# Statistics and Properties of Low-Frequency Vibrational Modes in Structural Glasses

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# Statistics and properties of low-frequency vibrational modes in structural glasses – supplemental material

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In this supplemental material we ($i$) describe the models and numerical methods used to obtain the results presented in the manuscript, and ($ii$) provide a short discussion regarding the statistics of minimal vibrational frequencies in our ensemble of slowly quenched solids.

## Numerical methods

We employ three popular glass forming models in three dimensions:

1. (HARM) A binary mixture of 'large' and 'small' soft spheres of equal mass $m$ interacting via a one-sided harmonic radially-symmetric pairwise potential of the form

$$\varphi_{\text{HARM}}(r_{ij}) = \left\{ \begin{array}{ll} \frac{1}{2}k\big(r_{ij} - (\sigma_i + \sigma_j)\big)^2 & , \quad r_{ij} \leq \sigma_i + \sigma_j \\ 0 & , \quad r_{ij} > \sigma_i + \sigma_j \end{array} \right. , \tag{1}$$

where $r_{ij}$ is the distance between the centers of the $i^{\text{th}}$ and $j^{\text{th}}$ spheres, $k$ is a stiffness constant, and $\sigma_i$ denotes the radius of the $i^{\text{th}}$ sphere. We used a 50:50 binary mixture, where half the particles have a radius of $0.5a_0$ and the other half of $0.7a_0$. The microscopic unit of length $a_0$ was chosen as the diameter of the small particles. $m$ denotes the units of mass, energies are expressed in units of $ka_0^2$, temperatures in units of $ka_0^2/k_B$ (with $k_B$ being the Boltzmann constant), pressure in units of $k/a_0$ and time in units of $\sqrt{m}/(ka_0)$. We prepared packings with a pressure $p = 10^{-1}$ and zero temperature by first equilibrating systems at the density $N/V = 1.0$ and temperature $T = 0.5$ for 100.0 microscopic time units, followed by an energy minimization using a combination of the FIRE algorithm [1] coupled to a Berensden barostat [2].

2. (KABLJ) The canonical Kob-Andersen binary Lennard-Jones system [4] is a binary mixture of 80% type A particles and 20% type B particles, interacting via the following radially-symmetric pairwise potential

$$\varphi_{\text{LJ}}(r_{ij}) = \left\{ \begin{array}{ll} \varepsilon_{ij} \left[ \left(\frac{\sigma_{ij}}{r_{ij}}\right)^{12} - \left(\frac{\sigma_{ij}}{r_{ij}}\right)^{6} + c_6 \left(\frac{r_{ij}}{\sigma_{ij}}\right)^{6} + c_4 \left(\frac{r_{ij}}{\sigma_{ij}}\right)^{4} + c_2 \left(\frac{r_{ij}}{\sigma_{ij}}\right)^{2} + c_0 \right] & , \quad \frac{r_{ij}}{\sigma_{ij}} \leq x_c \\ 0 & , \quad \frac{r_{ij}}{\sigma_{ij}} > x_c \end{array} \right. . \tag{2}$$

Energies are expressed in terms of $\varepsilon_{AA}$, then $\varepsilon_{AB} = 1.5$ and $\varepsilon_{BB} = 0.5$. The interaction length parameters are expressed in terms of $a_0 \equiv \sigma_{AA}$, then $\sigma_{AB} = 0.8$ and $\sigma_{BB} = 0.88$. $x_c = 2.5$ is the dimensionless distance for which $\varphi_{\text{LJ}}$ vanishes continuously up to 3 derivatives, and the density was set at $N/V = 1.2$. Temperature is expressed in terms of $\varepsilon_{AA}/k_B$ with $k_B$ the Boltzmann constant. Time is expressed in terms of $\sqrt{ma_0^2/\varepsilon_{AA}}$, with $m$ denoting the microscopic units of mass. With this parameter set the system experienced a computer glass transition at $T_g \approx 0.45$. Solids were prepared by equilibrating systems at $T = 1.0$ for 50.0 time units, followed by a rapid quench to $T = 0$ by means of a conventional conjugate gradient algorithm.

3. (3DIPL) A 50:50 binary mixture of 'large' and 'small' particles of equal mass $m$, interacting via radially-symmetric purely repulsive inverse power-law pairwise potentials, that follow

$$\varphi_{\text{IPL}}(r_{ij}) = \left\{ \begin{array}{ll} \varepsilon \left[ \left(\frac{\sigma_{ij}}{r_{ij}}\right)^{n} + \sum_{\ell=0}^{q} c_{2\ell} \left(\frac{r_{ij}}{\sigma_{ij}}\right)^{2\ell} \right] & , \quad \frac{r_{ij}}{\sigma_{ij}} \leq x_c \\ 0 & , \quad \frac{r_{ij}}{\sigma_{ij}} > x_c \end{array} \right. , \tag{3}$$

where $r_{ij}$ is the distance between the $i^{\text{th}}$ and $j^{\text{th}}$ particles, $\varepsilon$ is an energy scale, and $x_c$ is the dimensionless distance for which $\varphi_{\text{IPL}}$ vanishes continuously up to $q$ derivatives. Distances are measured in terms of the interaction lengthscale $a_0$ between two 'small' particles, and the rest are chosen to be $\sigma_{ij} = 1.18a_0$ for one 'small' and one 'large' particle, and $\sigma_{ij} = 1.4a_0$ for two 'large' particles. The coefficients $c_{2\ell}$ are given by

$$c_{2\ell} = \frac{(-1)^{\ell+1}}{(2q-2\ell)!!(2\ell)!!}\frac{(n+2q)!!}{(n-2)!!(n+2\ell)}x_c^{-(n+2\ell)} . \tag{4}$$

We chose the parameters $x_c = 1.48, n = 10$, and $q = 3$. The density was set to be $N/V = 0.82a_0^{-3}$. Temperatures are expressed in terms of $\varepsilon/k_B$ with $k_B$ the Boltzmann constant, and time in terms of $\sqrt{ma_0^2/\varepsilon}$, with $m$ denoting the microscopic units of mass. This system undergoes a computer glass transition at $T_g \approx 0.5$. Solids were created by first equilibrating system at $T = 1.0$, followed by a rapid quench from the melt to zero temperature by means of conjugate gradient. We have also created an ensemble of slowly quenched solids (see data and discussion in main text), cooled at a rate of $10^{-5}$ through the glass transition.

The stopping condition for our minimizations was set as follows; we calculate a characteristic interaction force scale $\bar{f} \equiv \left(\sum_\alpha f_\alpha^2/N\right)^{1/2}$ and a characteristic net force scale $\bar{F} \equiv \left(\sum_i |\vec{F}_i|^2/N\right)^{1/2}$, where $\alpha$ labels a pair of interacting particles, $f_\alpha \equiv -\frac{\partial\varphi}{\partial r_\alpha}$ is the force exerted between the $\alpha^{\text{th}}$ pair, $\vec{F}_i \equiv -\frac{\partial U}{\partial \vec{x}_i}$ is the net force experienced by the $i^{\text{th}}$ particle, and $N$ is the number of particles in the sample. We then terminate the minimization algorithm once the ratio $\bar{F}/\bar{f}$ drops below $10^{-10}$.

Normal modes were calculated both using Matlab [5], and following the methods presented in [6]. We have validated by comparison of the two methods and resorting to 128-bit precision that our analysis does not suffer from numerical inaccuracies.

We finally explain here how the spatial decay profile as shown in Fig. 4 of the main text was calculated. Given a mode $\hat{\Psi}$, we identify the mode's core as explained in [3]. We then calculate the median of the square of $\hat{\Psi}$'s components over a thin spherical shell, with thickness on the order of $a_0$, and of radius $r$ away from the mode's core. The decay profiles are defined as the square root of these medians.

## Statistics of low frequency modes in slowly quenched samples

In panel (a) of the figure below we show the direct calculation of the density of states $D(\omega)$ for the slowly-quenched 3DIPL systems with $N = 2000$. In panel (b) we show the distributions $P(\omega_{\min})$ of minimal vibrational frequencies calculated for the ensemble of slowly-quenched solids. In panel (c) we plot the same distributions, but this time as a function of the rescaled minimal frequencies $\omega_{\min}L^{3/5}$. The continuous magenta lines correspond to the Weibull distribution $W(y) \propto y^4 e^{-(y/y_0)^5}$, with $y_0 \approx 5.4$. The analysis is restricted to small systems due to poor statistics for larger systems. It is clear that the rescaling of the minimal vibrational frequencies by $L^{-3/5}$ leads to a very good collapse of the distributions. All aformationed data indicates that the $\omega^4$ law persists under a careful quench of structural glasses.
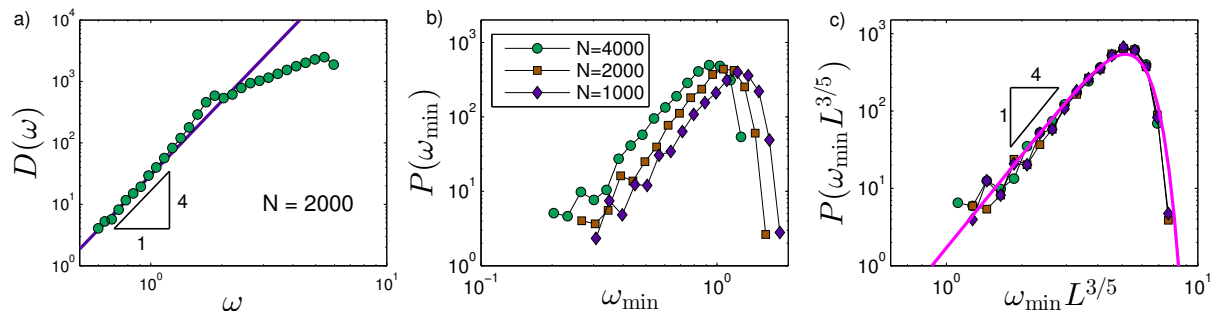
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