


Universality of the Nonphononic Vibrational Spectrum across Different Classes of Computer Glasses

David Richard^{1,*}, Karina González-López^{1,*}, Geert Kapteijns,¹ Robert Pater¹, Talya Vaknin², Eran Bouchbinder² and Edan Lerner¹

¹*Institute for Theoretical Physics, University of Amsterdam, Science Park 904, 1098 XH Amsterdam, The Netherlands*

²*Chemical and Biological Physics Department, Weizmann Institute of Science, Rehovot 7610001, Israel*

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It has been recently established that the low-frequency spectrum of simple computer glass models is populated by soft, quasilocalized nonphononic vibrational modes whose frequencies ω follow a gapless, universal distribution $\mathcal{D}(\omega) \sim \omega^4$. While this universal nonphononic spectrum has been shown to be robust to varying the glass history and spatial dimension, it has so far only been observed in simple computer glasses featuring radially symmetric, pairwise interaction potentials. Consequently, the relevance of the universality of nonphononic spectra seen in simple computer glasses to realistic laboratory glasses remains unclear. Here, we demonstrate the emergence of the universal ω^4 nonphononic spectrum in a broad variety of realistic computer glass models, ranging from tetrahedral network glasses with three-body interactions, through molecular glasses and glassy polymers, to bulk metallic glasses. Taken together with previous observations, our results indicate that the low-frequency nonphononic vibrational spectrum of any glassy solid quenched from a melt features the universal ω^4 law, independently of the nature of its microscopic interactions.

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Introduction.—It is common in condensed matter physics that dynamic and thermodynamic phenomena are controlled by low-energy excitations [1,2]. For example, in crystalline solids, phonon-phonon interactions control wave attenuation rates and heat transport [3]; dislocations (i.e., low-energy topological defects) mediate plastic deformation rates upon external mechanical loading [4]; the specific heat grows as the third power of temperature due to the $\sim\omega^2$ Debye distribution of phonon frequencies. The same principle is also seen to hold in glassy solids, in which soft two-level systems, and their interactions with phonons, are believed to control thermodynamic and transport properties below 10 K [5–7], and low-energy, quasilocalized excitations—often referred to as shear transformation zones [8]—govern elastoplastic responses [9]. Consequently, the complete understanding of the statistical mechanics of soft excitations in solids, and in particular in glasses, is of key importance.

Indeed, much attention has been devoted in the past few decades to understanding the low-frequency spectra of glassy solids [10–28]. It is now well accepted that soft, quasilocalized modes dwell at vanishing frequencies $\omega \rightarrow 0$ in simple computer glasses. These nonphononic excitations were shown to universally feature a disordered core of linear size of about ten particle diameters [25] (see examples in Fig. 1), decorated with algebraically decaying (mostly affine) displacement fields of magnitude $\sim r^{-(d-1)}$ at distance r away from the core, in d spatial dimensions. The

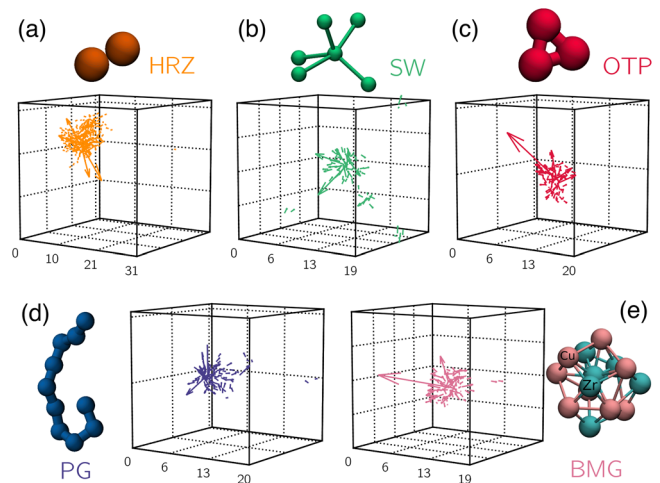


FIG. 1. In this work we study five realistic glass-forming models, each representing a different class of disordered solids, as illustrated by the cartoons. Visualizations of quasilocalized modes found in the employed models of (a) an elastic-spheres glass, (b) a network glass, (c) a molecular glass, (d) a polymer glass, and (e) a bulk metallic glass. For visualization purposes, only the largest 1% of components are shown.

frequencies associated with these excitations were shown to follow a universal distribution $\mathcal{D}(\omega) \sim \omega^4$ [22], independent of spatial dimension [23] or depth of supercooling prior to glass formation [24,25,27]. While these numerical observations are supported by various theoretical frameworks

[10,11], their relevance for laboratory glasses has not been well established; to the best of our knowledge, all computational investigations of the asymptotic functional form of low-frequency nonphononic spectra to date (with the exception of [29] put forward in parallel to this work) employed simple computer glass models, in which particles interact via radially symmetric, pairwise potentials.

In this work we create *in silico* ensembles of polymeric glasses, tetrahedral network glasses, elastic-sphere glasses, molecular glasses, and bulk metallic glasses (BMGs), which are considerably more realistic representatives of laboratory glasses compared to the simple computer-glass models investigated previously, in order to test whether the universal nonphononic spectrum observed in simple computer glasses remains relevant to laboratory glasses as well. Our main finding is that these realistic glass-forming models also feature the universal $\mathcal{D}(\omega) \sim \omega^4$ nonphononic vibrational density of states (VDOS), as seen in simple computer glasses. We thus extend the degree of universality of the ω^4 law, and lend substantial support to the assertion that any glass formed by quenching a melt—and, in particular, laboratory glasses—would feature the gapless ω^4 nonphononic VDOS.

Computer glass models.—We employ five computer glass models, each representing a different *class* of glassy solids. Here, we briefly review the employed models, keeping a complete description for [30]. Throughout this work we express frequencies in terms of c_s/a_0 , where c_s is the shear wave speed, and a_0 is the typical interparticle distance, both are precisely defined in [30].

The employed models are as follows: (1) An elastic-spheres glass model in which spherical particles interact via the linear-elastic Hertz contact law [31]. At low confining pressures, this model undergoes an unjamming transition [48–50]. We refer to this model as HRZ. (2) The Stillinger-Weber network glass model [32], which employs a three-body term in the potential energy that favors tetrahedral local structures. In some range of its parameters, this model mimics the behavior of amorphous silicon [51]. We refer to this model as SW. (3) A triatomic molecular glass model inspired by glass-forming models of orthoterphenyl [43,52], referred to in what follows as OTP. (4) A polymer-glass model of soft beads connected by finite extensible nonlinear elastic nonlinear springs [53], referred to in what follows as PG. Monomers between different polymers interact with a Lennard-Jones-like potential [33]. (5) A binary bulk metallic glass (BMG) alloy composed of Copper (Cu) and Zirconium (Zr) atoms according to $\text{Cu}_{46}\text{Zr}_{54}$ [34,35]. The interactions are calculated using the embedded-atom method (EAM), which gives rise to a spherically symmetric, many-body potential.

Detailed descriptions about how ensembles of glassy samples were created for each computer glass model are provided in [30]. Briefly described, we generate uncorrelated equilibrium configurations at temperatures much

larger than T_g , and perform an energy minimization on those configurations to obtain zero-temperature glassy solids.

For each generated glassy sample, we perform a normal mode analysis, which follows from a generalized eigenvalue problem: eigenvectors $\boldsymbol{\psi}$ and eigenfrequencies ω satisfy the equation

$$\sum_j \mathcal{M}_{ij} \cdot \boldsymbol{\psi}_j = m_i \omega^2 \boldsymbol{\psi}_i. \quad (1)$$

Here, m_i denotes the mass of the i th particle, the Hessian matrix reads $\mathcal{M}_{ij} \equiv (\partial^2 U / \partial \mathbf{x}_i \partial \mathbf{x}_j)$, where U denotes the potential energy and \mathbf{x}_i is the d -dimensional coordinate vector of the i th particle, and $\boldsymbol{\psi}_j$ is the d -dimensional Cartesian displacement vector of the j th particle. Note that no summation over i is implied on the right-hand side of Eq. (1). To obtain the eigenvectors $\boldsymbol{\psi}$ and eigenfrequencies ω , we solve the auxiliary eigenvalue problem

$$\sum_j \frac{\mathcal{M}_{ij}}{\sqrt{m_i m_j}} \cdot \phi_j = \omega^2 \phi_i, \quad (2)$$

where $\boldsymbol{\psi}_i = \phi_i / \sqrt{m_i}$. Details about the calculation of \mathcal{M} for the SW and BMG systems are provided at length in [30]. The system and ensemble sizes in our simulations were selected such that the lowest-frequency modes appear below the first phononic band, as explained in detail in [22,54].

Results.—Our key result is displayed in Figs. 2(a)–2(e), where we show the low-frequency regime of the VDOS of all simulated computer glasses. All models feature the universal form $\mathcal{D}(\omega) \sim \omega^4$, despite the stark qualitative differences between the microscopic interaction laws that define each model.

A quantitative comparison of the localization properties of quasilocated modes between our various computer models is made possible by studying those modes' participation ratio

$$e \equiv \frac{(\sum_i \boldsymbol{\psi}_i \cdot \boldsymbol{\psi}_i)^2}{N \sum_i (\boldsymbol{\psi}_i \cdot \boldsymbol{\psi}_i)^2}, \quad (3)$$

where $\boldsymbol{\psi}_i$ denotes the d -dimensional vector of a mode's Cartesian components pertaining to the i th particle. The participation ratio is expected to scale as $1/N$ for localized modes [55], and should be of order unity for extended modes (e.g., phonons). The product Ne is thus expected to reflect the core size of quasilocated modes, expressed in terms of the characteristic volume occupied by a single particle.

In Figs. 2(f)–2(j) we show the mean participation ratio \bar{e} of vibrational modes, scaled by system size N , binned over and plotted against frequency for all employed computer glasses. The first phonon band frequency $2\pi c_s/L$ is

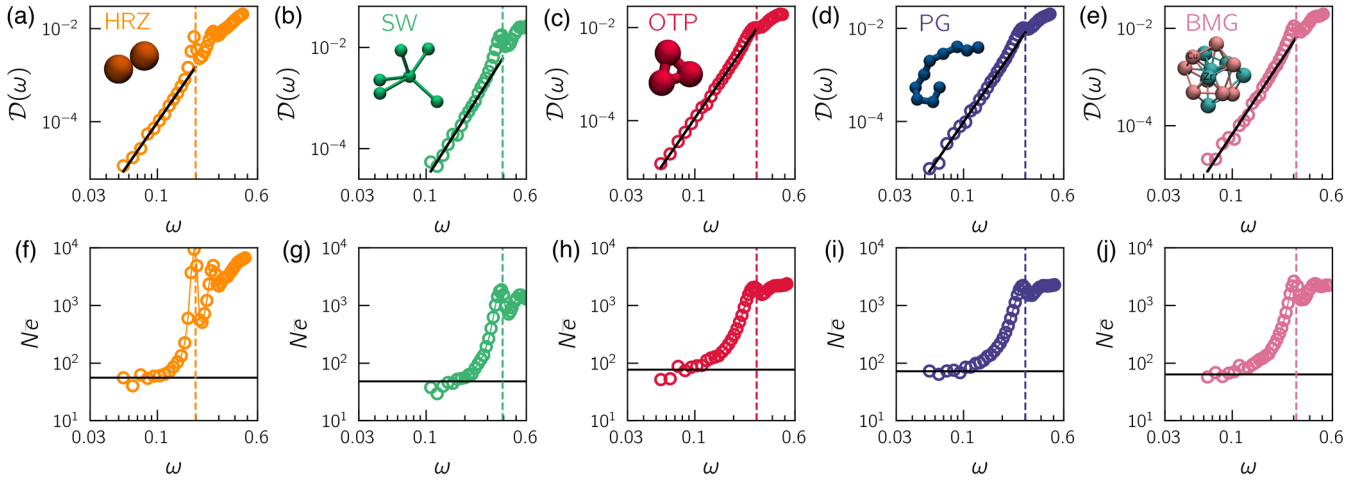


FIG. 2. Low-frequency vibrational modes' spectra and localization properties, measured in realistic computer glass models. We show $\mathcal{D}(\omega)$ vs frequency ω for a linear-elastic-spheres glass [HRZ, panel (a)], a network glass [SW, panel (b)], a molecular glass [OTP, panel (c)], a polymer glass [PG, panel (d)], and a bulk metallic glass [BMG, panel (e)]. The solid lines indicate $\mathcal{D}(\omega) \sim \omega^4$. Panels (f)–(j) show the average participation ratio \bar{e} [see definition in Eq. (3)], scaled by the number of particles N , binned over and plotted against frequency, for the same models of panels (a)–(e), respectively. The vertical dashed lines mark the first phonon band frequency $2\pi c_s/L$. The horizontal lines represent estimations Ne_0 of the low-frequency plateau, which capture the core size of soft quasilocated modes, see values reported in Fig. 3(a) and text for further discussions.

indicated by the vertical dashed lines, and features Ne of order of a few thousands, consistent with the system sizes employed. Approaching zero frequency, we see that $N\bar{e}$ plateaus at a typical value Ne_0 on the order of a few tens, as marked by the horizontal dashed lines. The estimated values of the plateaus Ne_0 are reported for all investigated computer glass models in Fig. 3(a). Remarkably, the variation of Ne_0 across the different models is very small, of less than a factor of 2 with respect to each other.

Finally, we note that the prefactor \mathcal{A}_g of the nonphononic VDOS, namely $\mathcal{D}(\omega) = \mathcal{A}_g \omega^4$, is an observable with dimensions of an inverse frequency to the fifth power. \mathcal{A}_g was discussed at length in [24,25], where it was argued to encompass information both about the number density of soft, quasilocated modes, and about their characteristic

stiffness. In those references it was shown that \mathcal{A}_g can be very sensitive to glass history, particularly for glasses that were deeply supercooled prior to their quench to the glass. Here, we compare $\mathcal{A}_g \equiv A_g/(a_0/c_s)^5$ across our different computer glasses. The results are displayed in Fig. 3(b); we find that \mathcal{A}_g is of order unity in all models, with the exception of the SW network glass model that features $\mathcal{A}_g \approx 0.25$.

We note that the quantities Ne_0 and \mathcal{A}_g generally depend on glass history [24,25,56]. However, these dependencies are most pronounced for glasses quenched from deeply supercooled liquids, and are generally weak or entirely absent for glasses quenched from high temperature liquid states [24,25,56]. Since in this work we indeed compare glasses quenched from high temperature liquid states (much higher than the computer glass transition temperature), the history dependence of Ne_0 and \mathcal{A}_g is expected to be weak or absent, as we demonstrate explicitly in Fig. S2 of [30]. Consequently, our comparison between these observables across different classes of glass-forming models is meaningful. We conclude that the energy landscapes of the computer glasses we investigate here share *quantitative* similarities that extend beyond the universal scaling of their nonphononic VDOS.

Summary and outlook.—In this work we have shown that the low-frequency nonphononic spectra of realistic computer glass models—including network glasses, polymer glasses, and molecular glasses—feature the universal gapless ω^4 law, as seen previously in simple computer glass models [22–27]. We thus expand the degree of universality of the ω^4 law to include several qualitatively different classes of realistic glass-forming models, and reinforce its

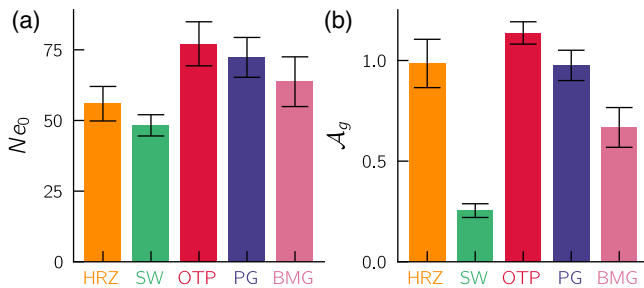


FIG. 3. Dimensionless characterizers of the nonphononic VDOS, compared across different classes of glassy solids. (a) Low-frequency plateau Ne_0 of the frequency-binned participation ratio \bar{e} scaled by N , which represents the core size of soft, quasilocated modes. (b) Dimensionless prefactors $\mathcal{A}_g \equiv A_g/(a_0/c_s)^5$ of the universal $\mathcal{D}(\omega) = \mathcal{A}_g \omega^4$ nonphononic VDOS.

relevance to laboratory glasses. Finally, our results support the description of glasses' vibrational properties via mesoscale, coarse-grained approaches that consider interacting oscillators and anharmonicities [11,12], in which the microscopic details play no role in determining the scaling with frequency of the nonphononic VDOS.

Our results underline the timeliness of formulating a first-principles theory that explains the observed universality of nonphononic spectra in glassy solids. Mean-field approaches that are based on a microscopic description [19–21] (rather than a coarse-grained one) predict that the nonphononic VDOS of glassy solids should scale as ω^2 , independent of spatial dimension. An important goal for future studies will be to consolidate the predictions of the mesoscopic [11,12] and microscopic [19–21] theoretical approaches.

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Note added.—We note that after the completion of this work, we became aware of the results obtained by Bonfanti *et al.* [29], which support our conclusions.

*These authors contributed equally to this work.

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