Mechanical properties of simple computer glasses

Edan Lerner
Institute for Theoretical Physics, University of Amsterdam, Science Park 904, 1098 XH Amsterdam, The Netherlands

Recent advances in computational glass physics enable the study of computer glasses featuring a very wide range of mechanical and kinetic stabilities. The current literature, however, lacks a comprehensive data set against which different computer glass models can be quantitatively compared on the same footing. Here we present a broad study of the mechanical properties of several popular computer glass forming models. We examine how various dimensionless numbers that characterize the glasses’ elasticity and elasto-plasticity vary under different conditions — in each model and across models — with the aim of disentangling the model-parameter-, external-parameter- and preparation-protocol-dependences of these observables. We expect our data set to be used as an interpretive tool in future computational studies of elasticity and elasto-plasticity of glassy solids.

I. INTRODUCTION

Computational studies of glass formation and deformation constitute a substantial fraction of the research conducted in relation to these problems. The attention drawn by this line of work has been on the rise recently due to several methodological developments that allow investigators to create computer glasses with a very broad variation in the degree of their mechanical and kinetic stability. These include the ongoing optimization of GPU-based algorithms \cite{1,2} that now offer the possibility to probe several orders of magnitude in structural relaxation rates in the supercooled liquid regime \cite{3}. Various sampling methods based on generalized statistical ensembles have been shown to yield well-annealed states \cite{4}. In groundbreaking work of Berthier and coworkers \cite{5}, inspired by previous advances \cite{6}, a glass forming model was optimized such to stupendously increase the efficiency of the Swap Monte Carlo algorithm, allowing the equilibration of supercooled liquids down to unprecedented low temperatures, while remaining robust against crystallization. In \cite{7} a model and algorithm were put forward that allows to create extremely stable computer glasses, albeit with a protocol which is not physical. Mechanical annealing by means of oscillatory shear was also recently shown to be an efficient protocol for creating stable glasses \cite{8}. Finally, numerical realizations of experimental vapor deposition protocols \cite{9} have shown good success in creating well-annealed glasses \cite{10,11}.

This recent proliferation of methods for creating stable computer glasses highlights the need for approaches to meaningfully and quantitatively compare between the various glasses created by these methods. In particular, it is important to disentangle the effects of parameter choices — both in the interaction potentials that define computer glass formers, and choices of external control parameters — from the effects of annealing near and below the models’ respective computer glass transition temperatures. In addition, in some cases it is useful to quantitatively assess the effective distance a given computer glass is positioned away from the unjamming point — the loss of rigidity seen e.g. upon decompressing ensembles of repulsive soft spheres \cite{12,13}.

This work is aimed towards establishing how elastic properties and elasto-plastic responses of simple computer glasses depend on various key external and internal control parameters, how they change between different models, and how they are affected by the preparation protocol of glasses. In order to disentangle annealing effects from model- and external-parameter dependences, we exploit the observation that creating computer glasses by instantaneous quenches of high energy states to zero temperature defines an ensemble of configurations whose elastic properties can be meaningfully and quantitatively compared between models and across different parameter regimes.

![Fig. 1](image-url) FIG. 1. (a) Sample-to-sample mean athermal shear modulus $G$ measured in inherent states that underlie liquid states at equilibrium parent temperatures $T_0$ of the POLY model, see Sect. II A 4 for model details. The vertical line approximates the crossover temperature above which several elastic properties saturate. (b) Sample-to-sample mean inherent state potential energy-per-particle $U/N$. Interestingly, while $G$ saturates above the crossover temperature, $U/N$ does not. In this work we focus on several observables that feature a saturation as seen for $G$ in panel (a).

The existence of the aforementioned ensemble is demonstrated in Fig. 1 where we plot measurements of the sample-to-sample mean athermal shear modulus (see definition below) of underlying inherent states of parent
equilibrium configurations (labelled by their equilibrium temperature $T_d$) of a simple glass-forming model (see details in Sect. II A below). This high-temperature saturation of elastic properties of very poorly annealed glassy states appears to be a generic feature of computer glasses [15, 16]. We therefore carry out in what follows a comparative study of elastic properties of different computer glass models created by instantaneous quenches from high energy states. Our analyses of elastic properties of instantaneously-quenched glasses are compared against the behavior of the same key observables measured in a variant of the glass forming model introduced in [2] that can be annealed very deeply below the conventional computer glass transition temperature. This allows us to compare the relative protocol- and parameter-induced variation in these key observables.

In the same spirit, we also investigate the elastoplastic steady state as seen by deforming our instantaneously-quenched glasses using an athermal, quasistatic shear protocol, that gets rid of any rate effects associated with finite deformation rate and finite temperature protocols. We anticipate our results to constitute a benchmark for quantitative assessment of other model glasses in future studies of elasticity, elastoplasticity and glass formation.

This paper is structured as follows; in Sect. II we spell out the models employed in our study, and list the physical observables that were calculated in those models. Sect. III presents various data sets that characterize the elasticity and elastoplasticity of the computer glasses we have investigated, and discusses various points of interest and connections to related previous work. Our work is summarized in Sect. IV.

II. MODELS, METHODS AND OBSERVABLES

In this Section we provide details about the model glass formers we employed in our study, and explain the methods used to create glassy samples. We then spell out the definitions of all reported observables.

A. Computer glasses

We have studied 4 model glass formers in $d=3$ dimensions. We have created ensembles of 1000 configurations of $N=8000$ particles for each model system, and for each value of the respective control parameter (see below).

1. Inverse-power-law

The inverse-power-law (IPL) model is a 50:50 binary mixture of 'large' and 'small' particles of equal mass $m$. Pairs of particles $i, j$ at distance $r_{ij}$ from each other interact via the inverse-power law pairwise potential

$$
\varphi_{\text{IPL}}(r_{ij}) = \left( \frac{\alpha_{ij}}{r_{ij}} \right)^{\beta},
$$

where $\varepsilon$ is a microscopic energy scale. Distances in this model are measured in terms of the interaction length-scale $\lambda$ between two 'small' particles, and the rest are chosen to be $\lambda_{ij} = 1.18a$ for one 'small' and one 'large' particle, and $\lambda_{ij} = 1.4a$ for two 'large' particles. For computational efficiency, we also employ a variant of the IPL model with a finite cutoff in the pairwise interactions, of the form

$$
\varphi_{\text{IPL}}(r_{ij}) = \left\{ 
\begin{array}{ll}
\varepsilon \left[ \left( \frac{\lambda_{ij}}{r_{ij}} \right)^{\beta} + \sum_{\ell=0}^{q} c_{2\ell} \left( \frac{r_{ij}}{\lambda_{ij}} \right)^{2\ell} \right], & r_{ij} \leq x_c, \\
0, & r_{ij} > x_c.
\end{array}
\right.
$$

where $x_c$ is the dimensionless distance for which $\varphi_{\text{IPL}}$ vanishes continuously up to $q$ derivatives. We chose the parameters $q = 2$ and $x_c = 1.5$. The coefficients $c_{2\ell}$, determined by demanding that $\varphi$ vanishes continuously up to $q$ derivatives, are given by

$$
c_{2\ell} = \frac{(-1)^{\ell+1}(\beta+2\ell)!!(\beta+2\ell)!!}{(2\ell-2\ell)!!(2\ell)!!} x_c^{-(\beta+2\ell)}. \quad (3)
$$

The control parameter of interest for this system is the exponent $\beta$ of the inverse-power-law pairwise interaction. Glassy samples were created by placing $N=8000$ particles randomly on a cubic lattice and minimizing the potential energy by a conjugate gradient minimization. The finite-cutoff variant was used for $\beta=12$, then we set the number density $\rho = N/V = 2.0$, for $\beta > 12$, then we set $\rho = 0.82$, and for $\beta = 4$, for which we set $\rho = 10.0$. In Sect. III A we motivate these number density choices.

2. Hertzian spheres

The Hertzian spheres model (HRTZ) we employ is a 50:50 binary mixture of soft spheres with equal mass $m$ and a 1:1.4 ratio of the radii of small and large particles. The units of length $\lambda$ are chosen to be the diameter of the small particles, and $\varepsilon$ denotes the microscopic units of energy. Pairs of particles whose pairwise distance $r_{ij}$ is smaller than the sum of their radii $R_i + R_j$ interact via the Hertzian pairwise potential

$$
\varphi_{\text{Hertz}}(r_{ij}, R_i, R_j) = \frac{2\varepsilon}{5^{2/3} x_c} \left( (R_i + R_j) - r_{ij} \right)^{5/2}, \quad (4)
$$

and $\varphi_{\text{Hertz}} = 0$ otherwise.

In this model we control the imposed pressure; glassy samples at target pressures of $p = 10^{-1}, 10^{-2}, 10^{-3}, 10^{-4}, 10^{-5}$ were created by combining a Berendsen barostat [17] into the FIRE minimization algorithm [18]. Initial states at the highest pressure were created by placing particles randomly on a cubic lattice, followed by minimizing the potential energy. Subsequent lower pressure glasses were created by changing the target pressure and relaunched the minimization algorithm.
3. Kob-Andersen binary Lennard-Jones

We employ a slightly modified variant of the well-studied Kon-Andersen binary Lennard-Jones (KABLJ) glass former [9], which is perhaps the most widely studied computer glass model. Our variant of the KABLJ model is a binary mixture of 80% type A particles and 20% type B particles, that interact via the pairwise potential

\[
\varphi_{\text{KABLJ}}(r_{ij}) = 4\varepsilon_{ij} \left( \left( \frac{r_{ij}}{\lambda_{ij}} \right)^{12} - \left( \frac{r_{ij}}{\lambda_{ij}} \right)^{6} + c_4 \left( \frac{r_{ij}}{\lambda_{ij}} \right)^4 + c_2 \left( \frac{r_{ij}}{\lambda_{ij}} \right)^2 + c_0 \right),
\]

if \( r_{ij}/\lambda_{ij} \leq 2.5 \), and \( \varphi_{\text{KABLJ}} = 0 \) otherwise. Lengths are expressed in terms of \( \lambda_{AA} \), then \( \lambda_{AB} = 4/5 \) and \( \lambda_{BB} = 22/25 \). Energies are expressed in terms of \( \varepsilon_{AA} \), then \( \varepsilon_{AB} = 3/2 \) and \( \varepsilon_{BB} = 1/2 \). Both particle species share the same mass \( m \). The coefficients \( c_4, c_2 \) and \( c_0 \) are chosen such that \( \varphi_{\text{KABLJ}}, \varphi'_{\text{KABLJ}} \) and \( \varphi''_{\text{KABLJ}} \) vanish at \( r_{ij}/\lambda_{ij} = 5/2 \). In this model we control the density \( \rho \equiv N/V \) with \( V \) denoting the volume.

4. Polydisperse soft spheres

The computer glass model we employed is a slightly modified variant of the model put forward in [3]. We enclose \( N \) particles of equal mass \( m \) in a square box of volume \( V = L^3 \) with periodic boundary conditions, and associate a size parameter \( \lambda_i \) to each particle, drawn from a distribution \( p(\lambda) \sim \lambda^{-3} \). We only allow \( \lambda_{\text{min}} \leq \lambda_i \leq \lambda_{\text{max}} \), forming our units of length, and \( \lambda_{\text{max}} = 2.22\lambda \). The number density \( N/V = 0.58\lambda^{-3} \) is kept fixed. Pairs of particles interact via the same pairwise interaction give by Eq. (2). We chose the parameters \( x_c = 1.4, n = 10, \) and \( q = 3 \). The pairwise length parameters \( \lambda_{ij} \) are given by

\[
\lambda_{ij} = \frac{1}{2}(\lambda_i + \lambda_j)(1 - n_a|\lambda_i - \lambda_j|).
\]

Following [3] we set the non-additivity parameter \( n_a = 0.1 \). In what follows energy is expressed in terms of \( \varepsilon \), temperature is expressed in terms of \( \varepsilon/k_B \) with \( k_B \) the Boltzmann constant, stress, pressure, and elastic moduli are expressed in terms of \( \varepsilon/\lambda^3 \). This model is referred to in what follows as POLY.

Ensembles of equilibrium states of the POLY model were created using the Swap Monte Carlo method [5, 6]; within this method, trial moves include exchanging (swapping) the size parameters of pairs of particles, in addition to the conventional random displacements of particles. For each temperature we have simulated 50 independent systems of \( N = 8000 \) particles, and collected 20 configurations for each system that were separated by at least the structural relaxation time (here time is understood as Monte-Carlo steps) as measured by the stress autocorrelation function, resulting in equilibrium ensembles of 1000 members for each parent temperature \( T_0 \). Ensembles of inherent states were created by performing an instantaneous quench of equilibrium states from each parent temperature by means of a conjugate gradient minimization of the potential energy.

We note that since particle size parameters are sampled from a rather broad distribution, and our simulated systems are of only \( N = 8000 \) particles, very large finite-size sampling-induced fluctuations of the equilibrium energy of different systems (which are entirely absent in e.g. binary systems such as the KABLJ) can occur; a description of how we reduced these fluctuations — which can affect various fluctuation measures described in what follows — is provided in Appendix [A].

B. Observables

In what follows we will denote by \( x_i \) the 3-dimensional coordinate vector of the \( i \)th particle, then \( x_{ij} \equiv x_j - x_i \) is the vector distance between the \( i \)th and \( j \)th particles, and \( r_{ij} \equiv \sqrt{x_{ij}^2} \) is the pairwise distance between them. We also omit the explicit mentioning of dimensional observables' units, for the sake of simplifying our notations; those observables should be understood as expressed in the appropriate microscopic units.

In all computer glasses considered in this work, pairs of particles \((i,j)\) interact via a radially-symmetric pairwise potential \( \varphi_{ij} = \varphi_{ij}(r_{ij}) \), then the potential energy reads

\[
U = \sum_{i<j} \varphi_{ij}.
\]

The (simple shear) stress in athermal glasses is given by

\[
\sigma = \frac{1}{V} \frac{\partial U}{\partial \gamma}.
\]

We also consider the shear and bulk moduli, defined as

\[
G = \frac{\partial^2 U}{\partial \gamma^2} - \frac{\partial^2 U}{\partial x^2} : \mathbf{M}^{-1} : \frac{\partial^2 U}{\partial x^2},
\]

and

\[
K = \frac{\partial^2 U}{\partial x^2} - \frac{\partial^2 U}{\partial x \partial \eta} : \mathbf{M}^{-1} : \frac{\partial^2 U}{\partial x \partial \eta} + p,
\]

respectively, where the pressure \( p \) is given by

\[
p = -\frac{1}{V} \frac{\partial U}{\partial \eta}, \quad \mathbf{M} = \frac{\partial^2 U}{\partial x \partial x}
\]

is the Hessian matrix, and \( \eta, \gamma \) parametrize the strain tensor

\[
\epsilon = \frac{1}{2} \begin{pmatrix}
2\eta + \eta^2 & \gamma + \eta \gamma & 0 \\
\gamma + \eta \gamma & 2\eta + \eta^2 + \gamma^2 & 0 \\
0 & 0 & 2\eta + \eta^2
\end{pmatrix}.
\]
To quantify the effect of nonaffinity on the bulk modulus $K$, we also consider the nonaffine term alone (cf. Eq. (10)), namely

$$K_{na} = \frac{1}{d^2V} \frac{\partial^2 U}{\partial \eta \partial \bar{x}} \cdot M^{-1} \cdot \frac{\partial^2 U}{\partial \bar{x} \partial \eta}.$$

The Poisson’s ratio is given by

$$\nu \equiv \frac{3K - 2G}{6K + 2G} = \frac{3 - 2G/K}{6 + 2G/K}$$

For every model studied in what follows, we also consider an “unstressed” potential energy

$$\mathcal{U} = \frac{1}{2} \sum_{i<j} \phi''_{ij}(r_{ij} - r_{ij}^{(0)})^2,$$

where $\phi''_{ij}$ is the second derivative of the $i,j$ interaction of the original potential, and $r_{ij}^{(0)}$ is the distance between the $i^{th}$ and $j^{th}$ particles in the mechanical equilibrium state $\partial U/\partial \bar{x} = 0$ of the original potential. The potential $\mathcal{U}$ can be understood as obtained by replacing the original interactions by Hookean springs whose stiffnesses are inherited from the original interaction potential, and that reside exactly at their rest lengths $r_{ij}^{(0)}$ so that the springs exert no forces on the particles. The observable we focus on is then the shear modulus $G \equiv V^{-1} \frac{\partial^2 \mathcal{U}}{\partial \eta^2}$ of the unstressed potential.

III. RESULTS

Here we present the various data sets of dimensionless observables that describe the mechanical properties of the glasses of different models and control parameters discussed in the previous Section.

A. Poisson’s ratio

We begin with presenting data for the Poisson’s ratio $\nu$ (see definition in Eq. (13)), which is a conventional dimensionless characterizer of the elastic properties of solids [20, 21], whether glassy [22] or crystalline [23]. Fig. 2(a)-(d) shows the sample-to-sample means of the Poisson’s ratio measured in our ensembles of the model glasses studied. To gain insight on the behavior of $\nu$, we also plot in panels (e)-(h) the ratio $G/K$ (cf. Eq. (13)) of the sample-to-sample means of the shear and bulk moduli.

Fig. 2(a) shows the Poisson’s ratio of the IPL model; we observe an interesting non-monotonic behavior of $\nu$ as a function of the exponent $\beta$ that characterizes the pairwise interaction. A corresponding non-monotonic behavior of the ratio $G/K$ is also observed (Fig. 2b); the decrease of $G/K$ at large $\beta$ is expected: in previous work [24] it
was shown that increasing $\beta$ is akin to approaching the unjamming point of repulsive soft spheres [12-14]. In [24] it is shown that $G/K$ is expected to vanish as $1/\sqrt{3}$, represented in Fig. 2, by the dashed line.

The decrease of $G/K$ at small $\beta$ seen in Fig. 2, that leads in turn to an increase in $\nu$ for small $\beta$, is however unexpected. While the careful investigation of the decrease of $G/K$ for small $\beta$ is left for future investigation, we postulate its origin to lie in the increasing relative importance of higher coordination-shell interactions with decreasing $\beta$.

![Interaction-cutoff-induced density dependence of the Poisson's ratio](image)

FIG. 3. Interaction-cutoff-induced density dependence of the Poisson’s ratio $\nu$ and the shear-to-bulk modulus ratio $G/K$ for the IPL model, see Sect. II A 1 for details. The dashed and continuous horizontal lines represent the values of $\nu$ and $G/K$ for $\beta=8$ and $\beta=6$, respectively, measured in the IPL model with no interaction cutoff. For $\beta\geq 12$ we see no appreciable dependence on density, while for $\beta<12$ a measurable dependence is observed, see text for discussion.

The importance of higher coordination shells for small $\beta$ can be appreciated by considering the density dependence of the Poisson’s ratio $\nu$ and the ratio $G/K$ for the IPL model with a finite interaction cutoff length (see Sect. II A 1), shown in Fig. 3. We note that the inverse-power-law interactions of the IPL model should imply the invariance of any dimensionless numbers to changes of the density [25]. However, upon introducing a cutoff (for computational efficiency) in the pairwise interactions, the invariance becomes only approximate. In particular, for $\beta<12$ a measurable dependence of $G/K$ (and therefore also of $\nu$) is seen over a broad range of densities, indicating the greater importance of higher coordination shells for small $\beta$ in the IPL model. We note that for $\beta=4$ some 10% of the solids we created without cutting off the potential were unstable (their Hessian matrix possesses at least one negative eigenvalue). We therefore use a finite interaction cutoff and fix $\rho=10.0$ to obtain approximations for $\nu$ and $G/K$ for $\beta=4$, which are represented by the open symbols in Fig. 2, i.e.

Fig. 2, shows the Poisson’s ratio $\nu$ measured in the HRTZ system, plotted against the imposed pressure $p$. As $p\to 0$ it appears that the incompressible limit $\nu=1/2$ is approached. As expected, this is a consequence of the aforementioned vanishing of the ratio $G/K$ upon approaching the unjamming point, as indeed seen in Fig. 2.

![Pressure to bulk modulus ratio](image)

FIG. 4. (a) Pressure to bulk modulus ratio vs. the density, for the KABLJ system. (b) The relative fraction of the nonaffine term of the bulk modulus, see text for definitions and discussion.

To better understand these observations in the KABLJ data, we plot in Fig. 4, the ratio of the pressure to bulk modulus of the KABLJ systems, vs. the density. As expected, the pressure decreases with decreasing density, and appears to vanish a bit below $\rho=1.2$ [27]. Accompanying the vanishing of pressure is a substantial increase in nonaffine nature of displacements under compressive strains, which we quantify via the nonaffine contribution to the bulk modulus $K_{\text{na}}$ defined in Eq. 12. Fig. 4, shows that the relative fraction that $K_{\text{na}}$ amounts to in the bulk modulus grows from nearly zero at $\rho \geq 2.0$ to about 13% at $\rho=1.15$. This increase in the nonaffine contribution to the moduli, together with the contribution of the negative pressure (cf. Eq. 10), can explain most of the increase of $G/K$, and the corresponding decrease of the Poisson’s ratio at low densities, in the KABLJ model.

Finally, in Fig. 2l we show the Poisson’s ratio measured in the POLY system, plotted against the equilibrium parent temperature $T_0$ from which the ensembles of glasses were quenched. The annealing at the lowest temperature leads to a decrease of slightly more than 8% in $\nu$. In terms of the ratio $G/K$, we observe an annealing-induced increase of over 55% above the high-$T_0$ plateau. For comparison, in [28] an increase of nearly 20% in $G/K$ was observed by varying the quench rate of a model of a Cu$_{64}$Zr$_{36}$ metallic glass over two orders of magnitude,
with an associated increase of $\approx 3.5\%$ in the Poisson’s ratio, whose typical values were found around $\nu = 0.41$.

We note that typical values for the Poisson’s ratio of metallic glasses ranges between 0.3-0.4 \cite{22, 28}, i.e. mostly lower than what we observe in our simple models, with the exception of the KABLJ model, discussed in length above. We attribute the higher values of $\nu$ seen in our models that feature inverse-power-law pairwise interactions (i.e. the IPL model, and the KABLI at high densities) to the relative smallness of the nonaffine term in the bulk modulus. This relative smallness results in relatively larger bulk moduli (compared to shear moduli), and, in turn, to higher Poisson’s ratios. Laboratory glasses experience a significant degree of annealing upon preparation, which would further reduce their Poisson’s ratio, as suggested by our measurements of the POLY system shown in Fig. 2h.

B. Degree of internal stresses

One of the hallmark features of glasses is their structural frustration. How can the degree of structural frustration of different computer glasses be compared? Here we offer to quantitatively compare different simple computer glasses via the following observable: consider a glassy sample that is comprised of $N$ particles; consider next replacing the fixed-shape box in which the glass is confined by a box that can undergo simple shear deformation, and consider fixing the imposed shear stress (instead of the box shape) at zero. Under these conditions, the internal residual stresses of the glass would lead to some shear deformation $\delta \gamma$ of the box, that can be estimated as $\delta \gamma \approx \sigma / G$, where $\sigma$ is the as-cast shear stress of the original glass. Since $\delta \gamma$ decays with system size $N$ as $1/\sqrt{N}$ (it is $N^{-1}$ times a sum of $O(N)$ random contributions, see Appendix B for numerical validation), we thus form a dimensionless characterization of glassy structural frustration by

$$\delta \tilde{\sigma} \equiv \sqrt{N} \delta \gamma = \sqrt{N} \delta \sigma / G,$$

where $\delta \sigma$ denotes the sample-to-sample standard deviation of the residual stresses.

In Fig. 5 we show $\delta \tilde{\sigma}$ measured in our ensembles of glasses. Interestingly, in the IPL and HRTZ models we see that $\delta \tilde{\sigma}$ tends to decrease upon approaching the unjamming point by increasing $\beta$ (for IPL) or decreasing the pressure (for HRTZ), respectively. In contrast with our observations for e.g. the Poisson’s ratio showed in Fig. 2, no non-monotonic behavior in $\delta \tilde{\sigma}$ is observed in the IPL model. At $\beta \gtrsim 16$ it appears that $\delta \tilde{\sigma} \sim \log \beta$.

The KABLJ and POLY models appear to agree at high densities and high $T_0$, respectively, showing $\delta \tilde{\sigma} \approx 0.22$ in those regimes. The POLY system exhibits a significant reduction of $\delta \tilde{\sigma}$ upon annealing (i.e. for lower $T_0$), up to roughly 40% below the high-$T_0$ plateau value.

C. Shear modulus fluctuations

We next turn to characterizing the degree of mechanical disorder of our simple computer glasses. We propose to quantify the mechanical disorder of a given ensemble of computer glasses by first measuring

$$\Delta G \equiv \sqrt{\text{median}_i [(G_i - G)^2]},$$

where the median is taken over the ensemble of glasses, and $G$ denotes the sample-to-sample mean shear modulus. In Appendix B we demonstrate that, as expected for an intensive variable (and see also \cite{29}), $\Delta G \sim 1/\sqrt{N}$. A dimensionless and $N$-independent quantifier of disorder is therefore given by

$$\Delta \tilde{G} \equiv \sqrt{N} \Delta G / G.$$  \hspace{1cm} (17)

In Fig. 6 we plot $\Delta \tilde{G}$ for our different computer glasses. We find that $\Delta \tilde{G}$ grows substantially in the IPL and HRTZ models as the respective unjamming points are approached, suggesting that $\Delta \tilde{G} \to \infty$ upon approaching unjamming.

While $\Delta G$ remains essentially constant at $\approx 2.5$ over the entire density range in the KABLJ model, in the

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**FIG. 5.** Sample-to-sample standard deviations of the shear stress $\delta \sigma$, scaled by $\sqrt{N}/G$ with $G$ the mean shear modulus.
POLY model we find a very substantial decrease of $\Delta G$ as a function of the parent temperature $T_0$, by over a factor of 3. The noise in our data is quite substantial; we nevertheless speculate based on our data that the variation of $\Delta G$ with harmonic pairwise interactions, and $\bar{G}/\tilde{G}$, in panel (b) we plot with a dotted line the cumulative distribution associated with the Gaussian fit of panel (a), shown for comparison.

The reason we choose to measure the median of fluctuations instead of the considering the more conventional standard deviation is that for small $N$ the distribution $p(G)$ of the shear modulus can feature a large tail at low values. This is demonstrated in Fig. 7, where we show the distribution of shear moduli measured in the IPL model for glasses of $N = 2000$ particles that were instantaneously quenched from high temperature states. Fig. 7 shows that the low-$G$ tail is substantial, leading to a large discrepancy between the full width at half maximum of the distribution of $G$ and its standard deviation. To overcome this discrepancy we opt for a measure which is based on the (square root of the) median of fluctuations rather than their mean. We note however that the large tail of $p(G)$ at low values of $G$ is expected to disappear as the system size is increased [29].

**D. Effect of internal stresses on shear modulus**

We conclude our study of the elastic properties of our computer glasses with presenting and discussion the effect of internal stresses on the shear modulus. To this aim we recall Eq. 14 which defines a modified potential energy $U$, constructed based on the original potential energy $U$ by connecting a relaxed, Hookean spring between all pairs of interacting particles, with stiffnesses $k = \varphi''|_{r_{ij}}$ adopted from the original pairwise potentials $\varphi$. An associated shear modulus $\tilde{G}$ is then defined as $V^{-1} \frac{d^2U}{d\gamma^2}$. In previous work [30] it has been shown using mean field calculations that $G/\tilde{G}$ indicates the distance of a system from an internal-stress-induced elastic instability. It is predicted in [30] that $G/\tilde{G} \approx 1/2$ in marginally-stable states with harmonic pairwise interactions, and $G/\tilde{G} > 1/2$ as glass stability increases. The ratio $G/\tilde{G}$ can also depend on statistical properties of interparticle interactions, as discussed in [31].

In Fig. 8 we show measurements of $G/\tilde{G}$ in our different computer glasses. In the IPL model we find that $G/\tilde{G} < 1/2$ in the entire $\beta$ range, but approaches 1/2 in the large $\beta$ limit at which the system unjams. In the HRTZ system we find $G/\tilde{G} \approx 1/2$ over most of the investigated pressure range, with a slight decrease at high pressures. In [31] we have compared $G/\tilde{G}$ with $G/\tilde{G}$ for a variety of glassy systems, and found that the ratio $G/\tilde{G}$ is an excellent indicator of the distance of a system from an internal-stress-induced elastic instability. In particular, we have found that $G/\tilde{G} \approx 1/2$ in marginally-stable states with harmonic pairwise interactions, and $G/\tilde{G} > 1/2$ as glass stability increases. The ratio $G/\tilde{G}$ can also depend on statistical properties of interparticle interactions, as discussed in [31].

**FIG. 6.** $\Delta \tilde{G} = \sqrt{N} \Delta G/G$ is a $N$-independent dimensionless quantifier of mechanical disorder, defined via Eqs. (16) and (17), and motivated in Sect. [IIIC]. The $p = 10^{-5}$ data point for which $\Delta \tilde{G} = 11.43$ was omitted for visual purposes.

**FIG. 7.** (a) The dotted line represents the probability distribution function $p(G)$ of the shear modulus of 20,000 computer glasses of $N = 2000$ particles of the IPL model with $\beta = 10$, made by a instantaneous quench from a high temperature liquid state. The continuous line is a fit to a Gaussian, that demonstrates the asymmetry of $p(G)$ about its mean. To better quantify the low-value tail of $p(G)$, in panel (b) we plot with a dotted line the cumulative distribution of $\int_{0}^{G'} p(G')dG'$; the continuous line represents the cumulative distribution associated with the Gaussian fit of panel (a), shown for comparison.
pressures. The KABLJ system shows that \( G/\rho \) can attain high values in the low density regime in which attractive interactions become dominant, and, similarly to as we have seen above, at large densities it agrees well with the \( \beta \approx 12 \) result for \( G/\rho \) of the IPL model. Finally, in the POLY system at high \( T_0 \), \( G/\rho \) agrees well with the IPL model for \( \beta \approx 12 \), as expected. Equilibration deep into the supercooled regime increases \( G/\rho \) by nearly 50%, bringing it to \( \approx 1/2 \) at the deepest supercooling.

E. Yield stress

Up until this point we have only discussed various dimensionless characterizations of the elastic properties of our computer glasses. In this last Subsection we present results regarding the simple shear yield stress of a subset of the models we have investigated, measured in athermal quasistatic deformation simulations. In particular, we exclude the POLY model from this analysis; its elastoplastic transient behavior was characterized in detail in [32], and its steady-flow state stress (referred to here as the yield stress) is expected to be independent of the key control parameter of the POLY model – the parent temperature \( T_0 \).

We employ the standard procedure for driving our glasses under athermal quasistatic deformation: the simulations consist of repeatedly applying a simple shear deformation transformation (we use strain steps of \( \Delta \gamma = 10^{-3} \)), followed by a potential energy minimization under Lees-Edwards boundary conditions [33]. As explained in Sect. [14] simulations of the HRTZ model involved embedding a barostat functionality [17] into our minimization algorithm, in order to maintain the pressure approximately constant during the deformation simulations, see further discussion in Appendix C.

In Fig. 8 we present the average yield stress \( \sigma_\infty \), defined here as the average steady-flow stress, taken after the initial elastoplastic transients, rescaled by the isotropic-states average shear modulus \( G \). Each point is obtained by averaging over the steady flow shear stress of 200 independent runs of each computer glass model, and for each control parameter value.

We find that in the IPL and HRTZ models \( \sigma_\infty/\rho \) decreases upon approaching their respective unjamming points \( \beta \to \infty \) and \( p \to 0 \). In the IPL model we observe \( \sigma_\infty/\rho \sim \log \beta \) at large \( \beta \); understanding this behavior is left for future investigations. In the HRTZ model one expects \( \sigma_\infty \sim p \) and \( G \sim p^{1/3} \) (it should scale with pressure similarly to the bulk modulus \( K \) of isotropic, as-cast states, see [34]), then \( \sigma_\infty/\rho \sim p^{2/3} \) is predicted. We cannot however confirm this prediction numerically: we postulate that the pressure range explored is not sufficiently close to the unjamming point in order to observe the asymptotic scaling. Finally, the KABLJ model features \( \sigma_\infty/\rho \cong 0.038 \) over the majority of the explored density range, with a slight increase as attractive forces become more dominant at low densities.

How do these numbers compare to more realistic computer glasses? In [28] values of around \( \sigma_\infty/\rho \cong 0.05 \) were reported for a model Cu$_{40}$Zr$_{60}$ metallic glasses that employs the embedded atom method [35], i.e. some 30% higher than what we find in e.g. the KABLJ model. Similar results were also found by [36] for model Cu$_{50}$Zr$_{50}$ and Cu$_{47.5}$Zr$_{47.5}$Al$_{5}$ metallic glasses. In [37] a value of \( \sigma_\infty/\rho \cong 0.03 \) was observed using the Stillinger-Weber model for amorphous silicon [38]. A value of \( \sigma_\infty/\rho \cong 0.11 \) can be estimated based on the stress-strain signals reported in [39] for computer models of sodium silicate glasses that employ the van Beest-Kramer-van Santen potential [40]. The spread in these values indicates that the simple computer models investigated in this work only represent a narrow class of amorphous solids.

IV. SUMMARY

The goal of this paper is to offer a comprehensive data set that compares — on the same footing — various dimensionless quantities of elastic and elasto-plastic properties of popular computer glass models. We build on the assertion that instantaneously quenching high-energy
FIG. 9. Yield stress $\sigma_\infty$ rescaled by the mean isotropic, as-cast shear modulus $G$. We reiterate that the open symbol in panel (a) represents an approximation obtained using the finite-cutoff variant of the IPL pairwise potential, see Sect. II A 1 for details. We further note that data points for $p=10^{-5}$ in the HRTZ model and $\beta=256$ in the IPL model could not be measured due to numerical convergence difficulties.

configurations to zero temperature defines an ensemble of glassy samples that can be meaningfully compared between different models. We aimed at disentangling the effects on mechanical properties of various features of the interaction potentials that define computer glass models, from those induced by varying external control parameters and preparation protocols. We hope that the various data sets presented in this work, and the dimensionless observables put forward in this work, will be used as a benchmark for future studies, allowing to meaningfully compare the mechanical properties of different computer glass models.

In addition to putting forward our various analyses of mechanical properties of computer glasses, we have also made a few new observations, summarized briefly here: we have identified an interesting nonmonotonicity in the Poisson’s ratio in the IPL model (see Fig. 2), as a function of the exponent $\beta$ of the inverse-power-law interactions. The shear-to-bulk moduli ratio $G/K$ echos this nonmonotonicity: $G/K$ decreases dramatically as $\beta$ is made small, in addition to its expected decrease at large $\beta$ – the limit at which the IPL model experiences an unjamming transition [24]. The decrease of $G/K$ at low $\beta$ indicates the proximity of an elastic instability, which, to the best of our knowledge, has not been addressed in previous literature. The numerical difficulties we encountered in our attempts to create glasses with $\beta=4$ without using a truncated pairwise potential further support that the non-truncated IPL model becomes unstable at low $\beta$.

Importantly, we have shown that the KABLJ model features a Poisson’s ratio that resembles that of laboratory metallic glasses, and, at density of order unity is generally lower than that seen for the purely repulsive and isomorph-invariant [25] IPL model; our study indicates that the increased nonadditivity of the bulk modulus at low pressures plays an important role in determining the Poisson’s ratio in the KABLJ model.

We offered a dimensionless quantifier of internal glassy frustration, $\delta\tilde{\sigma}$, shown to decrease by up to 40% in well annealed glasses compared to poorly annealed glasses. Even more remarkable is the annealing-induced variation in the sample-to-sample relative fluctuations of the shear modulus $\Delta G$ (cf. Eq. (17) and Fig. 6b), that decrease by over a factor of 3 between poorly annealed and well annealed glasses. Finally, an intriguing nonmonotonic behavior of $d(\delta G)/dT_0$ with equilibrium parent temperature $T_0$ was also observed.

An observable inaccesible experimentally but easily measured numerically is the ratio $G/G$ of the shear modulus $G$ to that obtained by removing the internal forces between particles, denoted here and above by $G$. A similar procedure was carried out in previous work in the context of the vibrational spectrum of glasses [30, 41, 42], and for the investigation of the lengthscale associated with the unjamming point [43]. In theoretical work [30, 31] some trends are predicted for $G/G$; however, since it varies both with stability and depends on details of the interaction potential, it usefulness as a characterizer of stability of a computer glass appears to be limited.

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Appendix A: Sample-to-sample realization fluctuations

The POLY model employed in this work considers soft spheres with polydispersed size parameters, which are drawn from a distribution $p(\lambda) \sim \lambda^{-3}$ sampled between $\lambda_{\text{min}}$ and $\lambda_{\text{max}}$ [3], see Sect. II A 4. Following [3], we chose $\lambda_{\text{max}}/\lambda_{\text{min}} = 2.22$; this choice can lead to large fluctuations between the energetic and elastic properties of dif-
different finite-size samples. To demonstrate this, we show in Fig. 10a the distribution of the mean energies (per particle), calculated over 1000 independent equilibrium runs at $T = 0.6$, each run pertaining to a different, independent realization of the particle-size parameters $\lambda_i$ drawn from the same parent distribution $p(\lambda)$, and with $N = 8000$ particles. The mean (over realizations) standard deviation of the energy per particle of individual runs was found to be $\approx 0.01$, whereas the standard deviation (over realizations) of the mean energy per particle is $\approx 0.16$, i.e. much larger than the characteristic energy per particle fluctuations of any given realization of particle-size parameters, for $N = 8000$.

In order to minimize the effects of these finite-size fluctuations, we selected the particular realizations whose mean equilibrium energy deviated from the mean over realizations (measured here to be $\approx 6.114$) by less than 0.5%, and discard of the rest. To test whether this selection protocol has any observable effect on the distribution of particle-size parameters measured only in the selected states. We find no observable effect of discarding of the realizations with too large or too small energies — as described above — on the distribution of particle-size parameters.

![FIG. 10.](a) Distribution of mean energy per particle, measured for 1000 independent realizations equilibrated at $T = 0.6$. (b) Distribution of post-selection particle size parameters, see text for details.]

### Appendix B: System size scaling of fluctuations

In Sections [I][II] and [III] we define two dimensionless measures of elastic properties of glasses: $\delta\sigma \equiv \sqrt{N}\delta\sigma/G$ and $\Delta G \equiv \sqrt{N}\Delta G/G$, respectively, where $\delta\sigma$ denotes the standard deviation of the as-cast shear stress $\sigma$, and $\Delta G$ is a measure of fluctuations that follows the definition given by Eq. (16). To establish that $\delta\sigma$ and $\Delta G$ are independent of system size $N$, in Fig. 11a we plot $\delta\sigma$ vs. system size $N$, and in Fig. 11b we plot $\Delta G$ vs. $N$. The model glass employed is the IPL model with $\beta = 10$ [15]. As asserted, both of these observables depend on system size as $1/\sqrt{N}$, implying the $N$-independence of $\delta\sigma$ and $\Delta G$.

![FIG. 11.](a) Sample-to-sample standard deviations $\delta\sigma$ of the as-cast shear stress, plotted against system size $N$. (b) The measure $\Delta G$ (cf. Eq. (16)) vs. system size $N$. Both measures of fluctuations decay as $1/\sqrt{N}$.

### Appendix C: Athermal quasistatic simulations of the HRTZ model at fixed external pressure

The key control parameter of the HRTZ model is the external pressure $p$; when creating glassy samples of this model, we incorporated a numerical scheme [17] that allows to specify the desired target pressure into our potential energy minimization algorithm. While this scheme does not fix the pressure exactly, it is sufficiently accurate for our purposes. The performance of the fixed pressure protocol in our quasistatic shear simulations can be gleaned from the example signals shown in Fig. 12.

![FIG. 12.](a) Stress vs. strain measured in a quasistatic shear deformation simulation of the HRTZ model at constant external pressure of $p = 10^{-2}$. (b) Pressure vs. strain in the same run shown in (a). Small fluctuations of less than 1% are still observed; our numerical scheme does not fix the pressure exactly, but rather only approximately.
[1] J. Glaser, T. D. Nguyen, J. A. Anderson, P. Lui, F. Spiga, J. A. Millan, D. C. Morse, and S. C. Glotzer, Comput. Phys. Commun. 192, 97 (2015).

[2] N. P. Bailey, T. S. Ingebrigtsen, J. S. Hansen, A. A. Veldhorst, L. Bohling, C. A. Lemarchand, A. E. Olsen, A. K. Bacher, L. Costigliola, U. R. Pedersen, H. Larsen, J. C. Dyre, and T. B. Schroder, SciPost Phys. 3, 038 (2017).

[3] D. Coslovich, M. Ozawa, and W. Kob, Eur. Phys. J. E 41, 62 (2018).

[4] F. Turci, C. P. Royall, and T. Speck, Phys. Rev. X 7, 031028 (2017).

[5] A. Ninarello, L. Berthier, and D. Coslovich, Phys. Rev. X 7, 021039 (2017).

[6] R. Gutiérrez, S. Karmakar, Y. G. Pollack, and I. Procaccia, Europhys. Lett. 111, 56009 (2015).

[7] G. Kapteijns, W. Ji, C. Brito, M. Wyart, and E. Lerner, Phys. Rev. E 99, 012106 (2019).

[8] P. Das, A. D. Parmar, and S. Sastry, arXiv preprint arXiv:1805.12476 (2018).

[9] M. D. Ediger, J. Chem. Phys. 147, 210901 (2017).

[10] S. Singh, M. D. Ediger, and J. J. De Pablo, Nat Mater. 12, 139 (2013).

[11] L. Berthier, P. Charbonneau, E. Flenner, and F. Zamponi, Phys. Rev. Lett. 119, 188002 (2017).

[12] C. S. O’Hern, L. E. Silbert, A. J. Liu, and S. R. Nagel, Phys. Rev. E 68, 011306 (2003).

[13] M. van Hecke, J. Phys.: Condens. Matter 22, 033101 (2010).

[14] A. J. Liu and S. R. Nagel, Annu. Rev. Condens. Matter Phys. 1, 347 (2010).

[15] E. Lerner and E. Bouchbinder, J. Chem. Phys. 148, 214502 (2018).

[16] L. Wang, A. Ninarello, P. Guan, L. Berthier, G. Szamel, and E. Flenner, Nat. Commun. 10, 26 (2019).

[17] H. J. C. Berendsen, J. P. M. Postma, W. F. van Gunsteren, A. DiNola, and J. R. Haak, J. Chem. Phys. 81, 3684 (1984).

[18] E. Bitzek, P. Koskinen, F. Gähler, M. Moseler, and P. Gumbsch, Phys. Rev. Lett. 97, 170201 (2006).

[19] W. Kob and H. C. Andersen, Phys. Rev. E 51, 4626 (1995).

[20] G. N. Greaves, A. L. Greer, R. S. Lakes, and T. Rouxel, Nat. Mater. 10, 823 (2011).

[21] K. K. Saxena, R. Das, and E. P. Calius, Adv. Eng. Mater. 18, 1847 (2016).

[22] W. H. Wang, J. Appl. Phys. 110, 053521 (2011).

[23] R. H. Baughman, J. M. Shacklette, A. A. Zakhidov, and S. Stafström, Nature 392, 362 (1998).

[24] S. Kooij and E. Lerner, Phys. Rev. E 95, 062141 (2017).

[25] J. C. Dyre, J. Phys. Condens. Matter 28, 323001 (2016).

[26] T. B. Schroder, N. Gnan, U. R. Pedersen, N. P. Bailey, and J. C. Dyre, J. Chem. Phys. 134, 164505 (2011).

[27] S. Sastry, Phys. Rev. Lett. 85, 590 (2000).

[28] Y. Cheng, A. Cao, and E. Ma, Acta Mater. 57, 3253 (2009).

[29] H. G. E. Hentschel, S. Karmakar, E. Lerner, and I. Procaccia, Phys. Rev. E 83, 061101 (2011).

[30] E. DeGiuli, A. Laversanne-Finot, G. During, E. Lerner, and M. Wyart, Soft Matter 10, 5628 (2014).

[31] E. DeGiuli, E. Lerner, C. Brito, and M. Wyart, Proc. Natl. Acad. Sci. U.S.A. 111, 17054 (2014).

[32] M. Ozawa, L. Berthier, G. Birolı, A. Rosso, and G. Tarjus, Proc. Natl. Acad. Sci. U.S.A. 115, 6656 (2018).

[33] M. P. Allen and D. J. Tildesley, Computer simulation of liquids (Oxford university press, 1989).

[34] M. Baity-Jesi, C. P. Goodrich, A. J. Liu, S. R. Nagel, and J. P. Sethna, J. Stat. Phys. 167, 735 (2017).

[35] M. S. Daw and M. I. Baskes, Phys. Rev. Lett. 50, 1285 (1983).

[36] B. Wang, L. Luo, E. Guo, Y. Su, M. Wang, R. O. Ritchie, F. Dong, L. Wang, J. Guo, and H. Fu, NPJ Comput. Mater. 4, 41 (2018).

[37] M. J. Demkowicz and A. S. Argon, Phys. Rev. B 72, 245205 (2005).

[38] F. H. Stillinger and T. A. Weber, Phys. Rev. B 31, 5262 (1985).

[39] G. Molnár, P. Ganster, A. Tanguy, E. Barthel, and G. Kerrouche, Acta Mater. 111, 129 (2016).

[40] B. W. van Beest, G. J. Kramer, and R. A. van Santen, Phys. Rev. Lett. 64, 1955 (1990).

[41] E. Lerner and E. Bouchbinder, Phys. Rev. E 97, 032140 (2018).

[42] H. Mizuno, H. Shiba, and A. Ikeda, Proc. Natl. Acad. Sci. U.S.A. 114, 5976 (2017).

[43] E. Lerner, E. DeGiuli, G. During, and M. Wyart, Soft Matter 10, 5085 (2014).