Exploring the complex free energy landscape of the simplest glass by rheology

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For amorphous solids, it has been intensely debated whether the traditional view on solids, in terms of the ground state and harmonic low energy excitations on top of it, such as phonons, is still valid. Recent theoretical developments of amorphous solids revealed the possibility of unexpectedly complex free energy landscapes where the simple harmonic picture breaks down. Here we demonstrate that standard rheological techniques can be used as powerful tools to examine non-trivial consequences of such complex free energy landscapes. By extensive numerical simulations on a hard sphere glass under quasi-static shear at finite temperatures, we show that, above the so-called Gardner transition density, the elasticity breaks down, the stress relaxation exhibits slow and aging dynamics, and the apparent shear modulus becomes protocol-dependent. Being designed to be reproducible in laboratories, our approach may trigger explorations of the complex free energy landscapes of a large variety of amorphous materials.

INTRODUCTION

Amorphous and crystalline solids have very different behaviors under external perturbations, especially rheological properties under shear deformations [1–18]. It is well known that by increasing the shear strain, a crystal displays a linear elastic response, followed by plastic deformation and yielding. However, experiments and numerical simulations show that this picture breaks down for amorphous solids, such as glasses [2, 5–8, 13, 16, 18, 19], granular matter [3, 11, 15], and foams [4], where the elastic behavior is mixed with plastic events. Such plastic events cause sudden drops in stress-strain curves, and are sometimes referred to as crackling noise [20], due to their similarities to avalanches in earthquakes. An apparent shear modulus or rigidity $\mu$, which is the ratio between the stress and strain, can be nevertheless defined and measured. Experiments on glassy emulsion systems [21, 22] show that $\mu$ scales linearly $\mu \sim P$ with the pressure $P$ both below and above the jamming density, while harmonic treatments predict $\mu \sim P^{1.5}$ (below) [23] and $\mu \sim P^{0.5}$ (above) [24] respectively. These contradictions reveal that amorphous solids can be strikingly softer than purely harmonic solids like crystals, even at sufficiently low temperatures where the harmonic expansion was conventionally expected to be valid.

On the theoretical side, the mean-field theory based on the exact solution in the large dimensional limit of the hard sphere glass has brought a more accurate and comprehensive picture beyond the harmonic description [10, 14, 25–30]. The main outcome is the prediction of a Gardner transition (see Fig. 1a), which divides the classical amorphous phase into two: in the stable phase (or normal phase), the state is confined in one of the simple smooth basins on the free energy landscape; once the system is compressed above the Gardner transition density $\varphi_G$ (or is cooled down below the Gardner transition temperature $T_G$), the simple glass basin splits into a fractal hierarchy of sub-basins and the glass state becomes marginally stable. Although similar ideas of complex energy landscapes have been conceived phenomenologically in earlier works [see 31 and references there in], the mean-field theory gives a firmer first principle ground for such a picture, with falsifiable predictions. In particular, the theory predicts that the elastic anomalies and non-trivial rheology should only appear in the marginally stable phase (or Gardner phase) [10, 14], which lies deep inside the glassy phase. However, the mean-field theory is exact only in the large dimensional limit, and its relevance in real systems is far from obvious. Here we test the theoretical proposal of the non-trivial rheology in physically relevant dimensions $d = 2$ and $3$, and compare quantitatively the theoretical predictions with our numerical data.

We design laboratory reproducible rheological protocols to examine the signatures of the intriguing complex free-energy landscape. Our protocols are applied on densely packed hard spheres, which is a simple and representative glass-forming model. Our result shows the anticipated anomalous rheology emerging at the Gardner transition, which turns out to be strikingly similar to the dynamical responses of spin glasses to an external magnetic field [32, 33]. The evidence of a complex free energy landscape in the Gardner phase is consistent with a previous numerical

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Preparation of stable glasses. To avoid crystallization, we work on a polydisperse mixture of hard spheres whose diameters are distributed according to a probability distribution \( P(D) \sim D^{-3} \), for \( D_{\text{min}} \leq D < D_{\text{min}}/0.45 \) [34, 36] (see Supplementary Note 1). A glass is typically obtained by a slow compression (or cooling) annealing from a dilute state, where it falls out of equilibrium at the compression (or cooling) rate-dependent glass transition density \( \varphi_g \) (or glass transition temperature \( T_g \)). Since we choose hard spheres as our working system, the density is the control parameter.

We design a numerical protocol to mimic a simple shear experiment of deeply annealed glasses (see Fig. 1). Our protocol includes three steps: We first use the swap algorithm [34, 36] to prepare a well-equilibrated, supercooled-liquid configuration at various densities \( \varphi_g \) (see Supplementary Methods and Supplementary Figure 1). The algorithm combines the Lubachevsky-Stillinger algorithm [37], which consists of standard event-driven molecular dynamics (MD) and slow compression, with Monte-Carlo swaps of particle diameters. The MD time is expressed in units of \( \sqrt{3mD^2} \), where the particle mass \( m \) and mean diameter \( D \), as well as the inverse temperature \( \beta \), are all set to unity. In other words, a particle travels over a distance of the order of the diameter within a unit MD time. From the thermodynamic point of view, the system is still in the liquid but we work at density \( \varphi_g \) sufficiently above the mode-coupling theory (MCT) crossover density \( \varphi_d \). Then once we switch off the particle swapping and return to the natural dynamics simulated by MD, the \( \alpha \)-relaxation time has become much larger than our MD simulation time scales so that the system behaves essentially as a solid. This glass is thus ultrastable, in a sense similar to those obtained by vapour deposition experiments [38–40]. At a given density \( \varphi_g \), we prepare many of such equilibrated configurations, which are statistically independent from each other, and we call them samples in the following.

Second, subsequently the equilibrated configuration is compressed up to a target density \( \varphi \) with a compression rate \( \dot{\varphi}_g = 10^{-3} \). From a single sample, that is a starting equilibrated configuration at \( \varphi_g \), we generate an ensemble of compressed glasses at \( \varphi \), obtained by choosing statistically independent initial particle velocities drawn from the Maxwell-Boltzmann distribution. We call each of such compressed glasses as a realization in the following. These realizations are out-of-equilibrium, since they no longer follow the liquid equation of state (EOS), but we consider that they remain in restricted equilibrium [29] for \( \varphi < \varphi_G \), i.e., they are equilibrated within the given glass state determined by the sample. The MD preserves the kinetic energy so that the system remains at the unit temperature throughout our simulations. The typical scale of the vibrations of the particles within the glass states depends on \( \varphi \). For instance it varies from \( 10^{-1} \) to \( 10^{-2} \) for \( \varphi = 0.645 \) to \( \varphi = 0.688 \) (for \( \varphi_g = 0.643 \), see Fig. 2 of [34]), so that particles make 10 to \( 10^2 \) collisions within a unit MD time.

Third, for a given realization, a simple shear is applied. The simple shear is modelled by an affine deformation of the \( x \)-coordinates of all particles, \( x_i \rightarrow x_i + \gamma z_i \), under the Lees-Edwards boundary condition [41] with fixed system volume. The shear strain is increased quasi-statically with a small constant shear rate \( \dot{\gamma} = 10^{-4} \), such that the shear rate dependence is negligible in the regime \( \varphi < \varphi_G \) (see Supplementary Figure 6 for a discussion on the \( \dot{\gamma} \)-dependence), and the shear stress \( \Sigma \) is measured at different \( \gamma \). The shear stress \( \Sigma \) and the pressure \( P \) are both calculated from inter-particle interactions due to collisions between hard sphere particles. For convenience, we introduce reduced pressure \( p = \beta P/\rho \) and reduced stress \( \sigma = \beta \Sigma/\rho \), where \( \rho \) is the number density of the particles (see Supplementary Note 1). Note that as the pressure, the shear stress is entirely due to momentum exchanges between the particles so that the rigidity is purely entropic in hard sphere systems. Furthermore, because shear stress and pressure has the same physical dimension, it is convenient to introduce a rescaled stress \( \tilde{\sigma} = \sigma/p \).

Breakdown of elasticity. Figure 1 shows the phase diagram for our polydisperse hard sphere model, and typical stress-strain curves of individual realizations in different density regimes. In the stable glass phase \( \varphi_g < \varphi < \varphi_G \) (Fig. 1b), the stress-strain curve shows a smooth linear (harmonic) response regime at small \( \gamma \), followed by a sharp drop of the stress \( \sigma \) signalling the yielding of the system. At yielding, a system-wide shear band emerges (see Fig. 1c), and the system is driven out of a free energy metastable glass basin. After yielding, the system enters a steady flow state, similar to those observed in athermal amorphous solids under quasi-static shear [6, 42]. In the Gardner phase \( \varphi_G < \varphi < \varphi_J \), where \( \varphi_J \) is the jamming density, the harmonic response is punctuated by mesoscopic plastic events.
(MPEs) that can happen at very small γ (see Fig. 1d). These MPEs correspond to sudden avalanche-like heterogeneous rearrangements of particle positions without formation of band-like patterns (see Fig. 1e). Similar MPEs have been observed in quasi-static shear simulations at zero temperatures [2, 6], but our simulations are performed at finite temperatures. Note that the details of the plastic events, including the locations of yielding, jamming, and MPEs, depend on the samples (see Supplementary Figure 8) and realizations (see Supplementary Figure 7). For the behavior of the stress-strain curves averaged over many realizations and samples, see Supplementary Figures 2, 3, and 4, as well as Supplementary Notes 2 and 3.

For large ϕ, the stress σ grows dramatically at large γ, and appears to diverge (see Fig. 1d). This shear jamming phenomenon is due to the dilatancy effect of hard sphere glasses under shear: the pressure p increases with γ when the system volume is fixed. Note that if p is kept as a constant when γ is increased, then the volume expands due to the dilatancy effect. In that case, shear jamming does not appear and shear yielding is recovered (see Supplementary Figure 5). While the switching from shear yielding to shear jamming with increasing ϕ is not a consequence of the Gardner transition, it implies that the system is trapped more deeply in the metastable basin, and that the activated barrier-crossing between metastable basins becomes forbidden. However, the emergence of sub-basins in the Gardner phase implies that even though the usual relaxation (α-relaxation) is frozen, an additional slow dynamics may appear. This aspect is explored below.

**Aging and slow dynamics.** We next show that in the Gardner phase, the relaxation of shear stress becomes complicated, accompanied by aging and a slow dynamics. Due to the similarity between the Gardner transition and the spin glass transition, here it is very useful to firstly recall what happens in spin glasses, which are essentially disordered and highly frustrated magnets [43, 44]. The mean-field spin glass theory has suggested complex free energy landscapes of spin glasses manifested as continuous replica symmetry breaking [45], much as what happens in the Gardner phase of hard sphere glasses [27, 28]. Remarkably, this feature is predicted to have a reflection in the dynamics, resulting in non-trivial dynamical responses to external magnetic field, and aging effects in the relaxation of magnetization [46–48]. In experiments, the simplest approach to examine the intriguing features of the dynamics is a combination of the so called zero-field cooling (zfc) and field cooling (fc) protocols. In the zfc protocol, one cools a spin glass sample from a high temperature in the paramagnetic phase down to a target temperature T, where a magnetic field h is switched on and one measures the increase of the magnetization. In the fc protocol, one first switches on the magnetic field h, and then subsequently cools the system down to the target temperature T and measures the remanent magnetization. The key point is that, in the two protocols, the order of cooling and switching on of the magnetic field is reversed. In such experiments [49, 50], the magnetizations observed in the zfc/fc protocols are the same if the working temperature T is higher than the spin glass transition temperature, while the fc magnetization becomes larger than the zfc magnetization if T is lower than the spin glass transition temperature. The anomaly, i.e., the difference between the zfc and fc magnetizations, is naturally explained by the mean-field theory [45]. Furthermore, examinations of the aging effects by these protocols give detailed information about the complex free energy landscape [32, 33, 46–48].

It has been pointed out theoretically that the shear on structural glasses plays a very similar role as the magnetic field on spin glasses [9, 51], and that the relaxation of the shear stress should also reflect the complex free energy landscape encoded by the continuous replica symmetry breaking solution in the Gardner phase (see Supplementary Figure 9, and Fig. 2 of [10]). The shear strain and stress in structural glasses correspond to the magnetic field and magnetization in spin glasses respectively. Furthermore, apparently compression corresponds to cooling in the hard sphere glasses. Therefore, inspired by the zfc/fc experiments in spin glasses, we design two distinct protocols which are combinations of compression and shear exerted in reversed orders: In the zero-field compression (ZFC) protocol, we first compress the configuration from φG to φ, and set the time to zero. Then we wait for time τw before a shear strain γ is applied instantaneously (see Supplementary Methods), and measure the relaxation of the stress σZFC(t, τw) as a function of the time τ = t − τw elapsed after switching on the strain. On the other hand, in the field compression (FC) protocol, we first apply an instantaneous increment of shear strain at the initial density φG, compress the configuration to φ, and set the time to zero. Then we measure the relaxation of the stress σFC(t) as the function of the elapsed time t.

For ϕ < ϕG, no aging effect is observed, and the dynamics is fast. The σZFC(t, τw) is stationary or time translationally invariant (TTI), i.e. σZFC(τ, τw) = σZFC(τ), depending only on the time difference τ = t − τw but not on the waiting time τw (see Fig. 2). After a time scale τG corresponding to the ballistic motions of particles [34], the ZFC stress σZFC(τ, τw) converges quickly to σFC(t) which is almost a constant in time.

In contrast, for ϕ > ϕG, σZFC(τ, τw) displays strong τw-dependent aging effects manifesting the out-of-equilibrium nature of the system, as well as a slow dynamics. In such a situation, different large time limits can emerge depending on the order of τ → ∞ and τw → ∞ [52]. An important feature which can be seen in Fig. 2 is that σZFC(τ, τw) exhibits a plateau suggesting the existence of a large time limit σZFC = limτ → ∞ limτw → ∞ σZFC(τ, τw) where τw → ∞ is taken before τ → ∞. On the other hand σFC(t) is again essentially constant in time t (for t > τG) and we shall denote it as σFC. In the reversed order of the large time limits, we expect that the ZFC shear stress decays to the FC one,
\[ \lim_{t_\infty \to \infty} \lim_{\tau \to \infty} \sigma_\text{ZFC}(\tau, t_\infty) = \sigma_\text{FC}. \] However, the convergence becomes slower as \( t_\infty \) increases, and its corresponding time scale could be beyond the simulation time window, as shown in the case of Fig. 2.

Apparently \( \sigma_\text{ZFC} \) is larger than \( \sigma_\text{FC} \) when \( \varphi > \varphi_G \), which implies the ergodicity breaking. The aging effect and the slowing down of dynamics show the similarities between the Gardner transition and the liquid-glass transition, which demonstrates that the Gardner transition could be considered as a “glass transition within the glass phase” (see also Supplementary Note 3). In a sharp contrast, because the Gardner transition is absent in a crystal, its shear stress relaxes faster when \( \varphi \) increases, and no aging is present.

**Protocol-dependent shear modulus.** The above observation suggests that the linear shear moduli measured by the two protocols should be distinct in the Gardner phase. We determine the apparent shear modulus \( \mu \) as \( \mu_\text{ZFC} = (\sigma_\text{ZFC} - \sigma_0)/\gamma \) and \( \mu_\text{FC} = (\sigma_\text{FC} - \sigma_0)/\gamma \), where \( \sigma_0 \) is the remanent shear stress at \( \varphi \) before \( \gamma \) is applied. The shear strain is increased quasi-statically with rate \( \dot{\gamma} = 10^{-4} \) up to a predetermined small target \( \gamma \). The shear stress is measured at \( \tau = 1 \) after waiting for \( t_\infty = 10 \). Details on the time and \( \varphi_0 \) dependences of the shear modulus is discussed in Supplementary Figures 10, 13, and 14.

Figure 3 shows that, while \( \mu_\text{ZFC} \) and \( \mu_\text{FC} \) are indistinguishable in the stable glass phase \( \varphi < \varphi_G \) (or \( p < p_G \)), they become clearly distinct in the Gardner phase \( \varphi > \varphi_G \) (or \( p > p_G \)). For a similar result of a two-dimensional bidisperse hard disk model, see Supplementary Figure 16. This behavior of shear modulus is a consequence of the time dynamics of the shear stress illustrated in Fig. 2: at the time scales used to measure the shear modulus (\( \tau = 1 \) and \( t_\infty = 10 \)), the two shear stresses \( \sigma_\text{ZFC} \) and \( \sigma_\text{FC} \) have converged to the same value for \( \varphi < \varphi_G \), but remain different for \( \varphi > \varphi_G \). The bifurcation point determines the Gardner transition threshold \( \varphi_G \) (or \( p_G \)). Within the numerical accuracy, the \( \varphi_G \) determined from this approach is fully consistent with the previous estimate based on particles’ vibrational motions and caging order parameters [34]. To further test this result, we perform detailed analysis on its dependence on the number of particles \( N \) and the shear strain \( \gamma \), as discussed below.

We find no appreciable finite size effects for \( \mu_\text{FC} \) (see Fig. 3a), which is in contrast to the observation in non-equilibrated systems, where \( \mu_\text{FC} \) decreases to zero in the thermodynamic limit [13]. It suggests that preparing deeply equilibrium configurations is the key to observe the non-vanishing \( \mu_\text{FC} \). While the shear moduli measured around the Gardner transition, and therefore the determination of \( \varphi_G \), are \( N \)-independent, stronger finite size effects are observed for \( \mu_\text{ZFC} \) at large \( p \) near the jamming limit: \( \mu_\text{ZFC} \) is lower in larger systems, suggesting a stronger non-linear effect. Nevertheless, the data of \( \mu_\text{ZFC}(p) \), with a fixed \( \gamma \), appear to converge for \( N \gtrsim 2000 \), which confirms that \( \mu_\text{ZFC}(p) \) and \( \mu_\text{FC}(p) \) remain distinguishable in the thermodynamic limit, for \( \varphi > \varphi_G \).

Regarding the \( \gamma \)-dependence, Fig. 3b shows that, within the numerical accuracy, \( \mu_\text{FC} \) is independent of \( \gamma \), as long as \( \gamma \) is sufficiently small. On the other hand, for \( \varphi > \varphi_G \) and a given \( N \), \( \mu_\text{ZFC} \) slightly increases with decreasing \( \gamma \). This result shows that in the Gardner phase, the non-linear effect on \( \mu_\text{ZFC} \) remains even for very small \( \gamma \), which is consistent with the observation of elasticity breakdown in Fig. 1. Such non-linear effects are observed for any \( N \) studied (see Supplementary Figure 12), and we expect that in the thermodynamic limit \( N \to \infty \), a pure linear behavior of \( \mu_\text{ZFC} \) can only exist in the limit \( \gamma \to 0 \) [53]. The vanishing of the pure elastic regime distinguishes the Gardner phase from the normal glass and crystalline phases. For a more detailed discussion on how the shear moduli depend on the strain \( \gamma \), the particle number \( N \), the initial density \( \varphi_0 \), and the waiting time \( t_\infty \), see Supplementary Note 4.

For \( \varphi > \varphi_G \), the mean-field theory predicts two power-law scalings in the large \( p \) limit [10]: \( \mu_\text{ZFC} \sim p^\kappa \) with \( \kappa = 1.41574, \ldots \), and \( \mu_\text{FC} \sim p \). The first scaling has also been derived semi-empirically by an independent approach [54]. We find good agreement between the theory and simulation on the scaling of \( \mu_\text{FC} \) (see Fig. 3). For \( \mu_\text{ZFC} \), a noticeable discrepancy is observed in the limit of large \( N \) for a fixed finite \( \gamma \) (Fig. 3a), but the discrepancy decreases when \( \gamma \to 0 \) for a fixed \( N \) (Fig. 3b), or when \( N \) is decreased for a fixed \( \gamma \) (Fig. 3a). This is because the mean-field \( \mu_\text{ZFC} \) is obtained in the pure linear response limit \( \gamma \to 0 \), while the non-linear effect caused by MPEs would increase with \( \gamma \) and \( N \), as discussed above. The scaling \( \mu_\text{FC} \sim p \) is consistent with the experimental observation in emulsions [21, 22]. Considering the experimental system is possibly not deeply equilibrated, we expect that the relaxation of experimental \( \mu(t) \) is sufficiently fast, and the measurement was performed in the long time limit \( \mu(t \to \infty) \to \mu_\text{FC} \) (see the discussion of Fig. 2).

**Interpretation of results.** The Gardner transition is a consequence of the split of glass basins in the phase space [28], and the split of particle cages in the real space (see Fig. 4). The schematic plot of the free energy \( F \) as a function of \( \gamma \) in Fig. 4 illustrates how a glass basin splits into many sub-basins once the system is compressed above \( \varphi_G \). Here we interpret our results based on this free energy landscape viewpoint. First, in the ZFC protocol, the system intends to remain in one of the sub-basins after compression (note that different realizations may end up in different sub-basins), but as \( \gamma \) increases in a quasi-static shear procedure (Fig. 1), it may become unstable where the shear stress drops abruptly, resulting in a MPE. The MPE could be interpreted as shear-induced barrier crossing between sub-basins, analogous to the barrier crossing between basins in a yielding event. Second, if \( \gamma \) is fixed, the shear stress relaxes with time, and according to the Arrhenius law, the emergence of barriers between sub-basins would result in a slowing down of the relaxation dynamics with \( \varphi \) (Fig. 2). The appearance of aging further reveals the emergence of complex structures within a basin, similar to the mechanism of aging in the glass transition [52].
Third, because in the FC protocol, the system can overcome the sub-basin barriers, the $\mu_{\text{FC}}$ always corresponds to the second order curvature of basins, rather than that of sub-basins as in the $\mu_{\text{ZFC}}$ case. This results in $\mu_{\text{FC}} < \mu_{\text{ZFC}}$ for the regime $\varphi > \varphi_G$ as observed in Fig. 3. Note that according to Fig. 4e, one should obtain a shear modulus close to $\mu_{\text{ZFC}}$ if an additional strain is applied after FC, as confirmed in Supplementary Figure 15. On the other hand, no basin split occurs and therefore two protocols are equivalent in the stable phase $\varphi < \varphi_G$. Previous study [13] has shown that $\mu_{\text{ZFC}} = \mu_{\text{FC}}$ for crystals. The similarity between crystals and stable glasses further confirms that their free energy basins are similarly structureless.

**DISCUSSION**

We wish to stress that, our data cannot exclude that the Gardner transition becomes just a crossover in finite dimensions, such that no real phase transition exists. Yet, irrespective of the sharpness of the Gardner transition, we rationalize here in a unified framework all the observations obtained on the rheological behavior of the simple hard sphere glass, and find quantitatively reasonable agreement between the theory and simulations. Thus, even if the Gardner transition is not sharp in the thermodynamic limit, for accessible sizes in numerical simulations, and likely for those in experiments as well [35], a behavior reminiscent of the transition can be clearly observed.

Finally, we make remarks on experimental consequences. It is an intriguing question to clarify whether the phase diagram presented in Fig. 1a is generic in a wide range of amorphous solids, ranging from different kinds of glasses to soft matter such as colloids (One can choose to change the temperature or pressure as the control parameter depending on specific systems). The crucial point is to keep track of the dynamical effects which might have been overlooked in some previous experiments, for the following two reasons. First, in reasonably stabilized dense systems, the liquid EOS (green line in Fig. 1a) and the Gardner line (red line) becomes separated enough, so that the liquid dynamics ($\alpha$-relaxation) and the intriguing internal glassy dynamics ($\beta$-relaxation induced by the Gardner transition) can be well separated in time scales. In this respect, recently developed experimental techniques, such as the vapor deposition [38–40] and the high pressure path [55], or the use of sufficiently old natural glasses [56], would provide ideal settings. If such an ideal setting is not possible, one could freeze the $\alpha$-relaxation out of the experimental time window, by working at sufficiently low temperatures or high densities. The second reason is that by experimentally studying the aging effects due to the internal dynamics of the amorphous solids, the complexity of the free energy landscape could become manifested as we demonstrated in the present paper.

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FIG. 1. Typical stress responses under quasi-static shear. (a) Illustration of the protocol on the polydisperse hard sphere glass phase diagram (adapted from Ref. [34]), where $k_B T / P = 1/\rho p$. The MCT dynamical crossover (yellow star) is located at $\varphi_d = 0.594(1)$ along the equilibrium liquid EOS (green line). Using the swap algorithm we first prepare equilibrium samples at various densities $\varphi_g$ (green squares) whose pressure obeys the Carnahan-Stirling empirical liquid EOS (solid green line) [34]. Next we switch off the swap algorithm, and perform compression annealing from $\varphi_g$ to jamming (blue triangles), producing realizations of compressed glasses at various densities $\varphi$. The system is now out-of-equilibrium and the pressure follows the glass EOSs $p \propto 1/ (\varphi_J - \varphi)$ (black dotted lines) [34]. The Gardner transition $\varphi_G$ (red circles and line) separates the stable (light yellow regime) and the marginally stable (light blue regime) glass phases. The insets show schematic depictions of free energy landscapes in these two different phases. As an example, an equilibrium configuration is prepared at $\varphi_g = 0.643$, and compressed (solid black line) up to $\varphi_J = 0.690(1)$. We show typical stress-strain curves under quasi-static shear with increasing $\gamma$, using a single realization of the compressed glass of $N = 1000$ particles, at (b) $\varphi = 0.670$ (pink cross) and (d) $\varphi = 0.688$ (pink plus), which are below and above $\varphi_G = 0.684(1)$ respectively. Curves in (b) and (d) are zoomed in (insets) for $\gamma \leq 0.025$, to show the different small-$\gamma$ behaviors in the two cases. The real-space vector fields of particle displacements are visualized in (c) for a yielding event (between the two red circles in (b)), and (e) for a mesoscopic plastic event (MPE) (between the two red circles in (d)), where each sphere is located at the equilibrium position before yielding/MPE, and each vector represents the displacement during yielding/MPE. We have subtracted the affine part caused by shear from the displacements, and only show top 20% particles with large displacements. A shear band around the middle of the $z$-axis is observed in (c). The sizes of particles are reduced by a factor of 0.4, and the vectors are amplified in length by a factor of 2 in (c) and a factor of 15 in (e). The color represents the magnitude of displacement.
FIG. 2. Relaxation of shear stress. Relaxations of the rescaled ZFC shear stress $\tilde{\sigma}_{ZFC} = \sigma_{ZFC}/p$ (filled symbols) and the rescaled FC shear stress $\tilde{\sigma}_{FC} = \sigma_{FC}/p$ (open symbols) show different behaviors at (a) $\phi = 0.670$ and (b) $\phi = 0.688$, corresponding to the pink plus and cross in Fig. 1 respectively (the Gardner transition density $\phi_G = 0.684(1)$ [34]). We show results for several different waiting time $t_w$, under an instantaneous increment of shear strain $\gamma = 10^{-3}$. Data are averaged over many realizations of compressed glasses obtained from a single equilibrated sample at $\phi_g = 0.643$ with $N = 1000$ particles. Here the rescaled remanent stress $\tilde{\sigma}_0$ is measured in the ZFC protocol at $\phi$, after the longest waiting time $t_w = 1000$ and before the shear strain is applied. The difference $\tilde{\sigma}_{ZFC}(\tau, t_w) - \tilde{\sigma}_{FC}(\tau, t_w)$ quickly vanishes and does not show significant $t_w$-dependence at (c) $\phi = 0.670$, while it decays much slower and shows a strong $t_w$-dependent aging effect at (d) $\phi = 0.688$. Note that by definition, $\tilde{\sigma}_{FC}(t)$ is a one variable function, but we plot it here as $\tilde{\sigma}_{FC}(\tau, t_w)$ in order to compare it with $\tilde{\sigma}_{ZFC}(\tau, t_w)$. The pressure $p$ is independent of time and protocol, in both cases (see Supplementary Figure 11). The error bars denote the standard error of the mean (s.e.m.).
FIG. 3. Protocol-dependent shear modulus. (a) The rescaled shear modulus \( \tilde{\mu} = \mu / p \), obtained from both ZFC (filled symbols) and FC (open symbols), is plotted as a function of \( p \), for \( \varphi_g = 0.643 \) and several \( N \). The data are obtained by using \( \gamma = 2 \times 10^{-3} \), and are averaged over \( N_s \approx 200 \) samples, and \( N_r \approx 100 \) individual realizations for each sample. The two shear moduli \( \mu_{ZFC} \) and \( \mu_{FC} \) coincide below \( p_G \) (vertical dashed line), and become distinct above, where \( p_G = 265 [34] \). The data are compared to the large \( p \) scalings predicted by the mean-field theory \( \mu_{ZFC} \sim p^{1.41574} \) and \( \mu_{FC} \sim p \) (black solid lines). The difference \( \mu_{ZFC} - \mu_{FC} \) is plotted as a function of \( \varphi \) in the inset, where the vertical dashed line represents \( \varphi_G = 0.684 [34] \). (b) Rescaled ZFC and FC shear moduli obtained from a few different \( \gamma \), for \( N = 1000 \) systems. The error bars denote the s.e.m.
FIG. 4. Illustration of protocols. We show the evolution of the free energy landscape and the state point \((\varphi, \gamma)\) under compression and shear. (a) In the ZFC protocol, the system is first compressed and then sheared, while the order is reversed in the FC protocol. (b) State point \((\varphi_g, 0)\): The schematic free energy \(F\) as a function of the strain \(\gamma\) at the initial density \(\varphi = \varphi_g\) before compression. We assume that the initial state point (black open circle) is located at the minimum of the parabola. To show an example of the real-space particle caging, we also plot three independent trajectories of the same tagged particle in the same two-dimensional sample (see Supplementary Note 5). (c) State point \((\varphi, 0)\): If the system is compressed first to \(\varphi\) (above the Gardner transition density \(\varphi_G\)), the free energy basin (red dashed line) splits into many sub-basins (blue line): the state point (blue solid circle) becomes trapped in one of the sub-basins. The dotted blue lines represent the metastable region of the sub-basins. The split of free energy basin corresponds to the split of cage in the real space (as an example, see the independent trajectories representing three split cages). (d) State point \((\varphi_g, \gamma)\): On the other hand, if the system is sheared first, the state point (red solid circle) is forced to climb up the parabola of the basin. (e) State point \((\varphi, \gamma)\): After both shear and compression, the state point can be located at different points in the same free energy landscape, depending on the order of the compression and shear. In the ZFC case, the state point (blue solid circle) is forced to climb up the sub-basin where it is trapped, while it can remain at lower energy state in the FC protocol (red solid circle). Because sub-basins are meta-stable (dotted blue line), MPEs occur with increasing \(\gamma\) in a quasi-static shear, and slow relaxation occurs for a fixed \(\gamma\) (green arrow). The shear stress \(\sigma\) is determined by \(\sigma \sim dF/d\gamma\) (right panel), and the shear modulus by \(\mu = d\sigma/d\gamma \sim d^2F/d\gamma^2\). The stress-strain curves show that for \(\varphi > \varphi_G\), \(\mu_{ZFC}\) (slope of blue line) is larger than \(\mu_{FC}\) (slope of dashed red line).
Supplementary Figure 1. Evolution of the reduced pressure $p$ under compression, for a system of $N = 2000$ particles. The data obtained from the swap algorithm agree with the Carnahan-Stirling empirical liquid EOS Eq. (Supplementary Equation 4) [34]. For comparison, we also plot data obtained from pure compression done by the LS algorithm without the swap for a few different compression rate $\delta_g$. The swap algorithm falls out of equilibrium at much higher $\varphi$, compared to the standard compressions.
Supplementary Figure 2. Quasi-static shear on equilibrium configurations prepared at a few different $\phi_g$. The systems consist of $N = 1000$ particles, and the data are averaged over $N_s = 100$ samples and $N_{th} = 10 - 20$ realizations for each sample. (a) The shear stress $\sigma$ and (b) the pressure $p$ are plotted as functions of the shear strain $\gamma$. At small $\gamma$, the stress-strain curve is fitted to a linear function $\sigma = \mu \gamma$ (lines), and the pressure-strain curve is fitted to a quadratic function $p = p_g + R \gamma^2$ (lines). The star marks the peak of the stress-strain curve, which represents the yielding point $(\gamma_y, \sigma_y)$, and the cross marks the peak $(\gamma_m, p_m)$ of the pressure-strain curve. The parameters $\mu$, $\gamma_y$, $\sigma_y$, $\gamma_m$, $p_m$, and $R$ are reported in Supplementary Figure 3 as functions of $\phi_g$. (c) The rescaled stress-strain curves and (d) the rescaled pressure-strain curves are compared to the mean-field theoretical predictions (black line) [29], for the equilibrium volume fraction $\phi_g = 2^d \phi_k / d = 7$, where the dimension $d = 3$. Here the solid line part is the stable 1-step replica symmetry breaking (1RSB) solution, and the dashed line part is the unstable 1RSB solution [29]. We have checked that the theoretical results are insensitive to $\phi_k$ on these rescaled plots.
Supplementary Figure 3. Elastic and yielding parameters of equilibrium configurations. We plot as functions of $\phi_g$, (a) the shear modulus $\mu$, (b) the dilatancy parameter $R$, (c) the yield strain $\gamma_y$ and the strain $\gamma_m$ at the maximum pressure in the pressure-strain curve, and (d) the yield stress $\sigma_y$ and the maximum pressure $p_m$.

Supplementary Figure 4. Quasi-static shear on out-of-equilibrium glass states. The states are compressed from $\phi_g = 0.643$ to target density $\varphi$ before the shear is applied. The systems consist of $N = 1000$ particles, and the data are averaged over $N_s = 100$ samples and $N_{th} = 10^{-30}$ realizations for each sample. (a) The shear stress $\sigma$ as a function of $\gamma$, for a few different $\varphi$ (from bottom to top, $\varphi = 0.645, 0.65, 0.66, 0.662, 0.665, 0.67, 0.675, 0.68, 0.683, 0.684, 0.685, 0.687, 0.688$), where $\phi_G = 0.684(1)$. The linear response regime is fitted to $\sigma = \mu \gamma$ (lines). (b) The shear modulus $\mu$ obtained from this fitting is compared to the ZFC shear modulus $\mu_{ZFC}$ (we use $\gamma = 2 \times 10^{-3}$, see the main text), both of which diverge approaching to the jamming limit $\phi_J = 0.690(1)$ (vertical dashed line). For all figures, the error bars denote the standard error of the mean.
Supplementary Figure 5. Quasi-static shear on the same glass state ($\phi_g = 0.643$, $\varphi = 0.688$, and $N = 1000$) under constant volume and constant pressure show different behaviors at large $\gamma$. We observe shear jamming in the constant volume simulation and shear yielding in the constant pressure simulation.

Supplementary Figure 6. The stress-strain curves of three different realizations of the compressed glass at (a) $\phi = 0.670$, and (b) $\varphi = 0.688$, obtained from the same equilibrated sample of $N = 1000$ particles at $\phi_g = 0.643$. They are driven by different strain rates $\dot{\gamma}$ as indicated by the legend.

Supplementary Figure 7. Stress-strain curves of three different realizations of the compressed glass at (a) $\phi = 0.670$, and (b) $\varphi = 0.688$, obtained from the same equilibrated sample of $N = 1000$ particles at $\phi_g = 0.643$. They are driven by the common strain rates $\dot{\gamma} = 5 \times 10^{-6}$ for $\phi = 0.670$, and $\dot{\gamma} = 10^{-4}$ for $\varphi = 0.688$. 
Supplementary Figure 8. Stress-strain curves on the compressed glasses at (a) \( \varphi = 0.670 \), and (b) \( \varphi = 0.688 \), obtained from three different equilibrated samples of \( N = 1000 \) particles at \( \varphi_g = 0.643 \). They are driven by the common strain rates \( \dot{\gamma} = 5 \times 10^{-6} \) for \( \varphi = 0.670 \), and \( \dot{\gamma} = 10^{-4} \) for \( \varphi = 0.688 \).

Supplementary Figure 9. Schematic illustration of the relaxation of ZFC shear stress \( \sigma_{ZFC}(\tau, t_w) \) after an instantaneous shear strain \( \gamma \) is applied, in the Gardner phase \( \varphi > \varphi_G \) [10]. The two shear moduli \( \mu_{ZFC} \) and \( \mu_{FC} \) correspond to the first and second plateaus respectively. Corresponding time scales for the black solid line are indicated. The dotted black line represents a shorter waiting time \( t_w \). See also Fig. 2 of [10].

Supplementary Figure 10. Relaxation of \( \tilde{\sigma}_{ZFC} = \sigma_{ZFC}/p \) (filled symbols) and \( \tilde{\sigma}_{FC} = \sigma_{FC}/p \) (open symbols) at (a) \( \varphi = 0.670 \) and (b) \( \varphi = 0.688 \), for a few different \( t_w \), under a quasi-static shear strain \( \gamma = 10^{-3} \). The system consists of \( N = 1000 \) particles, and is compressed from \( \varphi_g = 0.643 \). The data are obtained for one individual sample, but averaged over \( N_{th} \sim 1000 \) independent realizations of the compressed glass. The remanent stress \( \tilde{\sigma}_0 \) has been subtracted from \( \tilde{\sigma} \).
Supplementary Figure 11. Time evolution of the ZFC pressure $p_{ZFC}(\tau, t_w)$ and the FC pressure $p_{FC}(\tau, t_w)$ after an instantaneous increment of shear strain $\gamma = 10^{-3}$, at (a) $\varphi = 0.670$ and (b) $\varphi = 0.688$. For comparison, we also display the behaviour of the shear stress in the ZFC and FC protocols. Data are averaged over many realizations of compressed glasses obtained from a single equilibrated sample at $\varphi_g = 0.643$ with $N = 1000$ particles. The pressure values are rescaled by a factor of $1/100$. This plot shall be compared with Fig. 2 in the main text (only $t_w = 1000$ data are shown).

Supplementary Figure 12. $\gamma$-dependence on the ZFC and FC shear moduli. The data are obtained for (a) $N = 500$ and (b) $N = 2000$ particles, and are averaged over $N_s \approx 200$ samples and $N_t \approx 100$ individual realizations for each sample. The vertical dashed line represents the Gardner transition [34], and the solid lines are the mean-field predictions $\mu_{ZFC} \sim p^{1.41574}$ and $\mu_{FC} \sim p$ [10].

Supplementary Figure 13. $\varphi_g$-dependence on the ZFC and FC shear moduli. The data are obtained for $N = 1000$ particles, and are averaged over $N_s \approx 200$ samples and $N_t \approx 100$ individual realizations for each sample. To compute the shear modulus, $\gamma = 2 \times 10^{-3}$ is used. (a) The numerical data of the rescaled ZFC shear modulus $\hat{\mu}_{ZFC} = \mu_{ZFC}/p$ (filled symbols) and the rescaled FC shear modulus $\hat{\mu}_{FC} = p_{FC}/p$ (open symbols) with a few different $\varphi_g$, are compared to the mean-field theory (lines) with different $\varphi_g$, where $\varphi_g = 2^{d} \varphi_e/d$ with $d = 3$, following the convention used in Ref. [29]. Both numerical and theoretical results are rescaled by the reference values $\hat{\mu}_g = \mu_g/p_g$ and $p_g$ at $\varphi_g$. (b) The difference $\hat{\mu}_{ZFC} - \hat{\mu}_{FC}$ as a function of $\varphi$ for a few different $\varphi_g$, where the Gardner transitions $\varphi_G$ (values from Ref. [34]) are marked by vertical lines.
Supplementary Figure 14. The difference $\tilde{\mu}_{\text{ZFC}} - \tilde{\mu}_{\text{FC}}$ is plotted as a function of $\varphi$, for $\varphi_g = 0.643$ and $N = 1000$. The shear moduli are measured at $\tau = 1$ for a few different waiting time $t_w$. The data are obtained by using $\gamma = 2 \times 10^{-3}$, and are averaged over $N_s \approx 100$ samples and $N_r \approx 50$ independent realizations for each sample. The vertical dashed line represents $\varphi_G = 0.684$ [34].

Supplementary Figure 15. The reduced shear modulus obtained from the third protocol (FCS) is compared to $\tilde{\mu}_{\text{ZFC}}$ and $\tilde{\mu}_{\text{FC}}$, for $\varphi_g = 0.643$, $N = 1000$, and $\gamma = 2 \times 10^{-3}$. The data are averaged over $N_s \approx 200$ samples and $N_r \approx 100$ individual realizations for each sample.

Supplementary Figure 16. Protocol-dependent shear modulus of a bidisperse hard disk glass former, where the vertical dashed line marks the Gardner transition estimated independently in Ref. [34]. The data are obtained for $\varphi_g = 0.808$, and are averaged over $N_s \approx 100$ samples and $N_{1h} \approx 100$ realizations for each sample.
SUPPLEMENTARY NOTE 1 – MODEL AND OBSERVABLES

Polydisperse hard sphere system

We study an assembly of $N$ polydisperse HSs whose diameters are distributed according to a probability distribution $[34, 36],$

$$P(D) \sim D^{-3}, \quad D_{\text{min}} \leq D < D_{\text{min}}/0.45.$$  \hspace{1cm} \text{(Supplementary Equation 1)}

This distribution is chosen to optimize the swap algorithm so that denser equilibrium configurations can be obtained, while ensuring that crystallization is suppressed $[36].$ The control parameter is the number density $\rho = N/V$ or the volume fraction $\varphi = (\pi/6)\rho \int_{D_{\text{min}}/0.45}^{D_{\text{min}}} dDP(D)D^3,$ where $V$ is the volume of the system. The mode-coupling theory (MCT) dynamical crossover density is $\varphi_d = 0.594(1)$ $[34].$ The simulation time $t$ is expressed in units of $\sqrt{\beta mD^2},$ where the inverse temperature $\beta,$ the particle mass $m,$ and the mean particle diameter $D = \int_{D_{\text{min}}/0.45}^{D_{\text{min}}} dDP(D)D$ all set to unity.

Shear stress and pressure

For a HS system, the stress is entropic. The stress tensor is given by

$$\Sigma_{mn} = -\frac{1}{V} \sum_{i<j} r_{ij,m} f_{ij,n}$$  \hspace{1cm} \text{(Supplementary Equation 2)}

where $r_{ij,m}$ is the $m$-th component of the separation vector $r_{ij} = r_i - r_j$ between particles $i$ and $j,$ and $f_{ij,n}$ is the $n$-th component of the inter-particle force $f_{ij}.$ The force $f_{ij}$ is computed from the exchange rate of the momentum between $i$ and $j.$ In our shear protocols, we are interested in the $z$-$x$ element of the stress tensor (we omit the subscript),

$$\Sigma = -\frac{1}{V} \sum_{i<j} z_{ij} f_{ij,x}.$$  \hspace{1cm} \text{(Supplementary Equation 3)}

The pressure $P$ is the negative average of three diagonal elements of the stress tensor, i.e., $P = -(\Sigma_{xx} + \Sigma_{yy} + \Sigma_{zz})/3 = \frac{1}{3V} \sum_{i<j} r_{ij} \cdot f_{ij}.$ In this study, we report results in the units of reduced pressure $p = \beta P/\rho$ and reduced stress $\sigma = \beta \Sigma/\rho.$

SUPPLEMENTARY NOTE 2 – QUASI-STATIC SHEAR ON EQUILIBRIUM CONFIGURATIONS

First let us report data obtained by quasi-static shear on equilibrium configurations at a few different $\varphi_g$ (Supplementary Figure 2). As we noted in the main text, the system is in the liquid state in the thermodynamic sense (the Kauzmann density $\varphi_K,$ if any, is larger then $\varphi_g$), but the $\alpha$-relaxation time is much larger than our simulation time scales so that the system behaves as a solid. The stress-strain curve, averaged over many samples and realizations, shows a linear elastic regime at small $\gamma,$ followed by yielding. We define the location of the peak in the stress-strain curve as the yield strain $\gamma_y.$ Note that the definition of $\gamma_y$ is more ambiguous for the stress-strain curve of a single realization from a single sample (for example, see Fig. 1b in the main text). In this study, we do not attempt to precisely determine $\gamma_y$ for each single stress-strain curve. The shear modulus $\mu$ is determined by $\mu = \sigma/\gamma$ in the elastic regime. Both $\gamma_y$ and $\mu$ grow with $\varphi_g$ (Supplementary Figure 3). The yield strain $\gamma_y$ and the yield stress $\sigma_y$ (which is the stress at $\gamma_y$) appear to vanish continuously around $\varphi_g \approx \varphi_d.$ On the other hand, the shear modulus $\mu$ appears to remain finite at $\varphi_d,$ which implies a discontinuous jump of $\mu$ at $\varphi_d$ being consistent with the mean-field theory $[10].$ In the elastic regime, the dilatancy effect is observed: the pressure $p$ increases quadratically with $\gamma,$ i.e., $p = p_g + R\gamma^2,$ where $p_g$ is the pressure at $\varphi_g$ and $\gamma = 0,$ and $R$ is the dilatancy parameter. The onset of the peak in the pressure-strain curves lags behind the yielding, i.e. the peak of the stress-strain curves. We compare our numerical data to the mean-field theoretical prediction $[29],$ and find reasonable agreement on rescaled plots as shown in Supplementary Figure 2.
Next, let us present quasi-static shear data of out-of-equilibrium configurations. These configurations are obtained by compressing the equilibrium configurations from \( \varphi_g \) to a target density \( \varphi \), at a constant compression rate \( \delta_g = 10^{-3} \). Supplementary Figure 4a shows that at small \( \gamma \), the average stress-strain curve has a linear regime, which shrinks with increasing \( \varphi \). Note that the data presented here are obtained by averaging over many samples and realizations, while the data in Fig. 1 (main text) are for one single sample and one single realization. For \( \varphi > \varphi_G \), the apparent linear regime in Supplementary Figure 4a is not truly elastic, since it is averaged over many mesoscopic plastic events (MPEs) (Fig. 1). Thus the shear modulus defined as \( \mu = \sigma/\gamma \) in the linear regime is not only due to purely harmonic responses but also involves non-affine corrections due to the plastic events. With this point being clarified, we show that the shear modulus \( \mu \) obtained from fitting the data in the linear regime, is basically consistent with \( \mu_{ZFC} \) presented in the main text (Supplementary Figure 4b).

At larger \( \gamma \), we find that with increasing \( \varphi \), the shear yielding disappears and the shear jamming emerges (Supplementary Figure 4a), which can be also observed in Fig. 1. Note that the simulation is performed under the constant volume condition. If we instead fix the pressure and allow the volume to change, then the shear jamming does not appear and the shear yielding exists, even at large \( \varphi \) (Supplementary Figure 5). We also stress that the shift from the shear yielding to the shear jamming is not correlated to the Gardner transition. In fact, Supplementary Figure 5 shows that it is possible to observe both MPEs (at small \( \gamma \)) and yielding (at large \( \gamma \)) in the same stress-strain curve. The key difference is that, after a MPE, the system remains in the same basin although it escapes from the sub-basin, and therefore it still behaves like a solid, while after yielding, the system escapes from the basin and essentially behaves like a fluid.

We next discuss in detail how the measurements of stress-strain curves depend on factors such as the compression rate \( \delta_g \), the shear rate \( \dot{\gamma} \) (Supplementary Figure 6), the realization (Supplementary Figure 7), and the sample (Supplementary Figure 8). First of all, although these configurations are in principle out-of-equilibrium, they reach restricted equilibrium [29] for \( \varphi < \varphi_G \), i.e., they are nearly equilibrated within their glass basins. As shown in Ref. [34], neither structural relaxation nor aging can be observed within the simulation time scale. According to that, in this regime, the results presented here should be nearly unchanged if a slower compression rate is used. The situation is different for \( \varphi > \varphi_G \); because the time scale diverges in this regime, it is difficult to obtain even the restricted equilibrium configurations and the data would be \( \delta_g \)-dependent. Effectively, decreasing \( \delta_g \) is equivalent to increasing the waiting time \( t_w \). Since the \( \delta_g \)-dependence has been well studied in previous work [34], we do not repeat the analysis here. For other factors, in the regime \( \varphi < \varphi_G \), our results are independent of the shear rate (Supplementary Figure 6) and realizations (Supplementary Figure 7), although noticeable sample-to-sample variance (Supplementary Figure 8) is observed. In contrast, for \( \varphi > \varphi_G \), the stress-strain curve becomes realization-dependent. This observation is consistent with our basic expectation: the free energy landscape is complex in this regime, and the system could fall into different sub-basins after compression.

Relaxation of the shear stress: connection to the free-energy landscape

To better understand the relaxation of shear stress upon a instantaneous shear strain \( \gamma \), in particular the behavior of \( \sigma_{ZFC}(t, t_w) \) in the Gardner phase (Fig. 2b in the main text), for comparison we schematically plot \( \sigma_{ZFC}(\tau, t_w) \) anticipated from a theoretical point of view [10] (Supplementary Figure 9). The key feature is that, after an initial fast decay within the ballistic time scale \( \tau_b \), two steps of relaxations are expected: the \( \sigma_{ZFC}(\tau, t_w)/\gamma \) firstly relaxes to the plateau corresponding to \( \mu_{ZFC} \) at time \( \tau \sim \tau_b \), and then it further relaxes to the second plateau corresponding to \( \mu_{FC} \) at \( \tau \sim \tau_{mb} \). Here \( \tau_b \) and \( \tau_{mb} \) are the times needed to explore a single glass sub-basin and a glass meta-basin respectively. At even larger time \( \tau \sim \tau_\alpha \), the stress may eventually relaxes to zero due to \( \alpha \)-processes. In our study, the initial configurations at \( \varphi_g \) are deeply equilibrated, such that \( \tau_\alpha \) is far beyond our simulation time scale. Thus the \( \alpha \)-relaxation is irrelevant in our analysis. However, if the initial configurations are far away from equilibrium, then \( \tau_\alpha \) could be comparable to the simulation time scale. In that case, the last step of relaxation towards zero stress may be observed [13, 18]. On the other hand, the behavior of \( \sigma_{FC}(t) \) is much simpler. As a one parameter function, it is \( t_w \)-independent by definition, and \( \sigma_{FC}(t)/\gamma \) should converge quickly to \( \mu_{FC} \) after the initial ballistic processes.
Relaxation of the shear stress: the case of slow switching on of the shear strain

In the main text we discussed the relaxation of the stress after instantaneous shear. Here let us examine how the shear stress relaxes if a small shear strain \( \gamma \) is applied quasi-statically (Supplementary Figure 10). The data should be compared with those in Fig. 2, where an instantaneous shear strain is applied. For \( \varphi < \varphi_G \), we do not see aging effects within our numerical accuracy. The zero-field compression (ZFC) and the field compression (FC) shear stresses converge quickly to the same value. For \( \varphi > \varphi_G \), no converge is observed within our simulation time window. The \( \sigma_{ZFC}(\tau, t_w) \) displays a plateau for short \( \tau \), followed by slow decay. Note that the time scale \( \tau = 1 \) used in determining \( \mu_{ZFC} \) (see the main text) is in the plateau region.

Time evolution of the pressure

In the main text, we plot the rescaled shear stress \( \hat{\sigma} = \sigma/p \) in Figs. 2 and 3, since a simple scaling relation \( \sigma \sim p \) is expected in the normal glass phase. Here we examine whether the pressure \( p \) depends on time and protocol. Indeed, Supplementary Figure 11 shows that, in contrast to the stress, the pressure is nearly time-independent and protocol-independent after instantaneous shear, both below and above the Gardner transition. Therefore, \( \hat{\sigma} \) truly reveal the behavior of stress since we can treat \( p \) as a constant at any \( \varphi \).

SUPPLEMENTARY NOTE 4 – ADDITIONAL DATA ON THE PROTOCOL-DEPENDENT SHEAR MODULUS

Here we report supplementary data on the protocol-dependent shear modulus. We discuss how the ZFC and the FC shear moduli depend on the shear strain \( \gamma \), the number of particles \( N \), equilibrium density \( \varphi_E \), and the waiting time \( t_w \). We also measure the shear modulus using a third protocol.

Dependence on the shear strain \( \gamma \)

In our analysis, the shear modulus is measured by taking the ratio between the stress and the strain, i.e., \( \mu_{ZFC} = (\sigma_{ZFC} - \sigma_0)/\gamma \) and \( \mu_{FC} = (\sigma_{FC} - \sigma_0)/\gamma \), where \( \sigma_0 \) is the remanent shear stress. If \( \gamma \) is sufficiently small such that the non-linear corrections are negligible, the measured modulus should be independent of \( \gamma \). Our data show that the FC modulus \( \mu_{FC} \) is indeed in such a linear regime for the chosen \( \gamma \) (see Fig. 3b in the main text for \( N = 1000 \) systems, and Supplementary Figure 12 for \( N = 500 \) and \( N = 2000 \) systems). However, \( \gamma \)-dependence is observed for \( \mu_{ZFC} \): at large pressure \( p \) close to jamming, \( \mu_{ZFC} \) increases for smaller shear strain \( \gamma \). For smaller \( \gamma \), the large-\( p \) scaling \( \mu_{ZFC} \sim p^\gamma \) agrees better with the mean-field theory, for any \( N \) studied, but additional data are required to conclude if the mean-field result is coincided in the limit \( \gamma \to 0 \). Recently, a very careful study shows that the mean-field jamming exponents, which characterize the critical distribution of small inter-particle gaps and weak contact forces, are consistent with simulation data in finite dimensions, after removing localized buckling excitations [57]. Such an analysis in the \( p \to \infty \) limit is beyond the present numerical accuracy.

Dependence on the number of particles \( N \)

For a fixed \( \gamma \), Fig. 3 in the main text shows that at large \( p \), \( \mu_{ZFC} \) decreases with increasing \( N \). It suggests that the non-linear effect, associated to stress relaxation due to MPEs, is stronger in larger systems. Indeed, in Ref. [53], the authors found a finite-size scaling \( \delta \gamma_1 \sim N^\beta \) with \( \beta \approx -0.62 \), for the mean strain \( \delta \gamma_1 \) at which the first MPE takes place in amorphous solids. This scaling suggests that, MPEs are easier to occur in larger systems, and become unavoidable at any finite shear strain in the thermodynamic limit, because \( \delta \gamma_1 \to 0 \) as \( N \to \infty \). It is thus reasonable to see that data with smaller \( \gamma \) (for a fixed \( N \)) or smaller \( N \) (for a fixed finite \( \gamma \)) obeys better the mean-field scaling \( \mu_{ZFC} \sim p^{1.41574} \), because the theoretical \( \mu_{ZFC} \) is only concerned about the linear response [10].

In our data (Fig. 3b in the main text and Supplementary Figure 12), we do not find appreciable \( N \)-dependence of \( \mu_{FC} \). In contrast, a scaling relation \( \mu_{FC} \sim N^{-0.25} \) was reported in Ref. [13]. Here we discuss possible reasons for this discrepancy. In [13], the systems are quenched from completely random initial configurations. Compared to the case in well equilibrated systems, in such non-equilibrium systems, the stress relaxes much faster and eventually decays to zero, i.e., the system melts quickly [18] (see Supplementary Figure 9 for an illustration). Considering that larger
systems have an easier tendency to relax, we expect that in the thermodynamic limit, the system turns to a liquid within the simulation time scale used in Ref. [13], which is the reason why $\mu_{\text{FC}} \to 0$.

Dependence on the initial equilibrium density $\varphi_g$

We find that our basic observation – the bifurcation between the ZFC shear modulus $\mu_{\text{ZFC}}$ and the FC shear modulus $\mu_{\text{FC}}$ at the Gardner transition $\varphi_G$ – is independent of the initial equilibrium density $\varphi_g$ (see Supplementary Figure 13). Note that the value of $\varphi_G$ itself depends on $\varphi_g$. The large pressure ($\varphi \gg \varphi_G$) scalings, $\mu_{\text{ZFC}}$ versus $p$, and $\mu_{\text{FC}}$ versus $p$, are nearly unchanged for different $\varphi_g$. Additionally, we compare our simulation data with theoretical predictions for $\varphi < \varphi_G$. We plot $\bar{\mu}_{\text{ZFC}}/\bar{\mu}_g$ and $\bar{\mu}_{\text{FC}}/\bar{\mu}_g$ as functions of $p/p_g$ obtained from simulations, together with the mean-field state following theory [29], where $\bar{\mu} = \mu/p$ is the modulus rescaled by the pressure, and $\mu_g$ and $p_g$ are the shear modulus and the pressure at $\varphi_g$. Note that the theory does not distinguish between ZFC and FC moduli in this regime. On this rescaled plot, the theory and the simulation data show similar behaviors, both of which are insensitive to $\varphi_g$. We point out that the mean-field theory uses an over-simplified liquid EOS, that is only valid for mono-disperse hard spheres in the large dimensional limit. Thus a direct comparison between the theory and our simulation is impossible. However, once the effect of this mismatch on the liquid structure is removed by a proper rescaling with respect to the reference point at $\varphi_g$, the theory basically captures the general trend on how the system evolves under a slow compression annealing.

Dependence on the waiting time $t_w$

In Supplementary Note 3, we have discussed how $\sigma_{\text{ZFC}}(\tau, t_w)$ and $\sigma_{\text{FC}}(\tau, t_w)$ relax with $\tau$ under a quasi-static shear strain $\gamma$. Based on the data (Supplementary Figure 10), we choose time scales $\tau = 1$ and $t_w = 10$ to measure $\mu_{\text{ZFC}}$ and $\mu_{\text{FC}}$ (see the main text). The scale $\tau = 1$ is chosen within the first plateau regime of $\sigma_{\text{ZFC}}(\tau, t_w)$. Note that for larger $\tau$, the difference $\mu_{\text{ZFC}} - \mu_{\text{FC}}$ would decrease, and would eventually vanish as $\tau \to \infty$, even in the Gardner phase. On the other hand, in order to examine the $t_w$-dependence more carefully, we obtain additional data of $\mu_{\text{ZFC}}$ and $\mu_{\text{FC}}$ for a few different $t_w$ (we fix $\tau = 1$ in all cases). Interestingly, we found that the differences $\mu_{\text{ZFC}} - \mu_{\text{FC}}$ obtained by using different $t_w$, when plotted as a function of $\varphi$, collapse onto the same curve, within our numerical accuracy (Supplementary Figure 14). In particular, because the Gardner transition is determined as the bifurcation point between $\mu_{\text{ZFC}}$ and $\mu_{\text{FC}}$, this result shows that the location of the transition point is independent of $t_w$ in our approach.

A third protocol

To further verify and emphasize the protocol dependence on the shear modulus, we design a third protocol, in which we apply an additional shear strain after the FC procedure. In this protocol, we first apply a small quasi-static strain $\gamma$ at $\varphi_g$, compress the system to $\varphi$, and then after waiting for $t_w = 10$ and $\tau = 1$, we measure the stress $\sigma_{\text{FC}}$. This procedure is basically the same as the FC (see the main text). We then apply an additional quasi-static strain $\gamma$ at $\varphi$, and measure the stress $\sigma_{\text{FCS}}$ after waiting for $\tau = 1$. The FCS (FC+shear) modulus is defined as $\mu_{\text{FCS}} = (\sigma_{\text{FCS}} - \sigma_{\text{FC}})/\gamma$. Supplementary Figure 15 shows that this shear modulus is close to $\mu_{\text{ZFC}}$, and clearly different from $\mu_{\text{FC}}$. From the view point of free energy landscape, for $\varphi > \varphi_G$, $\mu_{\text{FCS}}$ represents the local curvature of the sub-basins at a finite given $\gamma$ (see Fig. 4 in the main text), while $\mu_{\text{ZFC}}$ is the local curvature of the sub-basins at $\gamma = 0$.

SUPPLEMENTARY NOTE 5 – PROTOCOL-DEPENDENT SHEAR MODULUS OF BIDISPERSE HARD DISKS

To test if the ZFC/FC approach can be applied to other systems, we also study a two-dimensional bidisperse hard disk model glass former. The system contains $N = 1000$ equimolar bidisperse hard disks with diameter ratio $D_1 : D_2 = 1.4 : 1$. The dynamical crossover density is $\varphi_d = 0.790(1)$ [34]. The example in Supplementary Figure 16 shows that the shear modulus becomes protocol-dependent for $\varphi > \varphi_G$. This signature can be used to determine $\varphi_G$, whose value is fully consistent with the previous independent estimate [34].
**Supplementary Methods – Numerical Protocols**

**Compression protocols**

*Generation of dense equilibrium liquids* – To prepare dense equilibrium configurations, we combine the Lubachevsky-Stillinger (LS) algorithm \[37\] with swap Monte Carlo moves \[34, 36\]. The LS algorithm consists of standard event-driven molecular dynamics (MD), and slow compression which is realized by variation of the diameter of particles. Our protocol consists of the following two steps:

1. Starting from an ideal gas configuration, we first compress it to \( \varphi_0 = 0.54 \), by growing spheres at a constant rate \( \delta \varphi = 10^{-3} \), such that \( D(t) = D(0)(1 + \delta \varphi t) \). Because the process is equivalent to compression, hereafter we call \( \delta \varphi \) as compression rate. This initial compression is fast enough to suppress crystallization, and slow enough to equilibrate the configuration up to \( \varphi_0 \).

2. Starting from the equilibrium configuration at \( \varphi_0 \), we switch to a slower compression rate \( \delta \varphi = 10^{-5} \), and compress the configuration to a higher density \( \varphi_g \). Swap attempts are introduced: we randomly pick a pair of particles and exchange their diameters if no overlap is created after the swap. The particle sizes do not change during swap moves. We perform 10\% swap moves and 90\% LS molecular dynamics steps. After the compression, we further relax the configuration for \( t = 1000 \) to check that the pressure does not change. The equation of state (EOS) of our equilibrium configurations agrees with the Carnahan-Stillinger (CS) expression (see Supplementary Figure 1) \[58\],

\[
p_{\text{CS}}(\varphi) = \frac{1}{1 - \varphi} + \frac{3s_1s_2}{s_3} \frac{\varphi}{(1 - \varphi)^2} + \frac{s_3^3 (3 - \varphi) \varphi^2}{s_3^2 (1 - \varphi)^3},
\]

(Supplementary Equation 4)

where \( s_k \) is the k-th moments of the diameter distribution function \( P(D) \).

*Generation of glasses* – The swap algorithm is switched off once the equilibrated configuration at the target \( \varphi_g \) is obtained. All the subsequent simulations are performed using the MD without swap. Since the \( \alpha \)-relaxation time has become much larger than our MD simulation time scales, we are left with a piece of glass. We call the configuration of the particle positions \( \{ r_i \} (i = 1, 2, \ldots, N) \) of such a glass at \( \varphi_g \) as a sample. From each of such a sample at \( \varphi_g \), we generate many realizations by setting random particle velocities \( \{ v_i \} (i = 1, 2, \ldots, N) \) drawn from the Maxwell-Boltzmann distribution. Each of such realizations is compressed by the LS algorithm to obtain compressed glasses at desired densities \( \varphi > \varphi_g \). Note that the kinetic energy is conserved so that the system remains at the unit temperature throughout our simulations.

**Shear protocols**

*Quasi-static shear* – In the quasi-static shear, the shear strain \( \gamma \) is increased with time at a constant rate \( \dot{\gamma} \), which is set small enough such that the system is quasi-equilibrated at each step. The protocol consists of the following steps:

1. Increase the shear strain \( \gamma \) instantaneously by an infinitesimal amount \( \gamma \rightarrow \gamma + \delta \gamma \) with \( \delta \gamma = 10^{-4} \). We perform an affine deformation to all particles, whose positions are shifted by \( x_i \rightarrow x_i + \delta \gamma z_i \), where \( x_i \) and \( z_i \) are the \( x- \) and \( z- \)coordinates of particles \( i \). This instantaneous shift could introduce overlaps between some particles, which are removed by using the the conjugated gradient (CG) method \[59\]. To use CG, a harmonic inter-particle potential \( \phi_{ij}(r) = (1 - r/D_{ij})^2 \) (zero when \( r > D_{ij} \)) is used, where \( D_{ij} = (D_i + D_j)/2 \) is the average diameter of particles \( i \) and \( j \). The boundary condition in the \( z \) direction satisfies the Lees-Edwards scheme \[41\], i.e., the \( x- \)position of the top (bottom) imaginary box is shifted by \( \delta \gamma L (-\delta \gamma L) \), where \( L \) is the linear size of the simulation box. After this step, we obtain an non-overlapping hard sphere (HS) configuration under shear strain \( \gamma + \delta \gamma \).

2. We switch the soft potential back to the hardcore potential, and equilibrate the system by using the event-driven MD to simulate the dynamics of HSs under fixed shear strain \( \gamma + \delta \gamma \). Again we emphasize that the dynamics preserves the kinetic energy so that the system remains at a constant temperature. The velocities are reinitialized after each step of the shear strain. The Lees-Edwards boundary condition is kept. We perform LS simulation for a duration \( \delta t \), such that \( \delta \gamma / \delta t = \dot{\gamma} \).

3. The above two steps are repeated until the shear strain reaches a target value.
To simulate quasi-static shear, we choose a sufficiently small $\dot{\gamma} = 10^{-4}$. We have checked that for $\varphi < \varphi_G$, the stress-strain response is independent of $\dot{\gamma}$ when $\dot{\gamma}$ is decreased from $10^{-3}$ to $10^{-5}$ (see Supplementary Note 3).

Instantaneous shear – To simulate instantaneous shear, we instantaneously increase the shear strain from 0 to $\gamma$. We then turn on the harmonic soft-potential, and use the CG algorithm to remove the overlaps. Different from the quasi-static shear, the system is generally far away from equilibrium after the instantaneous shear.

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