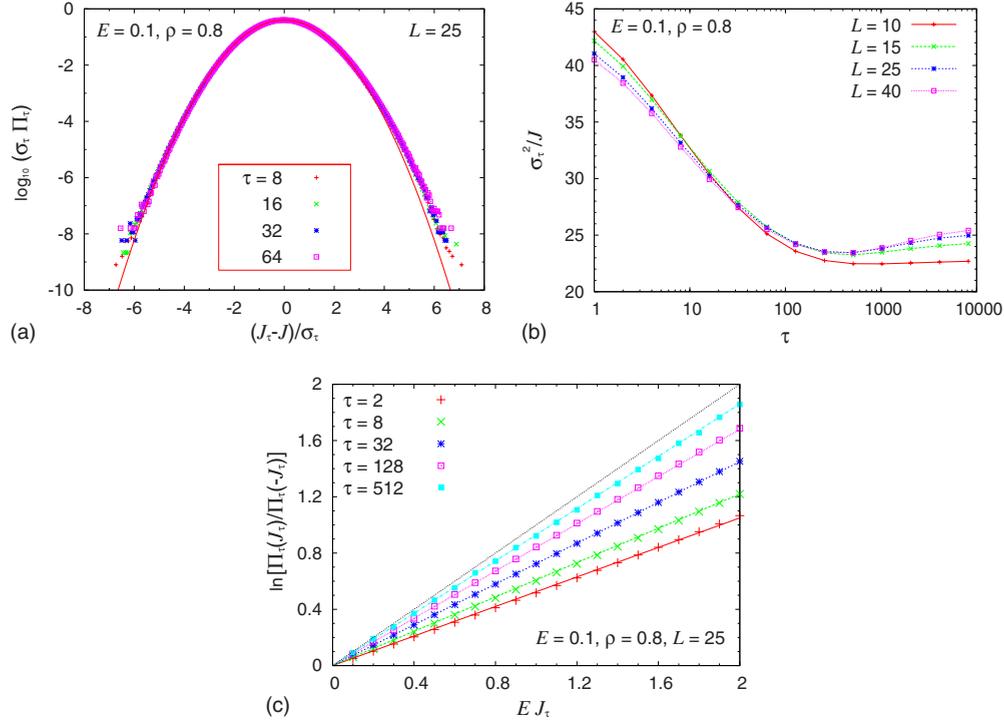


FIG. 1. (Color online) 2D constrained exclusion process driven by a small applied field $E=0.1$ at particle density $\rho=0.8$. The time τ is measured in MCs and L is the linear system size. (a) PDF of current fluctuations in normalized unit. Full line represents the Gaussian. (b) Temporal evolution of relative fluctuations for several system sizes. (c) Logarithmic probability ratio $\ln[\Pi_\tau(J_\tau)/\Pi_\tau(-J_\tau)]$ vs entropy production $W_\tau=EJ_\tau$ ($\beta=1$). Full line, with slope 1, represents the prediction of Eq. (1).

$=\ln[\mu_s(\sigma_\tau)/\mu_s(\sigma_0)]$ (where μ_s is the stationary measure) is subextensively small in τ , then the generating function of the action verifies $\langle e^{-\lambda W_\tau} \rangle = \langle e^{-(1-\lambda)W_\tau} \rangle$, which is just another form of FT. In particular, when μ_s is flat over fixed density configurations, then $B=0$ and FT holds at any time τ . This special case occurs in the asymmetric simple exclusion process (ASEP) to which our model reduces in the absence of constraints. In the presence of constraints, however, the NESS measure is not trivial: while the density profile is flat, particles are statistically more clustered in the transverse direction at larger field. This nonequilibrium fluctuation-induced attraction appears in the transverse pair-correlation function and is a consequence of the more hindered longitudinal transport at increasing field. Nonetheless, while the transverse diffusion slows down (due to the stronger attraction), longitudinal diffusion is enhanced and becomes superdiffusive over a growing range of times at increasing field. Neglecting border terms in this situation is not generally allowed even for large τ and that is the ultimate reason of the observed deviations from Eq. (1). The importance of border terms is discussed thoroughly in Ref. [10]. We now explore their physical implications for driven systems with glassy dynamics.

III. FLUCTUATION RELATION

To test Eq. (1), we perform Monte Carlo simulations of the above-driven stochastic dynamics. A system of linear size L is initialized with a uniform distribution of particles at fixed particle density ρ and is let to reach the NESS. Observ-

ables of interest are evaluated over time intervals of duration τ , along a trajectory of motion lasting $10^7 - 10^9$ Monte Carlo sweeps (MCSs), depending on the system size. In particular, we consider the particle current J_τ that is the signed number of jumps over τ along the applied field, and compute its probability density function (PDF), $\Pi_\tau(J_\tau)$. Since $w(\sigma, \sigma') = \Theta(\text{constraint}) \min\{1, e^{-\beta \vec{E} \cdot \vec{d}r}\}$, local detailed balance holds irrespective of local kinetic constraints. For mobile particles, one has $w(\sigma, \sigma')/w(\sigma', \sigma) = 1, e^{\pm \beta E}$, depending on the relative direction of unit displacement and applied field ($\vec{E} \cdot \vec{d}r = 0, \pm E$). Whereas for immobile particle, $w(\sigma, \sigma')/w(\sigma', \sigma) = 1$ no matter the value of $\vec{E} \cdot \vec{d}r$. The action functional (2) can then be easily identified as $W_\tau = \beta E J_\tau$, consistently with the standard definition of entropy production obtained from the time-dependent Gibbs entropy formula [5]. Thus, computing $\Pi_\tau(J_\tau)$ allows for a direct check of FT. Notice, that the absence of both potential- and kinetic-energy terms makes the PDFs of current, work, and heat exactly identical.

We have first checked that in the standard ASEP current fluctuations are Gaussian distributed and that Eq. (1) is obeyed for time as small as $\tau=1$ MCs, as expected because of flat measure. Similar behavior is found for constrained driven dynamics at small density. At moderately higher densities and small fields, the particle motion becomes weakly correlated and small non-Gaussian tails appear in the PDF of current fluctuations [see Fig. 1(a)]. Since the average current $J = \langle J_\tau \rangle$ does not depend on τ and the PDFs for various values of τ fall on the top of each other when plotted in normalized units, one might naively expect that the asymptotic large-

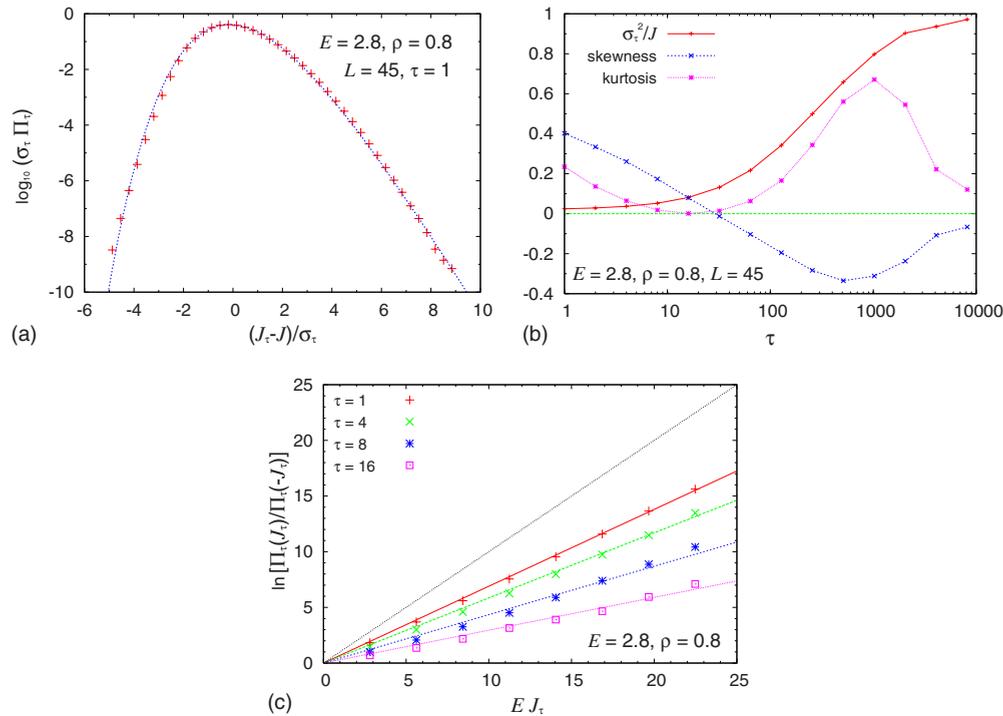


FIG. 2. (Color online) Same as in Fig. 1, but in the negative-resistance regime, with a large applied field $E=2.8$ and $L=45$. (a) The full line is a generalized Gumbel function, with fitting parameter $a=11$ (see [16]). (b) Temporal evolution of PDF high-order moments. Notice that relative fluctuations are rescaled by a factor 65. For systems of linear size $L=45$, the time scale for observing current-reversal events is at most of the order of 10^2 MCs. (c) Deviations from Eq. (1) here increase with τ .

deviation regime has been attained. Actually, current fluctuations, σ_τ^2 , still retain a dependence on τ and increasing the system size makes it harder to reach the asymptotic value [Fig. 1(b)]. The slow decay of current fluctuations leads to deviations from Eq. (1) which become smaller and smaller at increasing τ [see Fig. 1(c)]. Though such deviations are expected to disappear at longer τ , in fact recovering FT maybe difficult at larger density because the Ohmic regime shrinks [15].

On approaching the negative-resistance transport regime, which is a simple rheological analog of shear-thickening behavior [15], something more interesting takes place. First, current fluctuations becomes strongly non-Gaussian and asymmetrically distributed [see Fig. 2(a)]. Similar asymmetric PDFs have been observed in a wide range of systems [17]. Second, although the average current quickly attains its asymptotic value, the PDF keeps evolving with τ , as shown by the behavior of high-order moments in Fig. 2(b). Skewness and kurtosis generally display a nonmonotonous dependence on τ , which is suggestive of that found in the local correlations during the aging dynamics of glassy systems [18] and can be qualitatively understood as follows. On short-time scales, most particles are blocked and only a small fraction of them is able to move in the field direction, thus the PDF is left-skewed. Local rearrangements involving backward steps, however, are needed to sustain the system flow. Indeed, on intermediate time scales, current-reversal events become more probable and, correspondingly, the PDF is less asymmetric. On longer time scales, most particles have moved and the probability of observing negative

current is pretty small (although current-reversal events keep happening on shorter time scales). The PDF thus changes to a right-skewed shape before eventually reaching the Gaussian form typical of diffusive, uncorrelated motion. Figure 2(b) shows that the latter diffusive regime occurs at times that are much beyond the time-scale required for observing negative currents (at least 2 decades larger in the most favorable case, corresponding to parameters of Fig. 2). Therefore, the asymptotic regime of large deviation understood in Eq. (1) is hardly attained, even for such small systems. What type of deviations should we expect in the present case and what features of NESS they possibly encode? In spite of PDF asymmetry, we find that deviations from Eq. (1) are still well described by straight lines with a slope smaller than 1 and their temporal evolution is mainly determined by current fluctuations, σ_τ^2 . However, unlike the previously discussed linear transport regime, the slope appears to *decrease* at longer τ [see Fig. 2(c)]. The numerical extrapolation of the asymptotic slope is difficult in this regime because the accessible values of τ are limited by the larger system-size used. In fact, sampling the PDF requires some care at high density and large force: to avoid finite-size induced dynamical blocking effects (leading to bimodal PDFs), one needs to use moderately larger systems. Since, this obviously decreases the probability of observing current reversal, we consider density barely larger than the negative-resistance threshold. Even in this most favorable situation (in which the saturation current is finite [15]), current-reversal events become unobservable well before the asymptotic regime is attained. The trend of high-order moments thus

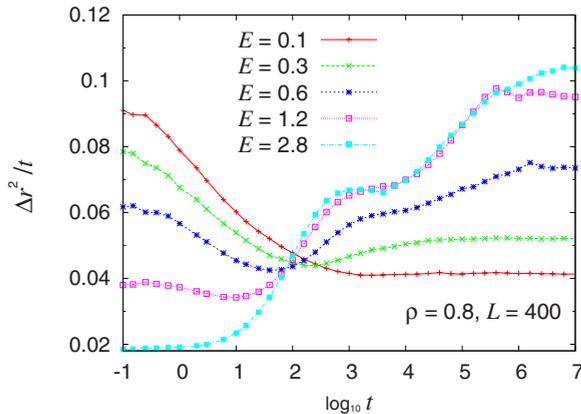


FIG. 3. (Color online) Time averaged longitudinal mean-square particle displacement, $\Delta r_{\parallel}^2/t$, vs time t .

makes us confident that Eq. (1) will never be recovered in any realistic simulation.

IV. HETEROGENEOUS ANOMALOUS DIFFUSION

Further insight into the nature of deviations from Eq. (1) is obtained by looking at the behavior of the longitudinal mean-square displacement relative to the center of mass, $\Delta r_{\parallel}^2(t)$. Since the latter quantity is proportional to σ_r^2 , the physical origin of the above behavior can be traced back to the existence of heterogeneous anomalous diffusion regimes (see Fig. 3). In the linear transport regime, $\Delta r_{\parallel}^2(t)/t$ first decreases with t and then reaches an asymptotic diffusive plateau that increases with the applied field. This enhanced diffusion behavior is also displayed by a driven particle probing an equilibrium glassy environment [19]. On approaching the negative-resistance regime, the early subdiffusion range tends to shrink and connects to the late normal (enhanced) diffusion through an intermediate (logarithmic) superdiffusive transient. Similar nonmonotonic changes of the effective anomalous diffusion exponent have been observed in simulations of biased Brownian motion in a crowded environment [20] and are consistent with some continuous-time random-walk models [21]. In the negative-resistance regime, subdiffusion is strongly suppressed and one only observes a long-lived superdiffusion behavior (see Fig. 3). Although, we expect normal diffusion to occur eventually for any finite system, interestingly, we find that the crossover time to normal diffusion increases with the system size. The appearance of superdiffusion is particularly intriguing in this context. It signals the simultaneous onset of long-time correlations in the motion of particles and the growth of their spatial correlations, as shown by the increasing peak of dynamical sus-

ceptibility [15]. A strikingly similar connection between anomalous diffusion and dynamic heterogeneity has been recently observed in vibrated granular packings, albeit in a distinct jamming/rigidity regime [22] (see also [23]). It is tempting to conjecture that the most peculiar feature distinguishing the approach to the near-jamming/shear-thickening/negative-resistance regime from the glassy behavior is that in the latter, diffusivity tends to vanish, while in the former it grows unboundedly.

Finally, it should be remarked that the results reported above are not specific of 2D: they were observed in three dimensions (3D) for different direction of the applied force/lattice geometry and for different types of kinetic constraints as well.

V. CONCLUSIONS

To conclude, our results generally show that for a large class of driven stochastic dynamics, the asymptotic regime in which the steady-state FT holds exceeds any reasonable physical time scale, even for systems of modest size. The presence of heterogeneous superdiffusion in the negative-resistance transport regime possibly suggests that the large-deviation function might not even exist in the usual sense, if the large-size limit is taken before the long-time one [24]. In spite of the above limitations, we find that deviations from Eq. (1) have several interesting features. First, they encode important physical properties, such as heterogeneous dynamics and anomalous diffusion. The strong “finite-time” deviations from Eq. (1) are a signature of growing long-range dynamical correlations both in time and space and are arguably a universal feature inherent to driven systems with glassy dynamics, such as shear-thickening/jamming fluids. Second, their linear form is the simplest one respecting the time-reversal invariance of Eq. (1), though the asymptotic limit understood in FT is not representative of the physical situation. Finally, they are highly suggestive of the notion of correlation-scale-dependent effective temperature [25]. While the occurrence of longitudinal superdiffusion seems to prevent the applicability of such an appealing concept, we find that transverse fluctuation dynamics is well described by a generalized Einstein relation (in some analogy to systems of driven vortices with random pinning [26]). Whether a modified form of Einstein relation [27,28] does hold for longitudinal fluctuations remains to be seen.

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- [1] R. J. Harris and G. Schütz, *J. Stat. Mech.: Theory Exp.* (2007) P07020.
 [2] F. Ritort, *Adv. Chem. Phys.* **137**, 31 (2008).
 [3] U. Marini Bettolo Marconi, A. Puglisi, L. Rondoni, and A.

- Vulpiani, *Phys. Rep.* **461**, 111 (2008).
 [4] J. Kurchan, *J. Phys. A* **31**, 3719 (1998).
 [5] J. L. Lebowitz and H. Spohn, *J. Stat. Phys.* **95**, 333 (1999).
 [6] M. Sellitto, e-print arXiv:cond-mat/9809186.

- [7] F. Zamponi, G. Ruocco and L. Angelani, Phys. Rev. E **71**, 020101(R) (2005).
- [8] F. Zamponi, F. Bonetto, L. F. Cugliandolo, and J. Kurchan, J. Stat. Mech.: Theory Exp. (2005) P09013.
- [9] A. Puglisi, P. Visco, E. Trizac, and F. van Wijland, Phys. Rev. E **73**, 021301 (2006).
- [10] A. Puglisi, L. Rondoni and Vulpiani, J. Stat. Mech.: Theory Exp. (2006) P08010.
- [11] A. Rákos and R. J. Harris, J. Stat. Mech.: Theory Exp. (2008) P05005.
- [12] D. Villamaina, A. Puglisi, and A. Vulpiani, J. Stat. Mech.: Theory Exp. (2008) L10001.
- [13] A. V. Chechkin and R. Klages, J. Stat. Mech.: Theory Exp. (2009) L03002.
- [14] F. Ritort and P. Sollich, Adv. Phys. **52**, 219 (2003).
- [15] M. Sellitto, Phys. Rev. Lett. **101**, 048301 (2008).
- [16] B. Portelli, P. C. W. Holdsworth, M. Sellitto, and S. T. Bramwell, Phys. Rev. E **64**, 036111 (2001).
- [17] M. Clusel and E. Bertin, Int. J. Mod. Phys. B **22**, 3311 (2008).
- [18] C. Chamon *et al.*, J. Chem. Phys. **121**, 10120 (2004).
- [19] R. L. Jack, D. Kelsey, J. P. Garrahan, and D. Chandler, Phys. Rev. E **78**, 011506 (2008).
- [20] D. Stauffer, C. Schulze, and D. W. Hermann, J. Biol. Phys. **33**, 305 (2007).
- [21] B. Hughes, *Random Walks and Random Environments* (Clarendon Press, Oxford, 1995).
- [22] F. Lechenault, O. Dauchot, G. Biroli, and J.-P. Bouchaud, Europhys. Lett. **83**, 46003 (2008).
- [23] D. A. Head, Phys. Rev. Lett. **102**, 138001 (2009).
- [24] In this respect, it would be interesting to extend to driven systems the Ruelle thermodynamic formalism, as done in J. P. Garrahan, R. L. Jack, V. Lecomte, E. Pitard, K. van Duijvendijk, and F. van Wijland, Phys. Rev. Lett. **98**, 195702 (2007).
- [25] L. F. Cugliandolo, J. Kurchan, and L. Peliti, Phys. Rev. E **55**, 3898 (1997).
- [26] A. B. Kolton, R. Exartier, L. F. Cugliandolo, D. Domínguez, and N. Gronbech-Jensen, Phys. Rev. Lett. **89**, 227001 (2002).
- [27] T. Hanney and M. R. Evans, J. Stat. Phys. **111**, 1377 (2003).
- [28] T. Speck and U. Seifert, Europhys. Lett. **74**, 391 (2006).