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LETTER

## The ratchet effect in an ageing glass

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**Abstract.** We study the dynamics of an asymmetric intruder in a glass-former model. At equilibrium, the intruder diffuses with average zero velocity. After an abrupt quench to  $T$  deeply under the mode-coupling temperature, a net average drift is observed, steady on a logarithmic time scale. The phenomenon is well reproduced in an asymmetric version of the Sinai model. The sub-velocity of the intruder grows with  $T_{\text{eff}}/T$ , where  $T_{\text{eff}}$  is defined by the response–correlation ratio, corresponding to a general behavior of thermal ratchets when in contact with two thermal reservoirs.

**Keywords:** structural glasses (theory), transport properties (theory), slow relaxation and glassy dynamics, diffusion

In an irreversible environment, thermal fluctuations can be rectified in order to produce a directed current. Following a few fundamental examples of historical and conceptual value [1, 2], in the last twenty years a huge number of devices and models—usually known as Brownian ratchets or motors—have been proposed [3]–[5]. The purpose of these models is often practical, e.g. the extraction of energy from a highly fluctuating environment, such as a living cell [6, 7]. But Brownian ratchets are also valid probes for the non-equilibrium properties of the fluctuating medium, the value of the current being sensitive to the interplay of different time scales as well as the different temperatures at work [8, 9].

The simultaneous breaking of spatial and time-reversal symmetries is a necessary condition for observing directed motion. A classical example of this mechanism (see for instance [3]), the so-called flashing ratchet, consists of a particle undergoing standard overdamped diffusion and also subject to a zero-average space-asymmetric (e.g. sawtooth) potential. If this potential is switched on and off according to a random/periodic time sequence, a current of particles (steady on average) can be observed. This current is driven by the energy injected into the system when switching on the potential. Another way of breaking the time-reversal symmetry is obtained by coupling the system with reservoirs at two different temperatures: an asymmetric intruder in such a multi-temperature environment displays an average drift, performing as a Brownian motor. For many-body systems this has been done, for instance, in [10, 11].

The present work, inspired from the latter scenarios with a continuous flow of energy in a non-thermalized medium, shows a study of the ratchet phenomenon in the ageing dynamics of fragile glass-formers. It is well known that such systems, when quenched below their mode-coupling temperature, display an out-of-equilibrium dynamics customarily described within a two-temperature scenario [12]–[15]. Fast modes are equilibrated at the bath temperature while slow modes remain at a higher effective temperature  $T_{\text{eff}}$ . Here we show that the energy flowing from slow to fast modes can be rectified to produce a directed motion. The properties of the observed current characterize the non-equilibrium behavior of the glass. In particular, a striking monotonic relation is observed, between the ratchet sub-velocity and  $T_{\text{eff}}/T$ . The experience with other kinetic models of ratchets [10, 11] teaches us that—when in the presence of a temperature imbalance—the heat flux also governs the ratchet velocity. These observations suggest the conjecture that one can obtain the effective temperature by replacing the measure of linear response and correlations with the simple measure of an average current. In what follows we test the reliability of this procedure.

The first of the numerical experiments proposed here involves the 3D soft-spheres model, which is a well known fragile glass-former [16]–[19]. The thermodynamic properties of soft spheres are controlled by a single parameter  $\Gamma = \rho T^{-1/4}$ , which combines the temperature  $T$  and the density  $\rho$  of the system, with  $\rho = N/V\sigma_0^3$  and  $\sigma_0$  the radius of the effective one-component fluid. We study a binary mixture (50:50) with radii ratio  $\sigma_1/\sigma_0 = 1.2$ . Particles interact via the soft potential  $U(r) = [(\sigma_i + \sigma_j)/r]^{12}$  and the dynamics is evolved via a local Monte Carlo algorithm. Time is measured in Monte Carlo steps (one step corresponds to  $N$  attempted MC moves). The model has a dynamical crossover at a mode-coupling temperature  $T_{\text{MC}}$  corresponding to the effective coupling  $\Gamma_{\text{MC}} = 1.45$  [16]. The ratchet is formed by the asymmetric interaction of a single particle, the intruder, with all the other particles of the system. Denoting with  $x_i, i > 0$ , the  $x$

coordinate of the  $i$ th particle, and with  $x_0$  the abscissa of the intruder, we choose

$$U(\mathbf{r}_0, \mathbf{r}_i) = \begin{cases} U(|\mathbf{r}_0 - \mathbf{r}_i|) & \text{if } x_i < x_0, \\ \varepsilon U(|\mathbf{r}_0 - \mathbf{r}_i|) & \text{otherwise,} \end{cases} \quad (1)$$

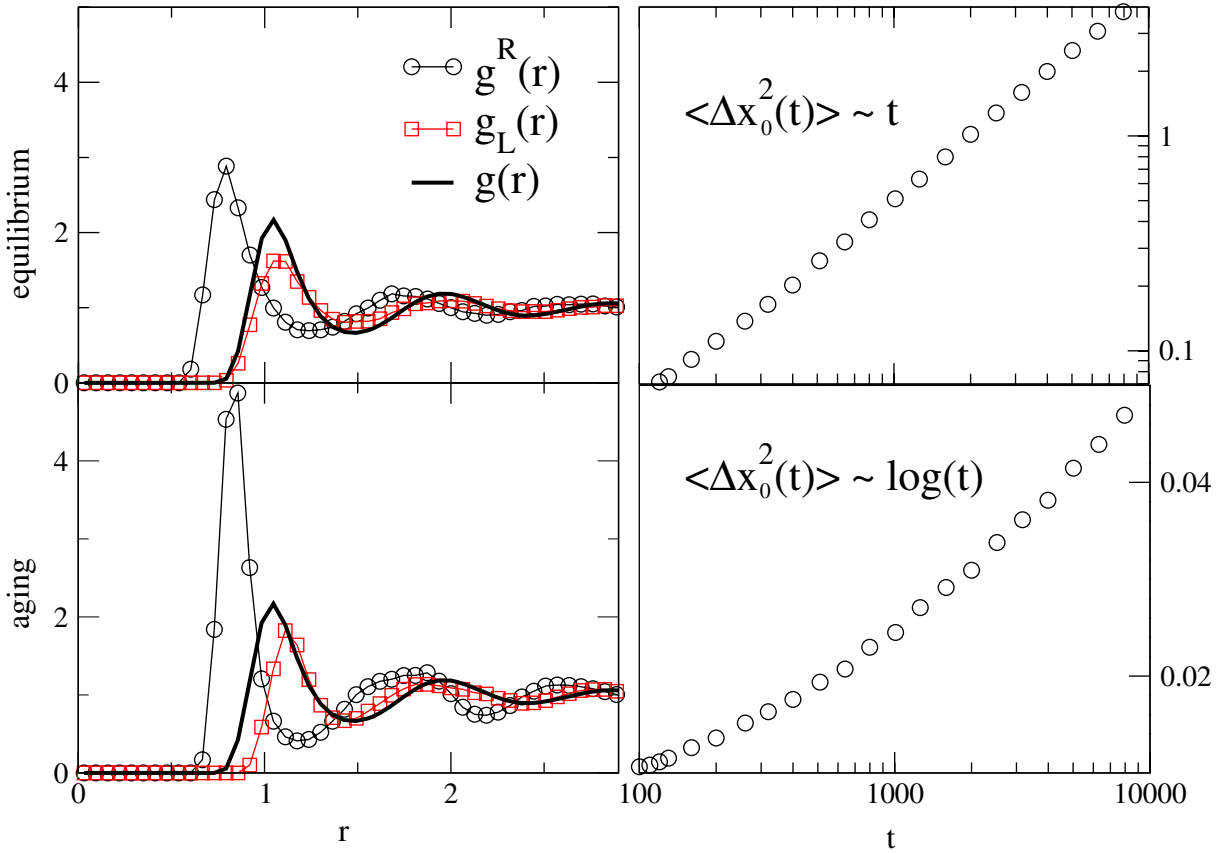
with  $\varepsilon = 0.02$ . The spatial symmetry along the  $x$  axis is therefore broken, fulfilling one of the requirements for getting a ratchet device. Let us stress that here, in analogy with the model proposed in [20], the asymmetry is inherent to a single object, the intruder, which is embedded in a symmetric environment, at variance with flashing and rocket ratchets [5]. Moreover, by exploiting condition (1), we are able to build an intruder with an intrinsic asymmetry, even if it is a point-like particle.

We equilibrate configurations embedding an asymmetric particle at a high temperature  $T_{\text{liq}} \gg T_{\text{MC}}$ . At  $T_{\text{liq}}$  the system has simple liquid behavior with fast exponential relaxation. The dynamics of the asymmetric intruder is then studied both along equilibrium trajectories and after quenches to different temperatures  $T < T_{\text{MC}}$ . The average displacements of the intruder along different axes are compared at and away from equilibrium, namely both  $\langle \Delta x_0(t) \rangle$  and  $\langle \Delta y_0(t) \rangle$  are studied, with  $\Delta x_0(t) = x_0(t) - x_0(t_0)$ . In particular  $t$  is the time elapsed since the instantaneous quench, namely we take  $t_0 = 0$ . The averages denoted by  $\langle \dots \rangle$  are realized by considering 5000 initial configurations with a single intruder, equilibrated at  $T_{\text{liq}} = 4.42 T_{\text{MC}}$ , each followed by an independent thermal history. Quenches are done to  $T = 0.67 T_{\text{MC}}$ ,  $0.53 T_{\text{MC}}$ ,  $0.42 T_{\text{MC}}$ ,  $0.31 T_{\text{MC}}$ .

Let us consider first the effect of the asymmetric interaction on the arrangement of particles around the intruder. We study the pair distribution function of neighbors to the right and to the left of the intruder,  $g_{\text{L}}^{\text{R}}(r|\delta x \gtrless 0) = \sum_{j|\delta x_j \gtrless 0} \delta(r_{j0} - r)$ , with  $r_{j0}$  the distance between the intruder and the  $j$ th particle, in equilibrium or at a fixed elapsed time after the quench (see figure 1, left panels). Right neighbors stay closer to the intruder due to the reduced repulsion. This asymmetric clustering of neighbors is slightly enhanced during ageing, but at this level of analysis there is no qualitative difference between the equilibrium and off-equilibrium regimes.

Dynamical measurements, in contrast, show important differences between the two regimes. Let us consider the mean square displacement around the average position at time  $t$  of the intruder particle (figure 1 right): it is linear with time at high temperatures, while it is logarithmic after a quench,  $\langle \Delta x_0^2(t) \rangle - \langle \Delta x_0(t) \rangle^2 \sim \log t$ . This behavior, typical of activated dynamics in a rough potential [21, 17, 22], is also observed for host particles. The behavior of the average displacement of the intruder,  $\langle \Delta x_0(t) \rangle$ , is more striking. At equilibrium (figure 2, black circles) there is no net displacement: this is because parity along the  $x$  axes is broken whereas time-reversal symmetry is preserved. The same happens for  $\langle \Delta y_0(t) \rangle$  (green diamonds) during ageing: in this case only (macroscopic) time-reversal symmetry is broken. But parity and time-reversal symmetry are both violated for  $\langle \Delta x_0(t) \rangle$  after the quench (red squares), and in this case a net average drift is found (figure 2), linear on a logarithmic time scale,  $\langle \Delta x_0(t) \rangle \sim \tau$  with  $\tau = \log^{1/2} t$ . The logarithmic time scale again points to a non-equilibrium phenomenon ruled by activated events. Moreover, the simple scaling relation  $\langle \Delta x_0(t) \rangle \sim \sqrt{\langle \Delta x_0^2(t) \rangle - \langle \Delta x_0(t) \rangle^2}$  between the displacement along the  $x$  axis and the m.s.d. around the average position is observed, in analogy with the Sinai model discussed below.

Our first exploration of ratcheting effects in glassy models leaves open one important question, namely that of the dependence of the drift on the parameters (e.g. temperature)

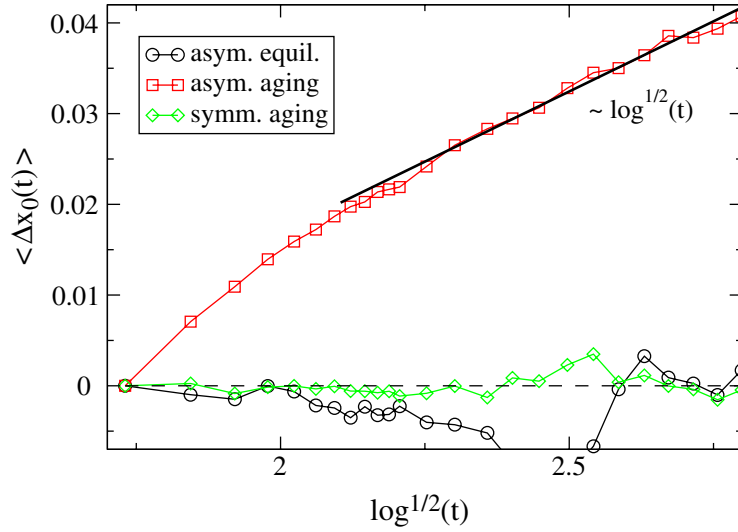


**Figure 1.** Left panels: pair distribution function centered on the intruder,  $g_L^R(r|\delta x \geq 0)$ , of the left (red squares) and right (black circles) neighbors, at equilibrium (top) and at a fixed elapsed time after the quench (bottom). Black line:  $g(r)$  for symmetric interactions. Right panels:  $\langle \Delta x_0^2(t) \rangle$  at equilibrium (top) and after the quench (bottom).

of the system. In order to sketch a preliminary answer, we need to extract from the curve  $\langle \Delta x_0(t) \rangle$  a synthetic observable. The finding of a logarithmic time scale  $\langle \Delta x_0(t) \rangle \sim \tau$  with  $\tau \sim \log^{1/2} t$  suggests defining an average sub-velocity as [23]

$$v_{\text{sub}}(t, t_w) = \frac{\langle \Delta x_0(t) \rangle - \langle \Delta x_0(t_w) \rangle}{\delta \tau} = \frac{\langle x_0(t) \rangle - \langle x_0(t_w) \rangle}{\delta \tau}, \quad (2)$$

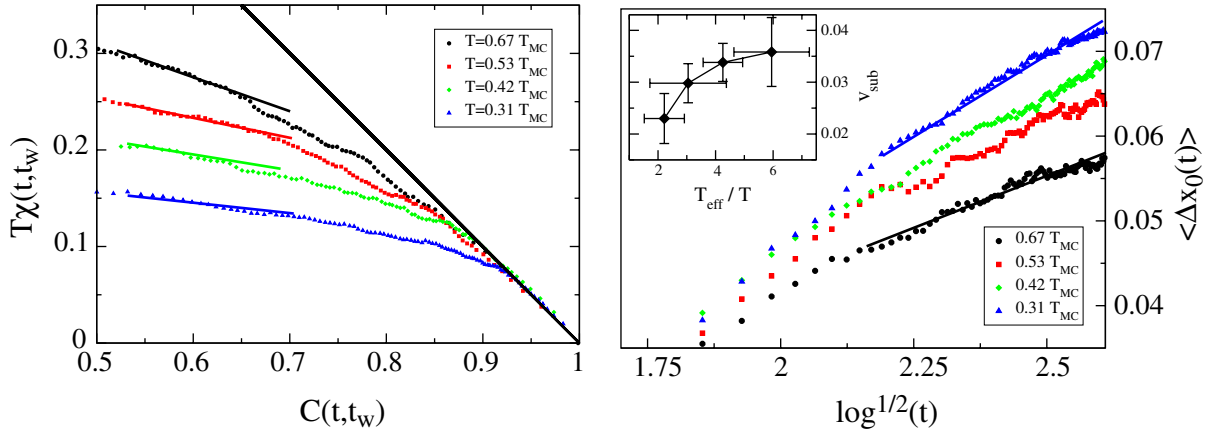
with  $\delta \tau = \log^{1/2} t - \log^{1/2} t_w$ . This average sub-velocity depends in general on both the running time  $t$  and the waiting time  $t_w$  elapsed since the quench. More precisely, considering figure 2, by fixing  $t_w$  and  $t$  we choose the time lag where the slope of the curve  $\langle \Delta x_0(t) \rangle$  is measured. Clearly, the instantaneous sub-velocity only depends on  $t_w$  and corresponds to a slowly decaying velocity  $v(t_w) = d\langle \Delta x_0(t_w) \rangle / dt_w \sim 1/t_w$  (with logarithmic corrections). For large enough waiting times we can define the ‘order parameter’  $v_{\text{sub}}$ , namely we find  $v_{\text{sub}}(t_w) \sim \text{const}$ , and then we probe its dependence upon the external parameters. Here we focus on the quench temperature  $T$ , drawing a connection between the drift of the glassy ratchet, which is a pure non-equilibrium effect, and the more customary equilibrium-like descriptions of ageing media in terms of effective



**Figure 2.** Average intruder displacement:  $\langle \Delta x_0(t) \rangle$  for equilibrium trajectories (black circles), and  $\langle \Delta x_0(t) \rangle$  and  $\langle \Delta y_0(t) \rangle$  after a quench far below  $T_{MC}$  ( $T = 0.42T_{MC}$ ; red squares and green diamonds respectively). The displacement  $\langle \Delta x_0(t) \rangle$  is measured in units of the average inter-particle distance, as obtained from the position of the first peak in the pair distribution function  $g(r)$  (see also figure 1).

temperatures. In the right panel of figure 3 we show the drift of the asymmetric intruder for four quench temperatures:  $T = 0.67T_{MC}$ ,  $0.53T_{MC}$ ,  $0.42T_{MC}$ ,  $0.31T_{MC}$ . We observe that on decreasing the quench temperature the drift grows in intensity, i.e. the sub-velocity increases. This is somewhat stronger evidence that the ratchet drift cannot be described as an equilibrium-like effect, e.g. trying to connect average kinetic or potential energy with a mobility: one would expect that at lower temperatures everything is slowed down, whereas we find that the ratchet drift is enhanced.

Typical examples of ratchets in (ideally) statistically stationary configurations are obtained by coupling the system with two or more reservoirs. It is the existence of different temperatures within the same system which allows the production of work without violations of the second principle of thermodynamics. But what is the second temperature in a glassy system? According to the well-established description of the ageing regime of glasses [13], it is the effective temperature defined as the violation factor of the fluctuation-dissipation theorem (FDT). The quench of a fragile glass, for instance in our soft-spheres model, below its mode-coupling temperature, produces ageing and violations of FDT. The factor  $X(t, t_w) < 1$  which allows one to write a generalized FDT,  $T\chi(t, t_w) = X(t, t_w)[C(t, t) - C(t, t_w)]$ , with  $\chi(t, t_w)$  the integrated response and  $C(t, t_w)$  the correlation, yields the definition of the effective temperature  $T_{\text{eff}}(t, t_w) = T/X(t, t_w)$ . The latter is usually higher than the bath temperature  $T_{\text{eff}}(t, t_w) > T$  and is understood as the temperature of slow, still not equilibrated, modes. Clearly, the ratio  $T_{\text{eff}}/T$  may be regarded as the parameter which tunes non-equilibrium effects and we study here how the glassy ratchet drift depends on it. We obtain  $T_{\text{eff}}$  from the parametric plot of  $C(t, t_w)$ , taken as the self-intermediate scattering function, versus the integrated response  $T\chi(t, t_w)$ ,



**Figure 3.** Left panel: parametric plot  $T\chi(t, t_w)$  versus  $C(t, t_w)$ . Right panel: asymmetric intruder drift for different quench temperatures; linear fits yield sublinear velocities. Inset:  $v_{\text{sub}}$  versus  $T_{\text{eff}}/T$ . In both panels colors of data correspond to different effective temperatures:  $T_{\text{eff}} = 1.497 T_{\text{MC}}$  (black),  $1.613 T_{\text{MC}}$  (red),  $1.792 T_{\text{MC}}$  (green) and  $1.819 T_{\text{MC}}$  (blue).

measured according to the field-free method of [24]. The parametric plot  $T\chi(t, t_w)$  versus  $C(t, t_w)$  is shown in the left panel of figure 3 for different temperatures.

At large times,  $v_{\text{sub}}$  exhibits finite-size effects, namely the drift saturates at a time that can be increased by increasing the size of the simulation box. This is why we compute  $v_{\text{sub}}$  by fitting data only in a relatively early time window, namely  $10 < t_w < 10^3$ . Accordingly, the measure of  $X(t, t_w)$  is obtained from the parametric plot  $T\chi(t, t_w)$  versus  $C(t, t_w)$  with  $t = 10^3$  and  $t_w$  ranging from 10 to  $10^3$ , namely the early ageing regime is considered. The inset of the right panel of figure 3 shows the behavior of  $v_{\text{sub}}$  versus  $T_{\text{eff}}/T$ , revealing that the sub-velocity increases when  $T_{\text{eff}}/T$  is increased. Namely the intensity of the ratchet effect, traced in the measure of  $v_{\text{sub}}$ , grows as the distance from equilibrium is increased.

Let us remark here that the mechanism governing our ratchet differs from that of a flashing ratchet [3]. Irreversibility is achieved by choosing an initial condition which is out of equilibrium with respect to the bath temperature  $T_{\text{liq}} \neq T$ . The extreme slowness of ageing of glass prevents the system from thermalizing, so energy continuously flows from fast to slow modes, supplying power to the Brownian ratchet. At variance with the flashing ratchet case, here there is no external time-dependent modulation of a potential. The movement of surrounding molecules produces fluctuations of the intruder potential, but these obey a globally conservative dynamics and cannot produce energy.

Indeed, simpler models can be conceived, where the intruder moves in a time-independent potential: as an enlightening example, in the following we discuss the Sinai model, where it is made clear how irreversibility comes solely from the choice of initial conditions. The Sinai model is one of the simplest describing the diffusion of a single particle through a random correlated potential [25]. Its long-time dynamics is ruled by activated events and is characterized by a logarithmic time scale: for this reason it appears to be a candidate well fitted for reproducing the previous experiment in a more controlled setup. In the original Sinai model the random potential is built from a random walk of the force on a 1D lattice. The force  $F_i$  at each lattice site  $i$  is an independent identically

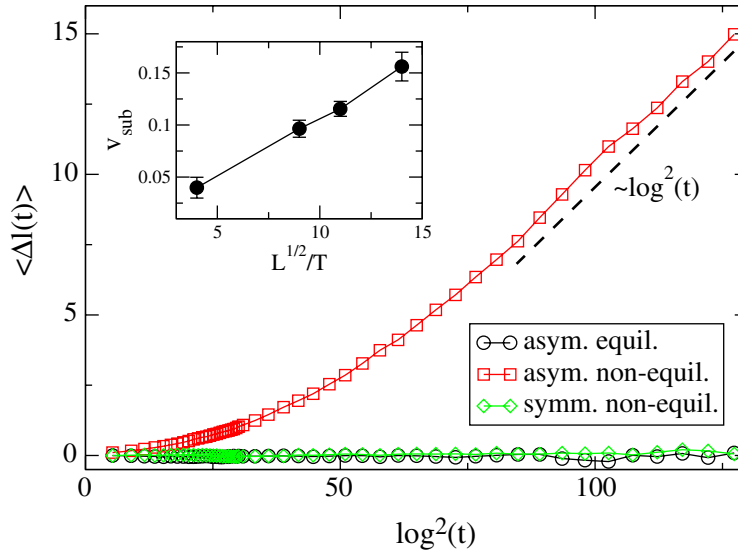
distributed random variable extracted from a zero-mean symmetric distribution  $p(F)$ . The potential on a lattice site  $n$  is given by  $U(n) = \sum_{i=1}^n F_i$ . Because  $\int dF p(F)F = 0$ , the average force experienced by a particle is zero. The potential excursion between two sites grows like  $\langle |U(i) - U(j)| \rangle \sim |i - j|^{1/2}$ . The above relation, together with the expression for the typical time needed to jump a barrier,  $\sim \exp(\beta\Delta U)$ , yields the long-time scaling of the mean square displacement of  $\langle \Delta l^2(t) \rangle \sim \log^4(t)$ , with  $l(t)$  denoting the site occupied by the particle at time  $t$  and  $\Delta l(t) = l(t) - l(0)$ . A logarithmic time scale for the growth of the domain size is quite ubiquitous in the low temperature regime of glassy systems, where activated processes dominate [26]. The time-reversal symmetry breaking is inherent in the Sinai model: by averaging over initial positions extracted from a flat distribution one reproduces an initial infinite temperature  $T_{\text{iq}} = \infty$ , while the quench to a glassy phase is reproduced by evolving the system at temperature  $T \ll \sqrt{L}$  where  $L$  is the linear size of the system. We propose a spatially asymmetric version of the Sinai model, where the symmetric force distribution  $p(F)$  is replaced with an asymmetric one  $\tilde{p}(F)$ , in analogy with the asymmetric potential of the glassy ratchet, in order to find a non-equilibrium drift. Namely the distribution  $\tilde{p}(F)$  is such that  $\tilde{p}(F) \neq \tilde{p}(-F)$ , but still  $\int dF \tilde{p}(F)F = 0$ . In particular, the random force is obtained according to the following procedure: at each site the sign of a random variable  $f_i$  is chosen with probability 1/2 and its modulus, in agreement with the sign, from either  $e^{-|f_i|/\lambda_-}$  or  $e^{-|f_i|/\lambda_+}$ , with  $\lambda_+ > \lambda_-$ . In the present simulation,  $\lambda_+ = 1$  and  $\lambda_- = 0.2$  have been used. The potential is then built as  $U(n) = \sum_{i=1}^n F_i$ , with  $F_i = f_i - (\lambda_+ - \lambda_-)/2$ . This amounts to a shift of the whole distribution such that  $\langle F \rangle = 0$ .

The results of the simulations of this model are shown in figure 4: they clearly show a behavior strikingly similar to those of figure 2 for the glass-former model: a drift is observed only when both time-reversal and spatial symmetry are broken. Indeed, as demonstrated by the black and red solid curves, it is sufficient that one of the two symmetries is restored to have a zero drift. In particular, to obtain an ‘equilibrium’ dynamics in this model it is sufficient to distribute all initial positions of the simulation according to a distribution  $\sim \exp[-\beta U(i)]$ : in this case, even with the asymmetric distribution of forces described above, the average drift is zero. The large time behavior of the drift is compatible with a squared logarithm,  $\langle \Delta l(t) \rangle \sim \log^2(t) \sim \sqrt{\langle \Delta l^2(t) \rangle}$ , in agreement with what was observed previously for the glassy ratchet.

The out-of-equilibrium dynamics of the Sinai model closely reproduces these observations: the inset of figure 4 shows the sub-velocity as a function of  $\sqrt{L}/T$ , with  $T \ll L^{1/2}$  the quench temperature and  $L$  the size of the linear chain, which fixes the most relevant energy scale of the model, i.e. the maximum depth of potential minima. The inset of figure 4 shows that  $v_{\text{sub}}$  grows monotonically with  $\sqrt{L}/T$ : when the last quantity is increased, particles condense at the bottom of the deepest valleys. The monotonic increase of  $v_{\text{sub}}$  with  $\sqrt{L}/T$  signals that also for the Sinai model the larger the distance from equilibrium the larger the velocity of the drift.

In conclusion, through numerical simulations in different models and different choices of the quench temperature, always chosen in the deep slowly relaxing regime, we have given evidence of the existence of a ‘glassy ratchet’ phenomenon. The drift velocity slowly decays in time and can be appreciably different from zero for at least three orders of magnitude in time. The overall intensity of the drift, measured in terms of a ‘sub-velocity’, is monotonically increasing with the distance from equilibrium, i.e. with the





**Figure 4.** Main: average displacement in the Sinai model. Black circles: diffusion in an asymmetric potential at equilibrium; green diamonds: diffusion during ageing in a symmetric potential (quenched from  $T = \infty$ ); red squares: diffusion during ageing in a asymmetric potential. Inset: the sublinear drift velocity  $v_{\text{sub}}$  grows with  $\sqrt{L}/T$ .

difference between the quench and effective temperatures. This observation supports the idea of regarding the ratchet drift as a ‘non-equilibrium thermometer’: it can be used as a device capable of indicating how far a system is from equilibrium. Nevertheless, for such a thermometer to be effective, a more accurate calibration procedure should be carried out. Namely, one should verify that  $v_{\text{sub}}$  is a function of  $T_{\text{eff}}$  and  $T$  only with a small number of parameters. In this case the calibration of our thermometer would require a small independent measure of  $T_{\text{eff}}$  to fix those parameters.

As an experimental realization of our glassy ratchet, one should consider a particle with an anisotropic interaction with the surrounding ones with a small magnetic dipole placed on it, orthogonal to the asymmetry axis. Then, switching on a constant magnetic field, the orientation of the two faces of the ‘Janus’ [27] particle is preserved with respect to a fixed reference frame, so the spatial symmetry of the interaction is broken. Recent theoretical and experimental advances in the study of functionalized or ‘patchy’ particles [28, 29] promise an experimental verification of our hypothesis in the near future.

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