What controls the Poisson’s ratio of highly incompressible metallic glasses?

Edan Lerner*

Institute for Theoretical Physics, University of Amsterdam,
Science Park 904, 1098 XH Amsterdam, the Netherlands

The manner in which metallic glasses fail under external loading is known to correlate well with those glasses’ Poisson’s ratio $\nu$: low-$\nu$ (compressible) glasses typically feature brittle failure patterns with scarce plastic deformation, while high-$\nu$ (incompressible) glasses typically fail in a ductile manner, accompanied by a high degree of plastic deformation and extensive liquid-like flow. Since the technological utility of metallic glasses depends on their ductility, materials scientists have been concerned with fabricating high-$\nu$ glassy alloys. To shed light on the underlying mechanism of high-$\nu$ metallic glasses, we employ computer simulations of a simple glass-forming model with a single tunable parameter that controls the interparticle-potential’s stiffness. We show that the presented model gives rise to ultra high-$\nu$ glasses, reaching $\nu = 0.445$ and thus exceeding the most incompressible laboratory metallic glass. We further show that our higher-$\nu$ computer glasses host relatively softer quasilocalized glassy excitations, and establish relations between their associated localization length, macroscopic elasticity, and mechanical disorder. Finally, we discuss the possible role of the so-called unjamming transition in controlling the elasticity of ultra high-$\nu$ glasses.

Introduction.—The Poisson’s ratio (PR) $\nu$ of a material represents the extent to which an applied uniaxial load results in deformation in the plane perpendicular to the loading axis. In three dimensions, an isotropic material is deemed incompressible if it features $\nu = 1/2$, in which case a uniaxial load will result in an elastic deformation that conserves the material’s volume perfectly [1]. Some soft, disordered materials such as rubber are nearly incompressible, featuring $\nu \approx 1/2$, while e.g. cork is highly compressible, with a PR that is close to zero [2]. In anisotropic, perfect-crystalline materials, the PR can span a much larger range [3], as is also the case for auxetic materials [4] and metamaterials [5].

The PR is an important material parameter; despite being defined based solely on linear-elastic moduli, it can serve as an indicator of a material’s nonlinear, dissipative mechanical response [6, 7], structural relaxation patterns [8, 9], and vibrational properties [10]. For instance, several investigations of the fracture toughness of metallic glasses have pointed out that it is highly correlated with the PR of those materials: metallic glasses featuring higher PRs are observed to undergo a sharp brittle-to-ductile crossover in their nonlinear mechanical response [6, 7, 11–13]. While the significance of the correlation between metallic glasses’ PRs and their failure patterns is still debated [14, 15], understanding the mechanism that controls a material’s PR remains an interesting challenge [16, 17]. In particular, different from polymeric, rubber-like materials in which elasticity is entropic in nature, the elastic properties of metallic glasses should be understood predominantly in terms of the interaction potentials between those materials’ constituent particles, and their associated emergent micromechanics.

Since the robustness of metallic glasses against catastrophic failure is important for their technological utility, designing ductile metallic glasses with high PRs is a burning challenge [18–22]. To date, the highest-PR metallic glass — a platinum-rich alloy — was fabricated by Demetriou et al. and reported upon in Ref. [21]; it features a PR of 0.43, and is therefore referred to in [21] as ‘liquid-like’. In addition, the same material is one of the stiffest metallic glasses recorded to date, boasting a bulk modulus of $K \approx 217$ GPa.

![FIG. 1. Poisson’s ratio (PR) $\nu$ is plotted vs. the parameter $Q$ that controls the interparticle stiffness (see inset) of the computer glass model put forward in this work. The dashed line corresponds to the PR of platinum-rich glasses studied in [21], which is the highest reported for a metallic glass to date. Inset: the interparticle potential employed (see Eq. (1)) for $Q = 10, 20, 40, 80, 160$ and 240.](image)

What microscopic mechanism is responsible for these ultra-high-PR-materials’ nearly incompressible character? One intriguing model of an elastic solid that can be driven to the incompressible-limit is soft-sphere packings [23]. These systems of repulsive, frictionless spheres are typically studied at vanishing pressures, such that they reside close to the unjamming transition [24, 25] – the point at which the relative shear rigidity $G/K \to 0$.

*e.lerner@uva.nl
(with $G$ denoting the shear modulus) under slow decompression, and consequently $\nu \to 1/2$ (and recall that $\nu \equiv (3 - 2G/K)/(6 + 2G/K)$). In this Letter we discuss and test the hypothesis that a similar mechanism that gives rise to incompressibility in soft-sphere packings near the unjamming point is also present in the high-PR glasses of soft-sphere packings studied in Refs. [19–21]. This hypothesis is supported by the high bulk moduli featured by those high-PR metallic glasses; these suggest that introducing stiffer repulsive interaction potentials between glasses’ constituent particles would enhance steric exclusion effects in the spatial organization of those glasses’ microstructures, thus inducing unjamming-like mechanisms, and high emergent PRs of those glasses.

To test this hypothesis, we employ a simple glass forming model whose particles interact via a pairwise potential in which the stiffness associated with the short-range repulsion can be tuned via a single parameter (denoted $Q$), as shown in the inset of Fig. 1. Using this model we create glasses — with finite tensile yield-stresses and at vanishing pressures — that feature ultra-high PRs, higher than the current highest-PR metal glass [21]. We investigate the mechanical disorder in our ultra-high PR glasses and show that they feature an abundance of very soft glassy defects, whose size increases with increasing PR. We then consider a measure of mesoscopic mechanical disorder (see precise definitions below), and establish a previously proposed relation between it and the density of glassy excitations. Finally, we discuss indications supporting that an unjamming-like mechanism is responsible for the ultra-high PRs of our computer glasses, and highlight an intriguing difference between the micromechanics of our ultra-high PR glasses and of soft-sphere packings.

**Model and methods.**—We employ a 50:50 binary mixture of ‘large’ and ‘small’ particles of equal mass $m$ interacting via the pairwise potential

$$\varphi(r_{ij}) = \begin{cases} \left( \frac{\lambda \nu}{r_{ij}} \right)^Q - \left( \frac{\lambda \nu}{r_{ij}} \right)^6 + \tilde{\varphi}_{\text{smooth}}(r_{ij}, Q), & \frac{\lambda \nu}{r_{ij}} < 2 \\ 0, & \frac{\lambda \nu}{r_{ij}} \geq 2 \end{cases}$$

(1)

where $\varepsilon$ represents a microscopic energy scale, $\lambda_{ij} = \lambda$, $1.18\lambda$, or $1.4\lambda$ for ‘small’–’small’, ‘small-large’ or ‘large-large’ interactions, respectively, and $\lambda$ forms the microscopic units of length. The function $\tilde{\varphi}_{\text{smooth}}(r_{ij}, Q)$ (see [26]) ensures that the potential and two derivatives vanish continuously at the dimensionless cutoff distance $r_{ij}/\lambda_{ij} = 2$. $\varphi(r)$ is presented in the inset of Fig. 1 for $Q = 10, 20, 40, 80, 160$ and 240. In order to mimic laboratory glasses at ambient pressures, we chose the dimensionless densities $N/V = 0.777, 0.737, 0.708, 0.694, 0.6802, 0.677$ for the values of $Q$ specified above, respectively, where $N$ denotes the number of particles and $V$ the simulation-box volume. The resulting mean pressure to bulk-modulus ratio $p/K$ of our glasses is on the order of $10^{-2}$ or smaller for all of our glass ensembles. Glasses were prepared by placing particles randomly on an FCC lattice, following by running $NVE$-dynamics for $10\tau$ where $\tau = \lambda\sqrt{m/\varepsilon}$ forms the microscopic units of time. We followed each independent $NVE$-run by an instantaneous quench to zero temperature by means of a standard nonlinear conjugate gradient method, to form a glass. We created about 1000 independent glassy samples of $N = 10976$ for each value of the parameter $Q$ as detailed above.

**Results and discussion.**—In Fig. 1 we show the PRs $\nu$ of our glass ensembles parameterized by $Q$. We find that $\nu$ grows roughly logarithmically with $Q$, reaching $\nu = 0.445$ for the largest $Q = 240$. The dash-dotted line represents the PR of the platinum-rich glasses reported by Demetriou et al. in Ref. [21], which is the largest PR reported to date (to the best of our knowledge) for a metallic glass.

Can the increase of $\nu$ in our model glass upon increasing the parameter $Q$ be understood on firmer grounds? If, for the sake of discussion, we neglect the attractive term in the pairwise potential, one would expect that $p/K \propto 1/Q$, as seen in the unjamming of soft discs (in two dimensions) interacting via a purely repulsive $x⁻Q$ pairwise interaction potential studied in Ref. [27]. Then, since in the unjamming scenario of repulsive spheres $G/K \sim \sqrt{p/K}$ is universally observed [28], we might expect that, in our glasses, $G/K \sim 1/\sqrt{Q}$. We test this expectation in Fig. 2. While the proposed scaling is certainly not satisfied, there is a clear trend in the data indicating that $\sim 1/\sqrt{Q}$ might be the correct asymptotic form for $G/K$ at large $Q$. We note that in Ref. [27] the scaling $G/K \sim 1/\sqrt{Q}$ is only observed for $Q > 200$, a regime not explored here.

![FIG. 2. The ‘unjamming’-based prediction $G/K \sim 1/\sqrt{Q}$ (see [27]) is not followed by our data (yellow symbols), but might be the correct asymptotic form of $G/K$ at large $Q$.](image-url)

Another hallmark of the unjamming transition of soft-sphere packings is the increase in the first peak of the pair correlation function $g(r)$ upon approaching the unjamming point, namely as $G/K \to 0$. Previous work [23] on decompressed soft-sphere packings has established that the first peak grows as $(G/K)^{-2}$; in Fig. 3 we plot the first peak of the pair correlation function measured in our glasses with various values of the parameter $Q$. We find that the first peak of $g(r)$ indeed increases with increasing $Q$ (and decreasing $G/K$). However, as shown in the
Figure 3: Pair correlations $g(r)$ of 'large'-'large' particles in our model glasses for various $Q$-ensembles as specified in the legend, plotted vs. the dimensionless distance $r/a_0$, where $a_0 \equiv (V/N)^{1/3}$ is an interparticle distance. These distributions behave similarly to those measured in soft-sphere packings near unjamming [23]. Inset: the maximum of $g(r)$ increases slightly more quickly than the $\sim (G/K)^{-2}$ scaling seen in soft-sphere unjamming.

Figure 4: Low-frequency vibrational spectra of our glassy ensembles measured for the same $Q$ parameters as described in the main text, and plotted against the rescaled frequency $\omega/\omega_0$, where $\omega_0 \equiv c_s/a_0$ and $c_s$ is the shear-wave speed. The continuous lines represent fits to the universal $\sim \omega^4$ nonphononic spectra seen quite generally in glassy glasses [29]. The prefactors $A_g$ are extracted from these fits, and used to construct a glassy lengthscale $\xi_g \equiv 2\pi a_0 A_g^{1/5} \omega_0$, which is plotted in the inset vs. $G/K$, see text for further discussion.
pertaining to two independent measurements (of $A_k$ and $\chi$) in Fig. 5a, where very good agreement is observed.

$$\omega^2 = k_+ + k_- + f_+ + f_- ,$$

where $k_+, f_+$ ($k_-, f_-$) are the stabilizing (destabilizing) contributions to an excitation’s energy due to interaction stiffnesses and forces, respectively, see precise definitions of these quantities in Ref. [43]. In purely repulsive soft-sphere packings, $f_+ = k_- = 0$, however these terms are generally nonzero once attractive interactions are introduced between the constituent particles.

In Fig. 5b we report the ratio $k_- / f_-$ for soft quasi-localized excitations analyzed using the nonlinear framework discussed in [44], for our various $Q$-ensembles. As mentioned above, we find that, for the highest PR glasses, the destabilizing terms pertaining to negative stiffness, $k_- / f_-$, dominate over the buckling-like terms ($f_-$) in softening the lowest-energy excitations. The signatures of this different softening mechanism on the anatomy of plastic instabilities under external loading [45] is left for future investigations.

*Summary and outlook.—*Inspired by experimental work on platinum-rich glasses [21], in this Letter we raise and test the hypothesis that the mechanism controlling the Poisson’s ratio in highly incompressible glasses is related to the unjamming transition of packings of soft repulsive spheres. To this aim we design a glass forming model — with a finite tensile yield-stress — whose repulsive stiffness can be tuned via a single parameter $Q$, and investigate the model’s elastic and vibrational properties under variations of $Q$. We find that at large $Q$ our glasses feature ultra-high PRs, exceeding the most incompressible laboratory metallic glasses. In addition, we highlight several unjamming-like phenomena observed in our model glasses; in particular, we find an increase in the first peak of the pair-correlation function $g(r)$, an increase of the prefactor $A_k$ of the universal frequency distribution of nonphononic, quasilocalized excitations [32, 33], and an increase in the linear size of those excitations [33], upon increasing the interparticle interaction stiffness (via the parameter $Q$). Finally, we point out an interesting, qualitative difference between our ultra-high PR glasses and soft-sphere packings in terms of the micromechanics of their respective soft, quasilocalized excitations.

Interestingly, not only do the platinum-rich glasses studied in Ref. [21] feature ultra-high PRs, but they also feature very high bulk moduli (of $\approx 217$ GPa), consistent with the mechanism discussed here that involves increasing the stiffness of the pairwise interaction potential of our model systems to obtain high-PR glasses. Future research should focus on more stringent tests of the unjamming-like picture described in this work, and its relevance to ultra-high PR laboratory glasses.

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