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Oscillatory athermal quasistatic deformation of a model glass

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We report computer simulations of oscillatory athermal quasistatic shear deformation of dense amorphous samples of a three-dimensional model glass former. A dynamical transition is observed as the amplitude of the deformation is varied: For large values of the amplitude the system exhibits diffusive behavior and loss of memory of the initial conditions, whereas localization is observed for small amplitudes. Our results suggest that the same kind of transition found in driven colloidal systems is present in the case of amorphous solids (e.g., metallic glasses). The onset of the transition is shown to be related to the onset of energy dissipation. Shear banding is observed for large system sizes, without, however, affecting qualitative aspects of the transition.

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Understanding the behavior of materials under mechanical deformation is of primary importance for many contexts. While the deformation behavior of crystals is theoretically well understood, no universally accepted framework exists to rationalize the behavior of mechanically driven amorphous systems, although significant progress has been made in recent years in developing a detailed understanding of how an amorphous solid responds to external stresses [1–4]. Considerable recent activity has been spurred by an interest in the mechanical behavior of metallic glasses, soft glassy materials, foams, and granular packings and has involved theoretical, computational, and experimental investigations [2,5–9]. Particular interest is understandably focused on the manner in which the response of an amorphous solid changes from a nearly elastic response at small applied stress to a state in which it will flow (or break) for large applied stress.

Many computational investigations have employed the approach of studying the zero-temperature behavior of amorphous solids under quasistatic conditions [using an athermal quasistatic (AQS) procedure; see, e.g., [10]]. In this procedure, systems are kept in local energy minimum configurations or inherent structures [11,12] while varying the strain. In previous work on model systems (binary mixture liquid with Lennard-Jones interactions) it has been shown that upon monotonically increasing the applied shear strain, the inherent structures evolve towards energies corresponding to the limit of high temperatures [13]. This and related phenomena are referred to as rejuvenation, in contrast to the well studied process of aging whereby (typically) a glassy material descends to deeper energy configurations as a function of the waiting time over which it relaxes at a given temperature. In contrast, when a cycle of strain is applied up to a maximum value that is then reversed [14], both ageing and rejuvenation are observed, with small amplitudes found to reduce the energy of samples ("overage" them), while larger-amplitude strains tend more often to increase the energy (thus "rejuvenating" the samples). Initial conditions of the samples in such cases matter: Samples with lower initial potential energy are rejuvenated more easily than those with higher energy [14].

It has been observed that the nature of rearrangements of colloidal particles in a suspension subjected to cyclic shear deformation depends on the amplitude of strain. For small amplitudes the particles become quiescent after a short transient, whereas for larger amplitudes a finite fraction of the colloidal particles move irreversibly. This observation has been made for low packing fractions [15,16] and for dense colloidal suspensions [17,18]. The latter is close to the present study, while the former is a radically different situation. It has been argued that such a nonequilibrium transition from a quiescent to an active state belongs to the universality class of conserved directed percolation [19].

In this Rapid Communication we report results of computer simulations for a three-dimensional binary Lennard-Jones model, wherein we subject samples to a large number of shear strain cycles (rather than to a single cycle as reported in [14]). For large enough amplitudes, our systems exhibit diffusive behavior and reach steady states that are independent of their initial conditions, characterized by a diffusion coefficient and an average energy that depend on the maximum strain only. For low amplitudes of the strain, different samples reach instead nondiffusive states, retaining a memory of their initial conditions. Our results thus indicate that sheared amorphous solids undergo the same type of quiescent to active transition as driven colloidal suspensions at moderate density [16], suggesting that the diffusion behavior observed in driven colloidal suspensions could occur in completely different cases, such as in fatigue experiments on metallic glasses [6,20,21]. In addition, we find that the critical amplitude γ_c coincides with a dramatic change in the amount of energy dissipated per deformation cycle (the area of the hysteresis curve), occurring close to but slightly below the yield strain γ_y of undeformed samples. Finally, we show that large systems subjected to deformation above the critical value present shear banding, but this leaves the dynamical transition picture unaffected.

We simulate a binary Lennard-Jones mixture with parameters and an interaction cutoff as in [14], using LAMMPS

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[22]. Cubic samples with $N = 500{,}4000{,}32\,000$ particles are equilibrated at two different reduced temperatures ($T = 1.0, 0.466$) at reduced density 1.2. The higher temperature corresponds to the onset of slow dynamics whereas the lower temperature, close to the lowest temperatures studied for this system, corresponds to a supercooled state [23]. Configurations sampled from the equilibrium trajectory are subjected to energy minimization, using the conjugate-gradient algorithm, to obtain sets of inherent structures typical of the liquid at temperature T. The inherent structure energies sampled by the liquid are lower for the lower temperatures [14].

Each such inherent structure is then subjected to two operations. First, particle positions are transformed affinely (in this case, we transform the x coordinates according to $r_x' = r_x + d\gamma r_y$ while keeping r_y and r_z the same), so that the shear strain γ of the samples is incremented by a small amount $d\gamma$, and Lees-Edwards boundary conditions [24] are updated accordingly, thus changing the original energy landscape (so that the initial configuration is no longer a minimum). Subsequently, energy minimization is performed using the conjugate gradient algorithm to obtain the inherent structure in the new energy landscape. These two operations represent an AQS step [10]. If AQS steps are iterated, the sample can be sheared to arbitrary values of strain. The dynamics under such a protocol can be assumed to mimic real shear deformation experiments when the temperature of the system is low enough to be neglected (hence the name athermal) and shear rates are low enough (quasistatic) to let the system relax fully before further deformation takes place [10]. In what follows, the shear strain γ is changed in steps of $d\gamma$ ($=10^{-3}$, 2×10^{-4}, and 2×10^{-4}, respectively, for $N = 500$, 4000, and 32 000) from 0 to a maximum value $\gamma_{\max}$ and then to $-\gamma_{\max}$ and back to 0. This operation is the elementary cycle of our dynamics. For practical purposes we define the accumulated strain γ_{acc} as the sum of the elementary absolute strains applied at all steps: $\gamma_{\text{acc}} = \sum_i |d\gamma_i|$. The accumulated strain plays the role of time in our analysis. At the end of each cycle, particle positions in configurations with $\gamma = 0$ (zero-strain configurations) are stored, so that the mean square displacement with respect to the initial configuration ($\gamma_{\text{acc}} = 0$) can be measured. Energy and all the components of the stress tensor are also recorded at every AQS step [but, except in Fig. 3(b), we report only the zero-strain values at the end of each cycle]. About 40, 10, 2 samples are subjected to the protocol above respectively for the systems with $N = 500$, 4000, 32000, so the quantities above can be averaged.

The average potential energy of the minima visited is presented in Fig. 1(a) as a function of the number of cycles. The behavior of the potential energy depends on the value of $\gamma_{\max}$. For low values of $\gamma_{\max}$, after a short transient, the energy curves reach a value that depends on the initial T. For high enough values of $\gamma_{\max}$, the energy converges to the same $\gamma_{\max}$-dependent value for both initial values of T and remains steady. We expect this result to hold for other temperatures as well. Whether overaging or rejuvenation will occur, therefore, depends on whether the initial energy of the undeformed inherent structure is lower or higher than the steady state energy. At intermediate values of $\gamma_{\max}$, the convergence of the energy to the asymptotic values requires more cycles than at the extreme $\gamma_{\max}$ values. To characterize this slow relaxation, we fit the transient behavior of the energy data in Fig. 1(a) with a stretched exponential, from which a characteristic relaxation strain $\tilde{\gamma}_{\text{acc}}$ is extracted. As can be seen from Fig. 1(b), the values of $\tilde{\gamma}_{\text{acc}}$ become high for intermediate $\gamma_{\max}$ and show behavior consistent with a divergence at a critical strain, similarly to what is observed in driven colloidal suspensions [16]. In order to understand the nature of particle motions that lead to the energy relaxation (or lack thereof), we calculate the mean square displacements (MSDs) from the initial configurations, averaged over the samples. The MSD curves shown in Fig. 2 indicate that for small values of $\gamma_{\max}$ the MSD curves saturate after an initial transient, whereas for large $\gamma_{\max}$ the particle displacements are diffusive, i.e., the MSD depends linearly on the accumulated strain beyond some value of γ_{acc}. In the case where a steady state is reached, the diffusivity can be extracted by a linear fit of the MSD calculated from an initial point in the steady state. In terms of the accumulated strain relative to

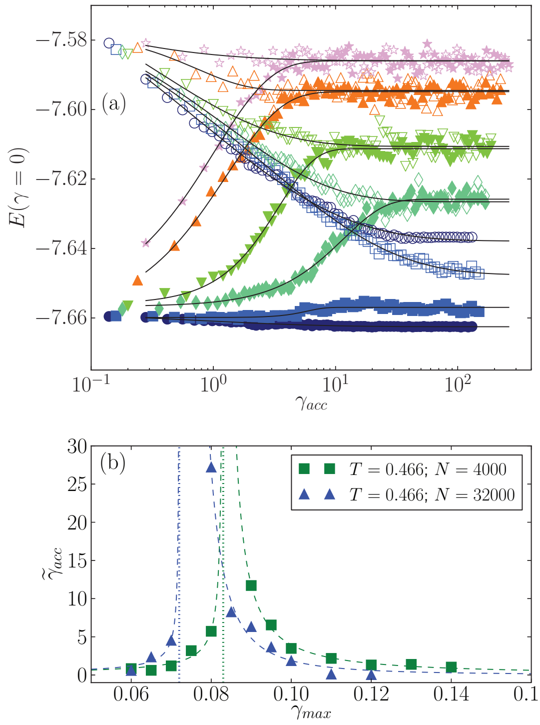

FIG. 1. (Color online) (a) Potential energy per particle E for zero-strain configurations, for different values of $\gamma_{\max}$ [0.07 (◯), 0.08 (□), 0.09 (◇), 0.1 (▽), 0.12 (△), and 0.14 (⋆)], averaged over different runs with samples of $N = 4000$ particles at $T = 0.466$ (closed symbols) and 1.0 (open symbols). The fits are obtained using a stretched exponential model. For the highest values of $\gamma_{\max}$, E fluctuates around a plateau after an initial transient, whose value increases with $\gamma_{\max}$ and is not dependent on the starting energy (curves with closed and open symbols of the same color and symbol merge), whereas for lower $\gamma_{\max}$ the memory of initial conditions is retained. For intermediate values our simulations are not long enough to determine which of the two behaviors eventually holds. (b) Values of characteristic strain $\tilde{\gamma}_{\text{acc}}$ (akin to relaxation time) obtained from stretched exponential fits of the energy relaxation like those in (a). The relaxation strain increases as one approaches the transition between the arrested and diffusive regimes. The lines through the data are guides to the eye. Vertical lines indicate critical strain amplitudes γ_c described in Fig. 3(a).

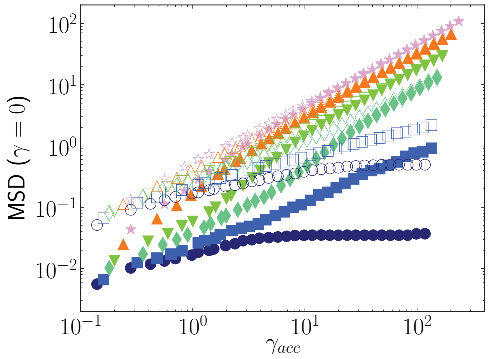

FIG. 2. (Color online) Mean square displacement for configurations, for different $\gamma_{\max}$, averaged over different runs with $N = 4000$ particles at $T = 0.466, 1.0$. Symbols are the same as in Fig. 1(a). Note the transition between an arrested and diffusive regime as $\gamma_{\max}$ is increased.

such a point γ_{acc}^* the MSD is given by

$$\mathrm{MSD} = D\gamma_{\mathrm{acc}}^*. \tag{1}$$

At intermediate values of $\gamma_{\max}$, the system reaches neither an immobile nor a diffusive steady state within the number of cycles we perform. The energy in Fig. 1(a) and the MSD data in Fig. 2, taken together, suggest that, in this regime, the system undergoes a transition from arrested (or quiescent) to diffusive behavior across a critical $\gamma_{\max}$, near which the relaxation of the system becomes very sluggish.

In Fig. 3(a) the values of D obtained for all the system sizes are plotted against $\gamma_{\max}$. For low $\gamma_{\max}$, D is zero and starts to increase rapidly around a critical value γ_c, which depends on the system size [Fig. 3(a)]. The values of D for $\gamma_{\max} > \gamma_c$ in Fig. 3(a) are reasonably well described by a law of the form (as in [16])

$$D = A(\gamma_{\max} - \gamma_c)^{\beta}. \tag{2}$$

The relevant fit values are mentioned in the caption of Fig. 3(a). Although better analysis is needed to be sure of the values of the exponent β and critical strain γ_c, the data shown are clearly consistent with a transition from a regime with zero diffusivity to one with finite diffusivity. Systems move diffusively in the large-strain regime whereas, in the low-strain regime, they become localized in configuration space. To test the robustness of the observation of a finite γ_c, we plot γ_c against different inverse system sizes $1/N$ in the inset of Fig. 3(a). This plot indicates that γ_c will be finite for $N \to \infty$.

Interestingly, the transition to the diffusive regime also coincides with the onset of energy dissipation in the system, as can be deduced by considering the stress-strain curves in the steady state, which are shown in the inset of Fig. 3(b). For small $\gamma_{\max}$ these curves do not enclose any significant area, indicating nearly elastic behavior (although involving small-scale plastic events even for small strain [3]). For large strain, however, the curves begin to clearly show hysteresis. As shown in Fig. 3(b), the $\gamma_{\max}$ values beyond which dissipation becomes finite agree with the critical strain γ_c for the corresponding system sizes. Therefore, the transition from a localized to a diffusive regime corresponds also to a transition from an almost nondissipative to a dissipative regime. It is interesting to ask if the critical strain is related to the yield strain γ_y in steady shearing conditions [we identify γ_y as the strain corresponding to the maximum stress obtained with our AQS procedure, as in the inset of Fig. 3(b); for thermal systems this value will be dependent on the strain rate, but in AQS deformation γ_y depends on the initial configuration only]. As shown in the inset of Fig. 3(a), the yield strain is close

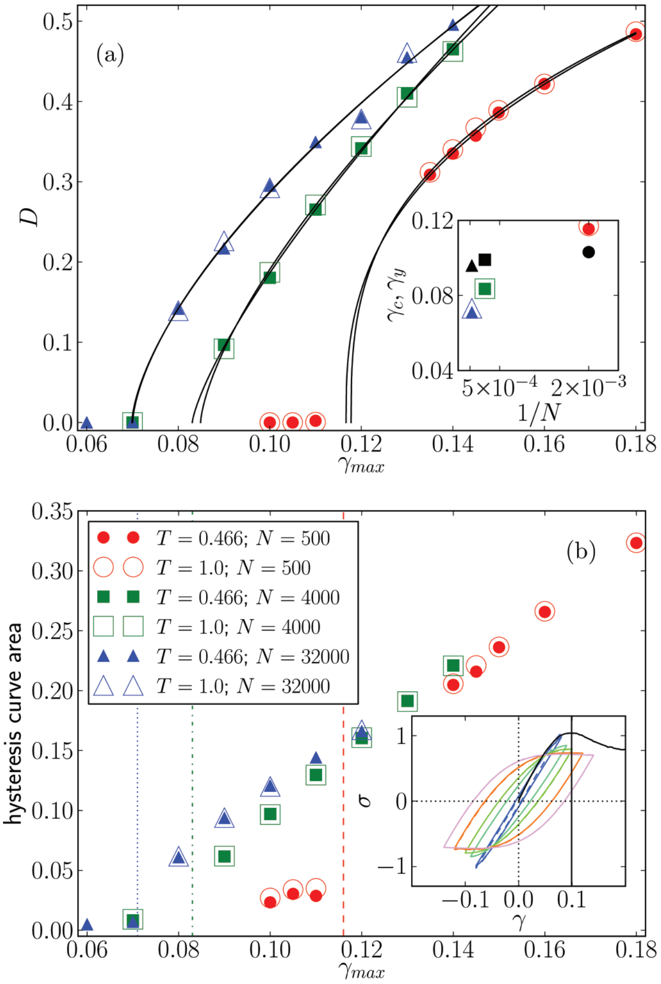

FIG. 3. (Color online) (a) Values of diffusivity D obtained by fitting the MSD, for the studied system sizes and temperatures. Superimposed on the data are power law fits [Eq. (2)] from which the critical strain γ_c values obtained [for $N = 500, 4000, 3200$, respectively, for ($T = 0.466$, $T = 1.0$)] are (0.115,0.117), (0.082,0.084), and (0.070,0.073). The corresponding exponent values β are (0.38,0.36), (0.75,0.76), and (0.61,0.54). The inset shows γ_c vs inverse system size $1/N$. Here γ_c extrapolates to a nonzero value as $1/N \to 0$. Also shown are the yield strain values γ_y (black closed symbols) obtained from stress-strain curves at $T = 0.466$ as shown in the inset in (b). (b) Stress-strain curves (shown in the inset for $N = 4000$) show hysteresis with finite enclosed area that increases with $\gamma_{\max}$ at high strain amplitude $\gamma_{\max}$. The stress-strain curve obtained under uniform shear is shown for reference, with the vertical line at the peak marking the yield strain γ_y. The areas enclosed by the hysteresis curves are shown in the main panel as a function of $\gamma_{\max}$ for all the different system sizes and temperatures studied. The areas, close to zero at low $\gamma_{\max}$, become finite above γ_c (indicated by vertical lines for each system size).

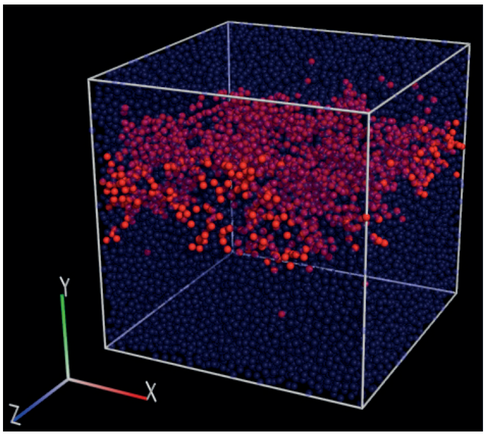

FIG. 4. (Color online) Snapshot of a $N = 32\,000$ configuration obtained in the steady state for $\gamma_{\max} = 0.08$. Particles in red are those that have undergone a scalar displacement greater than 0.6σ when the sample is subject to a full shear deformation cycle on the xy plane. The presence of a band is evident.

to but slightly higher than the critical strain (consistently with [17]), suggesting that the onset of irreversible events leading to critical behavior occurs at strain values smaller than the yield strain itself. Although our results for different system sizes show qualitatively consistent behavior, at the largest size studied, $N = 32\,000$, a new phenomenon is observed: Systems under strain display clear indications of shear banding, with particles that move the most in a single cycle clearly correlated in position (see Fig. 4). Shear bands are present in all the samples above the critical strain. However, despite the emergence of shear banding, the dynamical transition scenario discussed so far is not affected.

To summarize, we have shown that athermal oscillatory shear deformation drives, for large enough strains, dense samples of a model glass into steady states in which they explore their energy landscape in a manner dictated by the deformation amplitude $\gamma_{\max}$. Previous observations of overaging and rejuvenation via a single cycle of deformation in a model glass [14] are thus rationalized as initial steps towards a steady state that is independent of the initial state of the samples and depending only on $\gamma_{\max}$. Depending on whether the amplitude of the deformation is below or above a critical strain amplitude γ_c, these driven amorphous solids can be in an arrested (or localized) state or in a diffusive state. The relaxation to the steady state becomes very sluggish for $\gamma_{\max}$ values near the critical value. The transition from a localized to a diffusive steady state is very reminiscent of that observed in experiments on driven systems [16–18,25] and reported recently by other authors [26–28] on systems similar to ours. We emphasize that the similarity with the low-density colloidal suspension [16] is particularly remarkable given the differences in the microscopic origin of the reversibility for small strain amplitudes. The value of the amplitude γ_c at which this transition occurs depends on the system size but appears to remain finite in the limit of infinitely large systems. We find that the value of γ_c is in excellent agreement with the strain value at which hysteresis develops in the stress-strain curves, indicating a transition from a quasielastic to an elastoplastic regime. The critical strain values are close to but slightly lower than the yield strain. Shear banding occurs for our largest system sizes ($N = 32\,000$) in diffusing samples. Interestingly enough, the onset of shear banding at large N does not fundamentally affect the picture outlined above: Oscillatory strain, depending on the strain amplitude, is able to make amorphous solids trace almost nondissipative, localized orbits in their configuration space, or diffusive, dissipative trajectories, and such nonequilibrium transition occurs at a value of the strain amplitude γ_c that is close to but does not coincide with the yield strain.

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