Revealing a new scenario of sound damping in amorphous solids by probing glasses with different stabilities

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Understanding the difference between universal low-temperature properties of amorphous and crystalline solids requires an explanation of the stronger damping of long-wavelength phonons in amorphous solid1-3. A longstanding sound attenuation scenario, resulting from a combination of experiments,4,5 theoretical6-10 and simulative11-12 studies, leads to a quartic scaling of the attenuation coefficient with the wavevector, called Rayleigh scattering. Modern computer simulations offer conflicting conclusions regarding the validity of this picture12-14. Here, we simulate glasses prepared over an unprecedentedly broad range of glass stabilities, to perform the first microscopic analysis of sound damping in model glass formers with experimentally relevant preparation protocols. We demonstrate the existence of a new sound damping scenario, manifested in a quadratic dependence of the transverse sound attenuation coefficient on the wavevector over a range of wavevectors that becomes more prominent in more stable glasses. Our results uncover an intimate connection between glass stability and sound damping, and challenge existing theories.

Many theoretical treatments of sound attenuation in low temperature (athermal) amorphous solids predict a quartic scaling of the sound attenuation coefficient on the wavevector. Early arguments, used to explain the plateau in the temperature dependence of the thermal conductivity1, invoked the picture of scattering of sound waves by uncorrelated inhomogeneities that are much smaller than the wavelength, Rayleigh scattering. In several theories, these inhomogeneities have been modeled as local fluctuations of elastic constant10,11, and predict that the sound attenuation coefficient \(\Gamma_L(k) \sim k^4\) \((\lambda = L\) denotes longitudinal waves and \(\lambda = T\) denotes transverse waves) for small wavevector \(k\). This result is consistent with mean-field theories, which make the same prediction12,13. Another theoretical treatment, the soft-potential model, predicts that a quartic scaling regime exists due to phonons interacting with soft modes14.

Experimentally, longitudinal sound attenuation coefficients can be directly obtained from X-ray and light scattering. A compilation of many experimental results1-3,11,12,20 shows that the wavevector dependence of the longitudinal sound attenuation coefficient, \(\Gamma_L(k)\), can be divided into three regimes: (1) \(\Gamma_L(k) \sim k^2\) for low \(k\); (2) \(\Gamma_L(k) \sim k^4\) for an intermediate \(k\) regime; and (3) \(\Gamma_L(k) \sim k^2\) for large \(k\). While the intermediate wavevector quartic and the large wavevector quadratic scalings of the sound attenuation coefficient are well-documented, the small wavevector quadratic dependence was only seen in few experiments15,23. Because these experiments are performed at finite temperature, the latter regime can be ascribed to thermal and anharmonic effects.

Computer simulations offer a conflicting view of these results. Most computer studies address the problem of sound attenuation in the limit of zero temperature, to remove anharmonic effects. To our knowledge, no simulation reproduced the \(\Gamma_L(k) \sim k^2\) scaling observed at small wavevectors in experiment15,21. Regarding the quartic Rayleigh scattering regime, no firm conclusion can be drawn either. By simulating large glasses created by quenching configurations from a mildly supercooled liquid, Gelin et al. report a logarithmic correction to the quartic scaling13, \(\Gamma_L(k) \sim k^4 \ln(k)\). They further invoke the existence of correlated inhomogeneities of the elastic constants22 to rationalise this observation. However, a more recent, larger-scale study13 of harmonic spheres close to their unjamming transition confirms instead Rayleigh scattering. Therefore, the status of sound attenuation, even for simple glass-formers in the zero-temperature harmonic limit, remains unsettled.

All prior simulations studied sound attenuation in glasses with stabilities dramatically distinct from the ones of typical laboratory glasses. This constraint is imposed by the large preparation times required to equilibrate systems close to the experimental glass transition, which cannot be simulated using conventional techniques. In this work, we use an efficient swap Monte-Carlo algorithm23 that was recently developed24 to prepare glasses with a stability comparable to, or even exceeding the stability of experimental glasses. If we quantify the glass stability in terms of a cooling rate, the improvement due to the swap algorithm is equivalent to decreasing the cooling rate by more than 10 orders of magnitude, thus closing the gap between previous computer studies and realistic materials. In previous studies, it was demonstrated that both the low-frequency vibrational properties20 and the mechanical properties27 of these computer glasses dramatically evolve with increasing the glass stability over such a broad range.

We find that changing the glass stability over a broad range fully clarifies the various regimes of sound attenuation, and allows us to discover a novel attenuation regime that was not observed before. Generally, sound attenuation decreases with increasing stability, implying that more stable glasses are also less dissipative solids28. More
importantly, the wavevector dependence of the sound attenuation changes and becomes clearer with stability. For the longitudinal sound attenuation coefficient, with increasing stability we report an emerging wavevector region in which quartic scaling is observed. For the transverse sound attenuation coefficient, we also see a modest intermediate wavevector region with approximately quartic scaling with increasing stability, and a novel small wavevector quadratic regime which is more pronounced for more stable glasses and should be experimentally relevant. Finite temperature effects and anharmonicities will only come in addition to these regimes.

We produce glasses by instantaneously quenching a model glass-former equilibrated over a range of parent temperatures $T_p$, which uniquely control their stability. We study glasses with $T_p$ ranging from well above the onset of supercooling, denoted as $T_p = \infty$, down to $T_p = 0.062$, which is about 60% of the mode-coupling temperature. The parent temperature $T_p = 0.062$ is lower than the estimated experimental glass temperature $T_g \approx 0.072$ for this model and thus these glasses qualify as ultrastable. We note that for our range of system sizes the new sound attenuation regime is clearly visible only for $T_p < T_c$, and, since previous simulations could only equilibrate down to $T_p \gtrsim T_c$, these simulations were not able to observe this new regime.

We calculate the sound attenuation coefficient from two different quantities: the dynamic structure factor of $T = 0$ glasses, which is obtained from the normal mode analysis, $S_L(k,\omega)$, and the time-dependent current correlation function at temperature $T = 10^{-3}$. We find that both routes provide statistically identical results, demonstrating that our results are not affected by anharmonicity. We simulate systems ranging from 48000 particles to $10^6$ particles for $T_p = \infty$, and up to 192000 for $T_p = 0.062$. Details are provided in Methods.

In Fig. 1 we show a set of representative transverse dynamic structure factors $S_T(k,\omega)$ for several wavevectors $k$ calculated for our most stable glasses. For larger $k$, the peak broadens and shifts towards a larger frequency $\omega$. To find the sound attenuation coefficient $\Gamma_L(k)$ and the characteristic frequency $\Omega_L(k)$, we fit $S_L(k,\omega)$ to a damped harmonic oscillator model:

$$S_L(k,\omega) \propto \frac{\Omega_L^2(k)\Gamma_L(k)}{\omega^2 - \Omega_L^2(k)^2 + \omega^2\Gamma_L^2(k)}.$$

Representative fits are shown in Fig. 1 as solid lines.

In Fig. 2 we show the sound attenuation coefficients for our least stable, $T_p = \infty$, and most stable, $T_p = 0.62$, glasses. Three regimes can be distinguished for the $k$ dependence of $\Gamma_L(k)$. At small $k$, $\Gamma_L(k)$ shows a quadratic $k$ dependence irrespective of the glass stability, Fig. 2(a,b). However, the small wavevector quadratic regime extends up to larger $k$ in the more stable glass, but its amplitude is decreased by a factor of about 3, indicating much smaller dissipation.

For larger wavevectors there is a deviation from this quadratic scaling. To quantify this regime where $\Gamma_L(k)$ does not scale quadratically, we use a power-law description, $\Gamma_L(k) \sim k^\alpha$. We find that the glass stability controls the exponent $\alpha$. For $T_p = \infty$ we obtain $\alpha \approx 3.1$, Fig. 2(a,c), while $\alpha \approx 4.0$ for $T_p = 0.62$, Fig. 2(b,d) for both $\Gamma_T(k)$ and $\Gamma_L(k)$. Hence, we do not find evidence of a quartic Rayleigh scattering scaling of the sound attenuation coefficient with wavevector for any range of $k$ in our poorly annealed glasses, but we do find evidence for this regime, for a restricted $k$ range, in the more stable glasses.

To examine the possible presence of the $k^4$ scaling of the sound attenuation coefficient, we plot in Fig. 3
the quantity $\Gamma_L(k)/k^4$ for three representative $T_p$: our least stable glass $T_p = \infty$, a glass of intermediate stability where $T_p = 0.085$, and our most stable glass where $T_p = 0.062$. In this representation a straight line with a negative slope would indicate the $k^4 \ln(k)$ scaling suggested by Gelin et al.\[13,22\] For $\Gamma_T(k)$, Fig. 3(a) we can possibly identify regions that may be described by $\Gamma_T(k) \sim -k^4 \ln(k)$, but this does not provide a good description for a wide range of wavevectors. Instead we observe a distinct plateau, indicating that a purely quartic scaling without logarithmic correction may describe well the system with $T_p = 0.062$. This plateau is shown in the inset of Fig. 3(a) in a linear plot. We emphasize the absence of any finite size effects in those results, as demonstrated by the perfect agreement between the 11 different system sizes shown in Fig. 3. In Fig. 3(b), for our poorly annealed glass it is possible that $\Gamma_L(k)$ is described well by $\Gamma_L(k) \sim -k^4 \ln(k)$ at the smallest wavevectors we can access. However, we find again a distinct plateau for our most stable glass at $T_p = 0.062$, which indicates a purely quartic scaling of the sound attenuation coefficient. This change in the sound attenuation could be interpreted as a significant change in the spatial inhomogeneities of the local elasticity with the stability of the glass.\[13,22]\] It would be interesting to quantify those changes in future work.

To examine the temperature evolution of the crossover to $\Gamma_T(k) \sim k^2$ in the small wavevector regime, we plot $\Gamma_T(k)/k^2$ versus $k$ for the same representative glasses, $T_p = \infty$, $T_p = 0.085$, and $T_p = 0.062$, see Fig. 3(c). For $T_p = \infty$ the crossover to the low $k^2$ scaling may occur for $k \approx 0.1$ only, and thus we need system sizes of at least $N \approx 4 \times 10^6$ particles to convincingly discern this crossover. Since previous simulations examined attenuation for glasses quenched from high temperature configurations, it would indeed be difficult to observe this crossover due to the large system size needed. By re-examining published data from Marruzzo et al.\[11\] we conclude that they were also able to access this crossover for a system of $N = 10^5$ particles, a system ten times larger than our largest system. We reproduce their data in the inset of Fig. 3(c). The characteristic wavevector where this crossover occurs increases with increasing stability, which makes its observation much clearer in more stable glasses. The location of the crossover varies by about a factor of four over our range of stability. This indicates a crossover lengthscale that decreases with increasing stability, but the interpretation of this lengthscale is at present uncertain.

Figure 4 quantifies the evolution of the wavevector dependence of $\Gamma_L(k)$ with stability. Figure 4(a) shows the low-$k$ quadratic coefficient $H_T$ determined by the fits to $\Gamma_T(k) = H_T k^2$. Note that the glass stability increases with decreasing $T_p$.\[22\] The coefficient $H_T$ is almost independent of $T_p$ at high temperatures, down to the mode-coupling temperature $T_c$. The coefficient $H_T$ decreases approximately threefold between $T_c$ and our
lowest $T_p = 0.062$, indicating a significant decrease of the sound attenuation with increasing stability. We also find that $T_c$ separates two different $T_p$ dependencies of the exponent $\alpha$ of the power law, $\Gamma_T(k) \sim k^{\alpha}$, for our intermediate $k$ regime, Fig. 4(b). We find that $\alpha$ is approximately 3.1 until $T_c$, and then increases with increasing stability when the parent temperature decreases from $T_c$ down to $T_p = 0.066$. At the lowest parent temperatures, $T_p = 0.066$ and $T_p = 0.062$, the exponent reaches $\alpha = 4.0$, implying quartic scaling with the wavevector. Because of the modest range of $k$ where this power law holds, this temperature variation is not very significant.

In a previous study, the density of states $g(\omega)$ for the system studied here, we showed that low frequency modes in $g(\omega)$ could be divided into two contributions, $g(\omega) = A_D(T_p)\omega^2 + A_4(T_p)\omega^4$, where the $\omega^2$ term is the Debye contribution originating from extended phonon-like modes and the $\omega^4$ term is due to quasi-localized modes. We found that the shear modulus, which enters into the expression for Debye level $A_D(T_p)$, is approximately $T_p$-independent and then starts increasing as $T_p$ decreases below $T_c$. Also, we found that $A_4(T_p)$ is approximately $T_p$-independent and then starts rapidly decreasing as $T_p$ decreases below $T_c$. Combining the earlier study with the present one, we see a correlation between the decrease in the amplitude of the contribution from low frequency quasi-localized modes to the density of states and the nature of the sound attenuation. Furthermore, we found that the quasi-localized modes become more localized and sparse. If one is to consider the quasi-localized modes as defects, then the decrease of the stability length associated with the crossover from small wavevector $k^2$ scaling correlates with a decrease in the size of the defects in the glass. Further work is needed to transform these strong correlations into firm causal relations.

A theoretical description of the three regimes of sound attenuation reported here will be challenging. There are many descriptions that result in quartic dependence of the sound attenuation coefficient on the wavevector, but a full description of our results requires a crossover to $k^2$ scaling with decreasing wavevector, at least for transverse sound. Furthermore, such a theory would require a description of a stability-dependent intermediate wavevector regime. This stability-dependent intermediate regime could be due to changes of the internal stress, referred to as frustration, as shown in work by Angelani et al. There are two competing theoretical explanations for the quartic scaling of the sound attenuation coefficient. The first assumes that the phonon modes interact with additional soft modes. The second postulates that sound waves are scattered by local elasticity inhomogeneities. Gelin et al. demonstrated that correlations of local elasticity may be an important consideration in two dimensions, and future studies should examine these correlations in three-dimensional glasses. Similar studies of elastic correlations should also be performed upon approach to jamming in order to explain the differences between our results and Mizuno and Ikeda’s results, who did not find evidence of small wavevector $k^2$ scaling of the transverse sound coefficient. In this regime, the nature of the quasi-localised modes is also very different from typical findings in structural glasses.

The small-wavevector $k^2$ scaling of longitudinal sound attenuation coefficient that has been reported in scattering experiments is generally associated with anharmonic or thermally activated processes. Since these processes are not present in the zero-temperature harmonic approximation used in our analysis, the results presented here raise the possibility that at least part of the explanation for the quadratic scaling observed in experiments is not anharmonicity. This may shed some light on the departure from the low wavevector $k^2$ behavior reported by Devos et al. and Masciovecchio et al. Finally, it would be interesting to examine the stability dependence of sound attenuation using ultrastable glasses, now experimentally available via the method of physical vapor deposition to test further our simulational findings.

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I. METHODS

Simulation details

We perform computer simulations using a three-dimensional cubic system composed of polydisperse particles with equal mass $m = 1$. The distribution of particle diameters $\sigma$ follows $P(\sigma) = \frac{1}{\sigma^3}$, where $\sigma \in [0.73, 1.63]$ and $A$ is a normalization factor. The cross-diameter $\sigma_{ij}$ is determined according to a non-additive mixing rule, $\sigma_{ij} = \frac{\sigma_i^2 + \sigma_j^2}{2} (1 - c(\sigma_i - \sigma_j))$ with $c = 0.2$. The particle number $N$ varies between 48000 and 1000000 for glasses at a very high parent temperature, denoted as $T_p = \infty$, and between 48000 and 192000 for glasses with parent temperatures $0.62 \leq T_p \leq 0.120$. For all glasses studied the number density $\rho = N/V = 1.0$. The interaction between two particles $i$ and $j$ is given by the inverse power law potential, $V(r_{ij}) = \left(\frac{\sigma_{ij}}{r_{ij}}\right)^{12} + V_{\text{cut}}(r_{ij})$, when the separation $r_{ij}$ is smaller than the potential cut-off $r_{ij}^c = 1.25\sigma_{ij}$, and zero otherwise. Here, $V_{\text{cut}}(r_{ij}) = c_0 + c_2 \left(\frac{r_{ij}}{\sigma_{ij}}\right)^2 + c_4 \left(\frac{r_{ij}}{\sigma_{ij}}\right)^4$, and the coefficients $c_0$, $c_2$ and $c_4$ are set to guarantee the continuity of $V(r_{ij})$ at $r_{ij}^c$ up to the second derivative.

Zero-temperature glasses are produced by instantaneously quenching supercooled liquids equilibrated through a swap Monte Carlo algorithm at different parent temperatures $T_p$ to their local potential minima using the fast inertia relaxation engine minimization.$^1$ We calculate the normal modes by diagonalizing the Hessian matrix using Intel Math Kernel Library (https://software.intel.com/en-us/mkl/) and ARPACK (http://www.caam.rice.edu/software/ARPACK/).

Damping information

We calculate the $T = 0$ dynamic structure factors using the eigenvalues and eigenvectors of the Hessian matrix,$^2$

$$S_\lambda(k, \omega) = \left(\frac{k^2}{\hbar \omega^2}\right)^3 \sum_{n=1}^{3N-3} F_{n,\lambda}(k) \delta(\omega - \omega_n),$$

where $\lambda$ is $T$ for transverse or $L$ for longitudinal structure factor, $\omega_n$ is the frequency (square root of the eigenvalue) associated with the $n$-th eigenvector, and

$$F_{n,T}(k) = \left| \sum_{j=1}^{N} (\vec{e}_{n,j} \times \hat{k}) e^{i\vec{k} \cdot \vec{r}_j} \right|^2,$$
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Author contributions

L. W., E. F., and G. S. conceived the project. L. W. performed numerical simulations. L. B., E. F., and G. S. directed the project. All authors contributed to the discussion and analysis of the results. L. W., L. B., E. F., and G. S. wrote the manuscript.

Additional information

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