Supplemental Material for: “Anisotropic Structural Predictor in Glassy Materials”

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The goal of this document is to provide additional technical details regarding the results reported on in the manuscript.

S-1. Glass model, preparation and deformation

\textbf{Glass model} — We employ a glass-forming model system in two-dimensions composed of a 50:50 binary mixture of ‘large’ and ‘small’ particles of equal mass \( m \), interacting via radially-symmetric purely repulsive inverse-power-law pairwise potentials, that follow

\[
\varphi(r_{ij}) = \left\{ \begin{array}{ll}
\epsilon \left( \frac{A_{ij}}{r_{ij}} \right)^n + \sum_{\ell=0}^{q} c_{2\ell} \left( \frac{A_{ij}}{r_{ij}} \right)^{2\ell} & , \quad r_{ij} \leq x_c \\
\frac{r_{ij}}{x_{ij}} & , \quad r_{ij} > x_c
\end{array} \right.
\] (S1)

where \( r_{ij} \) is the distance between the \( i \)th and \( j \)th particles, \( \epsilon \) is an energy scale, and \( x_c \) is the dimensionless distance for which \( \varphi \) vanishes continuously up to \( q \) derivatives. Distances are measured in terms of the interaction lengthscale \( \lambda \) between two ‘small’ particles, and the rest are chosen to be \( \lambda_{ij} = 1.18\lambda \) for one ‘small’ and one ‘large’ particle, and \( \lambda_{ij} = 1.4\lambda \) for two ‘large’ particles. The coefficients \( c_{2\ell} \) are given by

\[
c_{2\ell} = \frac{(-1)^{\ell+1}}{(2q-2\ell)!} \frac{(n+2q)!}{(n-2)!} \frac{1}{(n+2\ell)!} a_c^{-(n+2\ell)}. \quad (S2)
\]

We chose the parameters \( x_c = 1.6, n = 10, \) and \( q = 4 \). The density has been set to be \( N/V = 0.86\lambda^{-2} \); this choice sets the scale of characteristic \( T = 0 \) interaction energies to be of order unity. This model undergoes a computer-glass-transition at a temperature of \( T_g \approx 0.5\epsilon/k_B \) for the chosen density. We note that while in this work we present results for a pairwise interaction potential, cf. Eq. (S1), our approach is completely general and applies also to 3-body (or higher) interaction potentials, as explained in the Supporting Information of \cite{1}.

\textbf{Preparation protocol} — We prepared an ensemble of glassy samples using the following protocol: first, systems were equilibrated in the high temperature liquid phase at \( T = 1.0\epsilon/k_B \). Then, the temperature was instantaneously set to a target value just below \( T_g \) of the model, where the dynamics have been run for a duration \( t_{\text{anneal}} = 100\tau_0 \), where \( \tau_0 \equiv \lambda \sqrt{m/\epsilon} \) is the microscopic units of time. This short annealing step is necessary to avoid generating unphysical ultra-unstable glassy configurations that could occur in an instantaneous quench, and is computationally advantageous compared to a continuous quench at a fixed quench-rate. After the annealing step we minimized the energy to produce glassy samples by a standard conjugate gradient method. Using this protocol, we have generated 10000 independent glassy samples, with \( N = 10000 \). This system size has been selected as it is sufficiently large to include several soft spots, yet sufficiently small in terms of the associated computational cost.

\textbf{Athermal Quasi-Static (AQS) deformation} — The performed athermal quasi-static (AQS) deformation simulations followed well-established two-step protocols of first imposing an affine transformation \( \mathcal{H}(\gamma) \) (either simple or pure shear, in either the positive or negative directions, see manuscript for more details) to the system and then minimizing its energy while enforcing Lees-Edwards boundary conditions, see e.g. \cite{2, 3}. These AQS simulations have been used both to validate the analytic linear response results and to test the predictive power of the proposed structural predictor against actual plastic rearrangements/events. For the latter, the energy of the system has been used to identify plastic rearrangements/events with strain precision up to \( 10^{-8} \) using backtracking methods. The plastic events have been automatically spatially localized by selecting the particle with the largest displacement value as a consequence of the energy minimization step at the occurrence of a plastic event.

S-2. Linear response coupling of the LHC to external deformation: Complete expression, numerical validation and visualization

\textbf{Complete analytic expression and numerical validation} — As explained in the manuscript, \( dc_{\alpha}/d\gamma \) for a given \( \mathcal{H}(\gamma) \) is obtained by operating on the LHC

\[
c_{\alpha} = \varphi''_{\alpha} \cdot \mathcal{M}^{-1} - f_{\alpha} \cdot \mathcal{M}^{-1} \cdot \mathcal{U}'' \cdot \mathcal{M}^{-1} \quad (S3)
\]

with the following differential operator

\[
\frac{d}{d\gamma} = \frac{\partial}{\partial \gamma} - \mathcal{U}'' \cdot \mathcal{M}^{-1} \cdot \frac{\partial}{\partial x}. \quad (S4)
\]

The result takes the form
where we used the shorthand notation $\Xi \equiv U^{(M)}$ for the mismatch force vector (recall that the superscript $U$ denotes the partial derivative $\partial / \partial \gamma$). Note that in the manuscript, cf. Eq. (2), only the leading order contribution in $M^{-1}$ has been reported and a compact tensorial notation has been used, while in Eq. (S5) we provide the complete expression in component/index form.

The dependence of $dc_\alpha / d\gamma$ in Eq. (S5) on a particular imposed deformation tensor $H(\gamma)$ is encapsulated in the partial derivative $\partial / \partial \gamma$ that can be expressed as

$$\frac{\partial}{\partial \gamma} = \sum_{i \in c} x_{ij} \cdot \frac{dH^T}{d\gamma} \bigg|_{\gamma=0} \cdot \frac{\partial}{\partial x_{ij}}, \quad (S6)$$

where $x_{ij}$ is the inter-particle vector connecting the positions of the $i^{th}$ and the $j^{th}$ particles (using $x_{ij}$ is natural as the pairwise potential energy $\varphi$, cf. Eq. (S1), depends on the pairwise distance between interacting particles, $|x_{ij}| = r_{ij}$). Therefore, the dependence on the applied deformation $H(\gamma)$ is fully contained in $dH^T / d\gamma$ (evaluated at $\gamma=0$).

In order to validate the analytic expression for $dc_\alpha / d\gamma$ in Eq. (S5), we deformed 1000 glass realizations under both simple and pure shear conditions to a strain of $\Delta \gamma = 10^{-5}$ and calculated the finite differences ratio $\Delta c_\alpha / \Delta \gamma$, where Eq. (S3) has been used to obtain $\Delta c_\alpha \equiv c_\alpha(\Delta \gamma) - c_\alpha(0)$. The value of $\Delta \gamma$ in the deformation simulations has been chosen to be small enough for the system to remain in the linear response regime (and in particular to avoid plastic rearrangements) and large enough for the results to be properly distinguished from the inherent noise in the calculations. In Fig. S1 we present the cumulative distribution function $C(Err)$, where the error is quantified by

$$Err = 100 \times \frac{|dc_\alpha / d\gamma - \Delta c_\alpha / \Delta \gamma|}{\min(|dc_\alpha / d\gamma|, |\Delta c_\alpha / \Delta \gamma|)} \quad (S7)$$

for both simple and pure shear. The results, which are as expected the same for simple and pure shear, quantitatively support the validity of the analytic result for $dc_\alpha / d\gamma$ in Eq. (S5).

**Visualization of interaction-wise fields** — In Figs. 1 and 3 in the manuscript we present visualizations of interaction-wise fields of both the LHC $c_\alpha$ and of the products $c_\alpha dc_\alpha / d\gamma$. The visualization has been carried out as follows: given an interaction-wise field $x_\alpha$ ($\alpha$ is an interaction index, and $x_\alpha$ represents either $c_\alpha$ or $c_\alpha dc_\alpha / d\gamma$), we first rescale the field such that the average of the absolute magnitude of the field values is unity. We then threshold the rescaled field $\tilde{x}_\alpha$ by setting $\tilde{x}_\alpha = \tilde{x}_0$ for all $\tilde{x}_\alpha > \tilde{x}_0$, and discard of all interactions for which $\tilde{x}_\alpha < 1$. The clearest visualization is obtained at $\tilde{x}_0=6.6$. The line widths presented in Figs. 1 and 3 of the manuscript are proportional to $\tilde{x}_\alpha$, and their color represent the sign of $\tilde{x}_\alpha$, with black (red) representing positive (negative) values. The exact same procedure as explained above was carried out for all presented interaction-wise fields; the differences between Fig. 3a and Figs. 3b-c stem from the different forms of the respective distributions of the different observables presented.

Finally, we note that while in the example presented in Figs. 1 and 3 the first plastic events in the 4 different de-
FIG. S2. The bin size $\xi$ dependence of $C(\lambda)$, defined in the manuscript and plotted in Fig. 4, for both (a) $c_\alpha d c_\alpha / d\gamma$ (under positive simple shear deformation) and (b) $|c_\alpha|$. The bin sizes shown are $\xi = 4$ (brown line and squares), $\xi = 5$ (orange line and diamonds, these results are identical to those presented in Fig. 4 in the manuscript), and $\xi = 6$ (green line and circles). Results without coarse-graining (i.e. no binning at all) are added as a reference (black line). The predictive power of both $c_\alpha d c_\alpha / d\gamma$ and $|c_\alpha|$ somewhat depends on $\xi$, and appears to improve with decreasing it in the range considered here. Note that no attempt has been made in this work to optimize the predictive power with respect to $\xi$.

**S-3. The predictive power of the structural predictor: Bin size effect and results for pure shear**

**Bin size effect** — As explained in the manuscript in detail, the metric we used to quantify the predictive power of structural indicators depends on a single, physically meaningful parameter $\xi$ that represents the typical linear size of soft spots. The latter can be estimated from the localization length of soft quasilocalized modes [4, 5], which is determined by the linear size of their disordered core, roughly composed of 10 particles in linear size. Since in our metric the value assigned to each bin is averaged over neighboring bins, the latter is consistent with a bin size of $\xi = 5$, which has been used in the manuscript. In Fig. S2 we quantify the effect of $\xi$ on the predictive power of both $c_\alpha d c_\alpha / d\gamma$ (panel a) and $|c_\alpha|$ (panel b), by showing $C(\lambda)$ for $\xi = 4, 5, 6$ (from the top curve to the bottom one, respectively, where the $\xi = 5$ results are identical to those presented in Fig. 4 in the manuscript) under simple shear in the positive direction. It is observed that $C(\lambda)$ somewhat depends on the value of $\xi$, with better quantitative results obtained for smaller values of $\xi$, and that both $c_\alpha d c_\alpha / d\gamma$ and $|c_\alpha|$ exhibit similar trends. Despite that the results appear to quantitatively improve as $\xi$ is reduced, we did not aim at optimizing the predictive power relative to $\xi$ and simply used the physically sensible choice of $\xi = 5$. We note in passing that bins of 5 particles have also been used in [6].

FIG. S3. The same as Fig. 4 in the manuscript, where the predictability function $C(\lambda)$ is shown (see manuscript for definitions), but for pure shear in (a) the positive direction and (b) the negative one. The symbols and color code are the same as in Fig. 4 in the manuscript, but for pure instead of simple shear. In order to further highlight the orientation-dependence of $c_\alpha d c_\alpha / d\gamma$, we also added the results for $C(\lambda)$ when the glass is deformed under positive (yellow solid line) and negative (black dashed line) simple shear.

**Results for pure shear** — In the manuscript, results for simple shear in both the positive and negative directions have been presented in Fig. 4. For completeness, we present here the corresponding results for pure shear deformation in Fig. S3. The figure demonstrates that the results for positive and negative pure shear deformation are essentially identical for those for positive and negative pure shear deformation, as expected from the isotropy of the initial glass state.