Scaling of the Non-Phononic Spectrum of Two-Dimensional Glasses

Lijin Wang\textsuperscript{1}, Grzegorz Szamel\textsuperscript{2}, and Elijah Flenner\textsuperscript{2}

\textsuperscript{1}School of Physics and Optoelectronic Engineering, Anhui University, Hefei 230601, China and
\textsuperscript{2}Department of Chemistry, Colorado State University, Fort Collins, Colorado 80523, USA

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Low-frequency vibrational harmonic modes of glasses are frequently used to understand their universal low-temperature properties. One well studied feature is the excess low-frequency density of states over the Debye model prediction. Here we examine the system size dependence of the density of states for two-dimensional glasses. For systems of fewer than 100 particles, the density of states scales with the system size as if all the modes were plane-wave-like. However, for systems greater than 100 particles we find a different system-size scaling of the cumulative density of states below the first transverse sound mode frequency, which can be derived from the assumption that these modes are quasi-localized. Moreover, for systems greater than 100 particles, we find that the cumulative density of states scales with frequency as a power law with the exponent that leads to the exponent $\beta = 3.5$ for the density of states independent of system size.

A striking feature of glasses is the excess of low-frequency vibrational harmonic modes over that predicted by Debye theory. Many computational studies analyzed low-frequency vibrational modes in glasses with the goal to characterize the excess harmonic modes, which are frequently referred to as non-phononic modes. Since the same low-temperature properties are found in different glasses, one is naturally tempted to look for universal features of the excess modes, independent of the model and the glass formation protocol. Importantly, these features should hold in the thermodynamic limit. In many studies the implicit hypothesis is that the excess modes density $D_{ex}(\omega)$ follows a power law, $D_{ex}(\omega) = A_{ex}\omega^{\beta}$.

Two methods are generally employed to determine $\beta$. One approach is to separate modes by their participation ratio with the assumption that the low-frequency excess modes have a significantly smaller participation ratio than the extended, plane-wave-like modes \cite{1,2}. This method was used to argue that $\beta = 4$ in three dimensions \cite{4,5}. However, this method fails in two dimensions since it is impossible to choose a suitable participation ratio cutoff to differentiate between the excess modes and the plane-wave-like modes in two dimensions.

The second method is to study small systems \cite{1,4}. The frequency range of the first band of plane-wave-like modes scales as $L^{-1}$ where $L$ is the simulation box size. The assumption is that the modes below this band of plane-wave-modes represent the excess modes and one can determine the scaling of $D_{ex}(\omega)$ from studying the harmonic mode spectrum for frequencies significantly below those of the first band. While conceptually straightforward, there are some subtle issues that must be kept in mind. First, every simulated glass has a slightly different shear modulus that results in a slightly different frequency expected for the first plane-wave-like mode, $\omega_{L1}$, for an ideal elastic body. In addition, the modes are not pure plane waves and there is a distribution of frequencies around $\omega_{L1}$ for each glass. For these reasons one can have a rather large range of frequencies where the first plane-wave-like modes exist. If one does not separate the plane-wave-like modes (like in the first method), one has to be careful to infer $D_{ex}(\omega)$ using only modes that do not correspond to plane-wave-like modes.

Another difficulty is that there is a reported finite size effect influencing the exponent $\beta$ in three-dimensional glasses \cite{4}. It has been reported that $\beta < 4$ for small systems and $\beta$ equals 4 for large enough systems \cite{4}. Recall that the plane-wave-like modes move to lower frequencies with increasing system size, and thus there is a smaller frequency range where the excess modes spectrum is found. In addition, in two dimensions there are very few excess modes \cite{3,7} and a very large number of glass realizations are needed to obtain good enough statistics to determine an accurate value for $\beta$. Analysis of $\beta$ in two dimensions is complicated by a reported finite size effect in the pre-factor $A_{ex}$, $A_{ex} \sim (\log N)^{(\beta+1)/2}$ with $N$ the number of particles in the system \cite{4}.

In a previous publication we stated that there was no clear evidence that there is a finite size effect for $\beta$ in simulations of two-dimensional glasses \cite{7}. In contrast, Lerner and Buchbinder argued very recently \cite{8} that $\beta$ increases with increasing system size and is four for a large enough (N=1600) aged glass.

Here we present analysis of our results of the cumulative density of states $I(\omega) = \int_0^{\omega} D(\omega')d\omega'$, where $D(\omega)$ is the full density of states, that lead to our conclusions that there is no system size dependence of $\beta$ and examine in detail some finite size effects of the density of states observed in two-dimensional glasses. We also argue that there is no clear evidence of systematic system size dependence observed in the data presented Ref. \cite{8}.

For an ideal elastic body, the frequency of the first transverse wave is given by $\omega_{T1} = \sqrt{(G/\rho)(2\pi/L)}$, where $G$ is the shear modulus, $\rho$ is the mass density, and $L$ is the linear size of the sample. The frequency of the first longitudinal wave is given by $\omega_{L1} = \sqrt{(G+K)/\rho(2\pi/L)}$, where $K$ is the bulk modulus. We start our analysis of the IPL-10 system discussed in Ref. \cite{7} for systems of $N \leq 100$. Shown in Fig. \cite{7} is the density of states $D(\omega) = (2N-2)^{-1} \sum_n \delta(\omega - \omega_n)$ for $N = 36$, 64, and 100. The frequencies $\omega_n$ are obtained by diagonalizing the Hessian
For these system sizes there are two distinct features at low frequencies, a shoulder and a peak. In Fig. 1(b) we scale the frequency by $\sqrt{N}$ and find that the two low frequency features align. The vertical line is the frequency $\omega_{T1}$ expected for an ideal transverse wave. For an ideal amorphous elastic body, $I(\omega)(2N-2) = 4$ at $\omega_{T1}$. The dotted green line is a fit to the $N = 36$ particle results and the dashed green line is a fit to the $N = 64$ particle results; the corresponding $\beta$ in the fitting function $I(\omega)(2N-2) \sim (\omega \sqrt{N})^{\beta+1}$ is indicated in the legend. Both fits are from $3 \leq \omega \sqrt{N} \leq 6$.

We examined the scaling of $I(\omega)$ for very small systems of $N \leq 100$ particles. Shown in Fig. 2(a) is $I(\omega)$ versus $\omega \sqrt{\log N}$, which is a scaling predicted from continuum elasticity due to a $r^{-1}$ decay of the non-phononic modes at large distances $r$. There is a systematic deviation from this scaling. In contrast, we find nearly perfect data collapse of $I(\omega)(2N-2)$ versus $\omega \sqrt{N}$, Fig. 2(b), which is a scaling motivated by the system size dependence of Debye theory. We are counting the average number of modes up to $\omega \sqrt{N}$ and seeing how many fall below $\omega_{T1}$. For an ideal elastic body, $I(\omega)(2N-2) = 4$ at $\omega_{T1} \sqrt{N}$ (vertical red line). For these very small systems we do not find any evidence of excess modes and do not believe that the excess mode scaling can be inferred from systems less than 100 particles. This analysis also highlights the difficulty in determining the frequency range to use to study excess (non-phononic) modes.

We fit $I(\omega)(2N-2)$ versus $\omega \sqrt{N}$ for $3 \leq \omega \sqrt{N} \leq 6$ for $N = 36, 64$ and 100 to $a(\omega \sqrt{N})^{\beta+1}$. We find that $\beta$ varies from 3.38 for $N = 36$ to 3.61 for $N = 64$, and that $\beta = 3.48$ for $N = 100$. We show the fits for $N = 36$ and $N = 64$ in Fig. 2(b). It is impossible to determine from the fits if one value of $\beta$ should be preferred over other.
that \( \beta \geq 3 \) is consistent with all our results for systems over 100 particles.

Shown in Fig. 4 is \( I(\omega)/\omega^{4.5} \) (corresponding to \( \beta = 3.5 \)) for the IPL-10 system for different system sizes discussed in Ref. [7]. The low frequency data suggests that \( \beta \approx 3.5 \) for every system size except for \( N = 786 \). As pointed out in Ref. [8], the \( N = 786 \) particle result shows an upturn at small frequencies suggesting a \( \beta \) different than the other system sizes. We find that this system size for the IPL-10 system is an outlier and it does not provide sufficient evidence of a finite size effect. As we show below in Fig. 6 with improved statistics we find that \( \beta \) for \( N = 786 \) is consistent with our results for other systems and other system sizes for the same system.

To determine the exponent \( \beta \) for systems of more than 100 particles we fit \( \log[I(\omega)] = (\beta + 1) \log(\omega) + a \). For each system size we fit three to four frequency ranges to obtain \( \beta \) and average \( \beta \) obtained from these fits. The uncertainty is chosen to include each value of \( \beta \) for a given system and system size. We exclude the results for the Lennard-Jones systems presented in Ref. [7] since there is a systematic downturn at small \( \omega \) for each system size that is not seen for the other systems. Shown in Fig. 5 are the results for the remaining systems analyzed in Ref. [7] (closed symbols).

Figure 5 does not show any clear, systematic system size dependence of the exponent \( \beta \). For the harmonic sphere system (HARM), \( \beta \) does appear to grow with system size, but the growth is within the uncertainty of the calculation. The two inverse-power-law systems (IPL-10 and IPL-12) show no systematic change in \( \beta \) with system size.

For the data presented in Fig. 4 there is one outlier, and that is the IPL-10 system with 786 particles, see also Fig. 4. We increased the number of glass samples for this system and system size from the 0.79 million samples used in Ref. [7] to 2.4 million samples. Shown in Fig. 6 is \( I(\omega)/\omega^{4.5} \) for \( N = 786 \) with the improved statistics.

We can see that the upturn starts at lower \( \omega \), where the statistics are poor, and there is a larger range of \( \omega \) where
\( \beta \approx 3.5 \) is a reasonable description of the data. We redid the fitting for \( \beta \) for this system and the result is shown in Fig. 5 as the open circle. With the improved statistics \( \beta \) is consistent with the other results.

The change of \( \beta \) with increasing the number of glass samples highlights the difficulty of obtaining an accurate value of \( \beta \) for two-dimensional systems. There are so few excess modes that many glass samples are needed to obtain accurate results to do a direct calculation of the density of states. This was pointed out in Ref. [11] where they utilized non-linear mode analysis [9, 12] to analyze two-dimensional systems.

We find that all systems where \( N > 100 \) result in \( \beta \approx 3.5 \) for all our systems and we cannot rule out \( \beta \approx 3.5 \) for \( N \leq 100 \) with our data alone. We note that we do not believe that one can obtain the scaling of the non-phononic density of states for \( N < 100 \). We also find that these conclusions are consistent with the data presented in Ref. [8]. To argue for a finite size effect the authors of Ref. [8] fit the density of states, as opposed to the cumulative density of states, and obtained different values of \( \beta \) for the power law \( D_{ex}(\omega) = A_{ex}\omega^\beta \). However, when the density of states is rescaled there is very convincing data collapse, which suggests that all the results correspond to the same function and \( \beta \) is the same for each system size. To see convincing evidence of a system size dependence of \( \beta \) one would need to see deviations from the data collapse at low frequencies where we expect that the plane-wave-like modes do not contribute to the density of states, and these deviations are not present in the density of states presented in Ref. [8].

We also don't find clear evidence for any system size dependence of \( \beta \) in the cumulative density of states shown in Fig. 2 of Ref. [8]. We argued above that the scaling of the non-phononic density of states cannot be inferred from systems of 100 particles or less. If we focus on the systems of greater than 100 particles we find that Fig. 2(d) of Ref. [8] shows that \( \beta \) is the same for \( N = 196 \) and \( N = 1600 \) poorly annealed system. According to Fig. 1(c) of Ref. [8] \( \beta \approx 3.6 \) for these systems. This value of \( \beta \) is consistent with our results as well as the low frequency scaling of the cumulative density of states for the \( N = 196 \) and \( N = 1600 \) well-annealed system of Ref. [8].

All the systems discussed in this note and in Ref. [8] are fairly small, and it is possible to study much larger systems. The difficulty with larger systems is that the first plane-wave-like modes get pushed to lower frequencies and the frequency range where only excess modes are expected to be found becomes smaller. This difficulty is combined with the very limited number of non-plane-wave-like modes in two-dimensions. We examined a 30 000 particle system for the same IPL-10 model as used in Ref. [9] and found inconclusive results for the value of \( \beta \) at the lowest frequencies examined, see Fig. 3(b). These results were similar to those for three-dimensional systems in Ref. [13] in that \( \beta \) was smaller than 3.5 at lower frequencies. This puzzling result persisted despite having about 0.23 million configurations of 30 000 particles. Examination of these large systems deserves more work.

Non-linear excitation analysis has been used as a proxy to measure exponent \( \beta \) in two-dimensional glasses [9–12]. Many properties of glasses are determined by the harmonic modes [14–17], and thus it is important to understand how the specific features of the harmonic modes influence these properties. We believe it would be ideal to directly measure \( \beta \) using harmonic modes, but find that this is a very difficult task in two dimensions. The results presented here strongly suggest that a harmonic modes-based calculation of \( \beta \) for modes below the first sound mode results in \( \beta \approx 3.5 \) rather than \( \beta = 4.0 \) for two-dimensional glasses.

Two-dimensional solids and liquids have characteristics different than their three-dimensional counterparts [7–9]. One characteristic for two-dimensional glasses is the very small density of excess modes, which makes studying the excess modes difficult. There is evidence that the density of the excess modes scales as \( \omega^{2} \) for excess modes above \( \omega_{T1} \) for two-dimensional systems [7]. Since
$\omega T_1 \to 0$ in the thermodynamic limit, it is unclear that the scaling of the modes below $\omega T_1$ is important in the understanding of the behavior of two-dimensional glasses. Future work needs to focus on the influence of excess harmonic modes on the low-temperature, thermodynamic properties of glasses.

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