

Observation of High-Frequency Transverse Phonons in Metallic Glasses

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Using inelastic neutron scattering and molecular dynamics simulations on a model Zr-Cu-Al metallic glass, we show that transverse phonons persist well into the high-frequency regime, and can be detected at large momentum transfer. Furthermore, the apparent peak width of the transverse phonons was found to follow the static structure factor. The one-to-one correspondence, which was demonstrated for both Zr-Cu-Al metallic glass and a three-dimensional Lennard-Jones model glass, suggests a universal correlation between the phonon dynamics and the underlying disordered structure. This remarkable correlation, not found for longitudinal phonons, underscores the key role that transverse phonons hold for understanding the structure-dynamics relationship in disordered materials.

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Introduction.—Normal vibration modes and vibrational density of states (VDOS) are fundamental for understanding many of the physical properties of materials, such as dynamical excitations, and mechanical and thermal transport properties [1]. In crystalline materials, the normal modes, known as phonons, are quantized plane-wave solutions of the elemental modes of vibration [2], which can be well characterized experimentally by, e.g., inelastic neutron scattering (INS) [3]. In amorphous materials, however, the phonon modes become far more complicated and are very different from those in crystals [4–13]. Most notably, the phonon spectra become broadened, and the extent of broadening depends on the degree of the disorder [14,15]. Still, in amorphous materials, phononlike dispersions have been predicted, for example, by analytical theories for a three-dimensional disordered system, first proposed by Hubbard and Beeby [4] and further developed by Takeno and Goda [6,16]. An extended review can be found in Yoshida and Takeno [17], with applications to both classical and quantum liquids. Calculations based on these theories show that the longitudinal mode has a strong dispersion, whereas the transverse mode is dispersionless at large wave vectors [4,6,16,17].

In amorphous materials, the transverse rather than the longitudinal phonon mode is more sensitive to disorder [7,18]. The transverse phonons are also responsible for many of the properties associated with the disorder. As an example, the velocity of transverse phonons is proportional to the square root of the shear modulus, which largely determines the deformation behavior of disordered materials [9,19]. Moreover, the transverse phonons have been linked with the Boson peak [7,8], or the excess VDOS, which is a universal feature in disordered materials. Recent simulations have found that for transverse phonons, the full-width-at-half-maxima (FWHM) of the phonon energy spectrum increases sharply with the momentum transfer Q [7]. At a certain Q , the phonon wavelength and mean free path becomes comparable. This is the so-called Ioffe-Regel limit, beyond which the transverse phonons were believed to be extremely damped and therefore could no longer propagate [7].

Thus, it is of great interest, from both fundamental and practical perspectives, to understand transverse phonons, especially at large Q where the effect of disorder becomes particularly pronounced. Yet, in spite of their importance, transverse phonons are difficult to determine

experimentally [7,18,20]. For this reason, little is known about the nature of transverse phonons, e.g., the contribution by different atomic species, or whether they exist at all. INS and lately inelastic x-ray scattering (IXS) are methods of choice for the experimental study of phonons [3,5,8,10,20–23]. While longitudinal phonons are straightforward to determine, measurements of transverse phonons require more considerations. In crystalline materials, the scattering intensity for transverse phonons vanishes in the first Brillouin zone, due to the scattering geometry factor, so the measurements of transverse phonons are typically done in the second Brillouin zone and beyond [22,23]. In disordered materials, however, the disorder-induced phonon broadening [14] makes it difficult to determine transverse phonons due to the crossover of longitudinal and transverse branches. Thus, although a few IXS studies reported the measurements of transverse acoustic excitations in metallic liquids and glasses [5,24,25], the data are limited to small wave vectors (below $Q \sim 1 \text{ \AA}^{-1}$) and with significant uncertainties. INS is capable of reaching to larger Q without suffering from the form factor dropoff as in IXS. Still, previous INS data have been plagued with inadequate precision due to the broad phonon spectrum and limited neutron beam flux [5,20].

The new chopper spectrometers at the state-of-the-art pulsed neutron sources provided opportunities to investigate the transverse phonons in disordered materials [26]. The high flux brought forth by the powerful sources, coupled with advances in neutron instrumentation, has enabled high-precision measurements to large Q , with fine energy resolution. Here, we report INS data for a ternary Zr-Cu-Al metallic glass (MG), where evidence of high-frequency transverse phonons was observed at an intermediate wave vector, around the boundary of the second quasi-Brillouin zone (QBZ) [20], see Fig. 1(a) for definition. The experimental observations are supported by molecular dynamics (MD) simulations, which not only confirms the existence of the transverse phonon branch at high-frequency but also shows how individual atoms contribute to the extended transverse mode. These findings present a challenge to the viewpoint that transverse phonons exist only at low energies, below the Ioffe-Regel limit [7,18]. MD simulations answer the paradox, by showing that the transverse phonon peak width follows the static structure factor, rather than increasing monotonously with the phonon frequency.

Experimental and simulation methods.—Sample preparation and characterizations have been described elsewhere [27]. The INS experiment was carried out to measure the atomic dynamics in $\text{Zr}_{46}\text{Cu}_{46}\text{Al}_8$ MG, using the time-of-flight wide angular-range chopper spectrometer (ARCS) [28] at Spallation Neutron Source [29]. The measurements were performed with incident neutron energies of $E_i = 50$ and 80 meV at room temperature (RT). For each E_i , the dynamic structure factor $S(\mathbf{Q}, E)$, where $\mathbf{Q}$ and E are the

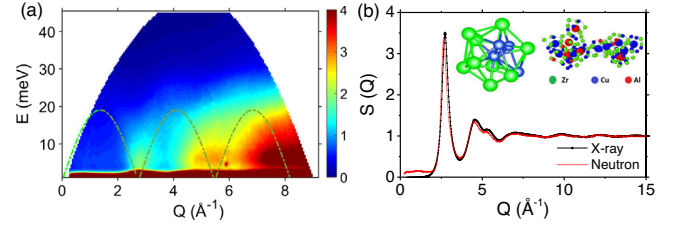


FIG. 1. The dynamic and static structure factors in $\text{Zr}_{46}\text{Cu}_{46}\text{Al}_8$ MG. (a) Dynamic structure factor $S(\mathbf{Q}, E)$, measured by INS with $E_i = 50$ meV on ARCS at RT. The dashed curve is calculated by the function, $E_0 |\sin[\pi(Q/Q_{\max})]|$, which defines the quasi Brillouin zone (QBZ), a term analogous to the Brillouin zone in crystalline materials in order to facilitate discussions of the phonon dynamics over different Q regions. Here, $Q_{\max} = 2.74 \text{ \AA}^{-1}$ is the position of the first sharp diffraction peak and $E_0 = 19.02$ meV is an estimated Einstein frequency for vibration. (b) Static structure factor $S(Q)$, measured by synchrotron x-ray and neutron total scattering at RT, demonstrated the fully amorphous nature of the sample. The inset shows a schematic configuration of the cluster-centered structure in $\text{Zr}_{46}\text{Cu}_{46}\text{Al}_8$ MG.

momentum and energy transfer, respectively, was generated using standard software MantidPlot [30]. One-dimensional “ Q cuts” were taken along the E axis to obtain the phonon spectra at specific Q points by DAVE [31].

Classical MD simulations were performed for $\text{Zr}_{46}\text{Cu}_{46}\text{Al}_8$ MG with the realistic embedded atom (EAM) method potential [32] using LAMMPS software package [33]. The glassy sample containing 10 000 atoms at 300 K was obtained by fast quenching the equilibrated liquid at 2000 K with a cooling rate of 10^{12} K/s. Periodic boundary conditions were applied in three directions. In the cooling process, the isobaric and isothermal ensemble was employed and the sample size was adjusted to give zero pressure. This was followed by the canonical ensemble MD at 300 K for data collection and analysis. The simulated static structure factor was benchmarked with experimental measurements [27]. A three-dimensional Lennard-Jones binary glass was also simulated for comparison. The dynamic properties were analyzed in terms of van Hove correlation function (VHF), dynamic matrix method (DM), and velocity correlation function method (VCF), respectively. More details on simulation can be found in Supplemental Material [27].

Results.—Figure 1(a) shows a representative INS data set of the $S(\mathbf{Q}, E)$ for $\text{Zr}_{46}\text{Cu}_{46}\text{Al}_8$ MG, obtained with $E_i = 50$ meV [27]. The full glassy nature of the samples was confirmed by the static structure factors $S(Q)$, measured with neutron and synchrotron x-ray diffraction, shown in Fig. 1(b).

The measurement results were analyzed in terms of the generalized Q -dependent phonon density of states (GDOS), $G(\mathbf{Q}, E)$, which is related to $S(\mathbf{Q}, E)$ by the following equation [20,22,34],

$$G(\mathbf{Q}, E) = [1 - e^{-E/k_B T}] \frac{E}{Q^2} S(\mathbf{Q}, E), \quad (1)$$

where $[1 - e^{-E/k_B T}]$ describes the Bose-Einstein statistics, k_B is the Boltzmann constant, and T the temperature. The GDOS was reduced from the INS data using DAVE-mslice software [31]. Note that GDOS is slightly different from the current spectrum, defined as $C(Q, E) = (E^2/Q^2)S(Q, E)$, which has also been frequently used in analysis of the excitation spectrum in glasses and liquids [5]. The high-resolution experimental Q -dependent GDOS (with $E_i = 50$ meV) for $\text{Zr}_{46}\text{Cu}_{46}\text{Al}_8$ MG is presented in Fig. 2(a). A characteristic dispersion relationship can be readily seen. The phonon spectrum is significantly broadened in energy, with the center band located at ~ 10 to 30 meV. The corresponding data for $\text{Zr}_{46}\text{Cu}_{46}\text{Al}_8$ MG obtained with $E_i = 80$ meV are similar [27]. The phonon spectrum is dominated by the acoustic modes; no optical mode can be seen in Fig. 2(a), even in Q -dependent GDOS

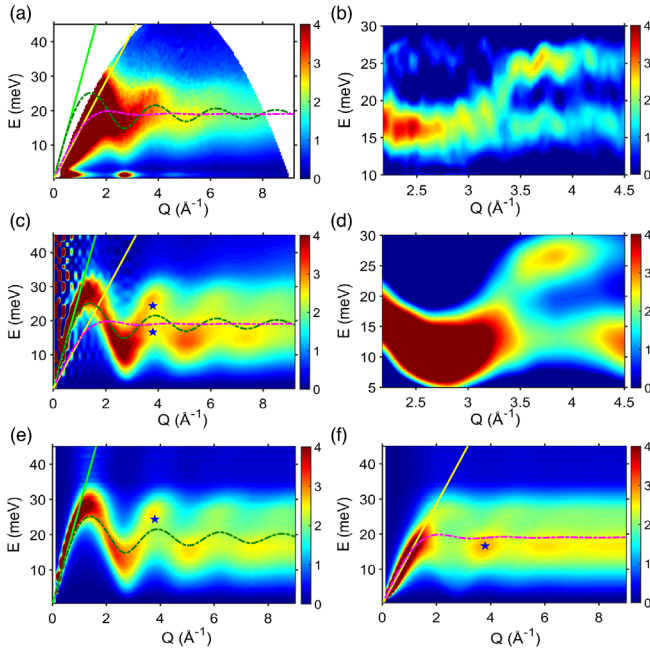


FIG. 2. Phononlike dispersion relationship in $\text{Zr}_{46}\text{Cu}_{46}\text{Al}_8$ MG. (a),(c) Phonon Q -dependent GDOS reduced from INS data and the corresponding neutron-weighted MD simulation results based on the VHF method. (b),(d) Maps of the second derivatives of (a) and (c) in the vicinity of the second QBZ, respectively. Only the absolute values are plotted to trace the peaks, following the practice in Refs. [35,36]. (e),(f) The longitudinal and transverse Q -dependent VDOS obtained from MD simulations by the DM method. The results clearly show that the transverse branch has an intensity maximum around $Q = 3.8 \text{ \AA}^{-1}$. The olive and magenta curves in (a),(c),(e),(f) are calculated longitudinal and transverse phonon dispersions based on the analytical theory for a disordered system [4,6,16,17,27]. The green and yellow lines in (a), (c),(e),(f) are calculated dispersions based on longitudinal and transverse sound velocities measured by the RUS method. These data are consistent with each other in the low Q regime. The blue stars in (c),(e),(f) marks the boundary of the second QBZ, where the transverse phonons manifest. For the purpose of comparison, the color bars are plotted in relative intensities.

obtained with higher incident energies [27]. To examine the acoustic nature of the vibration modes, we performed resonant ultrasound spectrometer (RUS) measurements [27]. The green and yellow solid lines in Fig. 2(a) are calculated dispersions at small Q based on sound velocities measured by RUS, $V_L = 4218$ m/s and $V_T = 2165$ m/s, for longitudinal and transverse sound modes, respectively.

The dashed curves in Fig. 2(a) are analytical calculations for dispersion relations in a disordered system [4,6,16,17,27], with the parameters of the Einstein frequency and inter-particle distance taken from experimental $G(Q, E)$ and $S(Q)$, respectively [27]. Well separated longitudinal and transverse phonon branches are predicted by this model at QBZ boundaries and centers, e.g., at $Q \sim 2.7 \text{ \AA}^{-1}$ (the center) and $Q \sim 3.8 \text{ \AA}^{-1}$ (the boundary) of the second QBZ. Unfortunately, it is difficult to identify different branches from the experimental Q -dependent GDOS, because the phonon spectra are too broad and the branches overlap. The simulated results analyzed by the VHF are shown in Fig. 2(c). Here, a hint of branch separation can be seen at $Q \sim 3.8 \text{ \AA}^{-1}$, the second QBZ boundary. The blue stars superimposed in Fig. 2(c) are the longitudinal and transverse phonon peak positions extracted by fitting the Q -cut experimental GDOS at $Q \sim 3.8 \text{ \AA}^{-1}$ [27].

To confirm the separation of the longitudinal and transverse modes at $Q \sim 3.8 \text{ \AA}^{-1}$, the second-derivative method, a powerful data analysis technique to separate overlapping branches, was employed. This method, also known as the curvature method, has been successfully used in angle-resolved photoemission spectroscopy (ARPES) experiments to identify subtle features in the electronic density of states [35,36] from, e.g., Weyl fermions in semimetals, and recently in electron energy loss spectroscopy experiments to identify the plasmons in a topological insulator [37]. In this study, the second derivative with respect to E was calculated and examined. The results are visualized in Figs. 2(b) and 2(d) in the vicinity of the second QBZ, for the INS and MD simulated spectra. Two branches can be seen to separate starting from $Q \sim 3.5 \text{ \AA}^{-1}$. Linking together the experimental results, MD simulation, and the calculations by the analytical model, we can identify that at $Q \sim 3.8 \text{ \AA}^{-1}$, the upper branch at $E \sim 24$ meV corresponds to the longitudinal mode, whereas the lower branch at $E \sim 17$ meV corresponds to the transverse phonons.

In spite of the apparent success and expanding applications, caution must be exercised when applying the second-derivative method. A shift in peak position was sometimes noted when the second-derivative method was applied to the ARPES data. This was discussed in detail by Zhang *et al.* [38], and a correction method has been proposed. The peak produced by the second-derivative method may also be subject to large error, if the original data are of poor quality, e.g., due to inadequate counting statistics. Naturally, the second-derivative method is most effective

when discussed with reliable models, where the simulation results provide guidance on what the original and second-derivative data might look like.

To further verify and characterize the identified phonon modes, the DM and VCF methods were employed independently to compute the contributions of longitudinal and transverse branches. A comparison of the pros and cons of these three methods is discussed in Supplemental Material [27]. Overall, the calculations by different methods are consistent. The Q -dependent VDOS results calculated by the DM method are presented in Figs. 2(e) and 2(f), while those by the VCF method are shown in Supplemental Material [27]. Both methods show a dispersive longitudinal branch, while the transverse branch is flat beyond $Q \sim 2 \text{ \AA}^{-1}$, consistent with the calculations by analytical theories [4,6,16,17,27]. Distinctively, however, the transverse spectrum calculated by both methods exhibits a high concentration of strong intensities at $E \sim 17 \text{ meV}$ around $Q \sim 3.8 \text{ \AA}^{-1}$, exactly where the second derivative of the experimental Q -dependent GDOS shows a separation.

More importantly, MD simulations by DM provided atomistic insights on the nature of the different phonon modes. In Fig. 3(a), the VDOS and the contributions from individual atoms are plotted. We emphasize here that the VDOS and GDOS obtained in MD simulations are almost the same [27], because Cu and Zr have similar coherent scattering lengths and the contribution from Al is small. Thus, a comparison can be made between the calculated VDOS and experimental GDOS. Three peaks are identified in the MD simulations, which can be mapped to the

experimental GDOS data. The MD simulations indicate that both Zr and Cu atoms contribute to the peaks at 17 and 24 meV. There is no contribution from Al atoms, however. This result also means that the experimentally observed phonon modes, at 17 and 24 meV (both transverse and longitudinal), involve Zr and Cu atoms only. On the other hand, the higher energy modes of 32 to 50 meV are mostly due to Al atoms.

To quantify the extent of a vibrational mode, the participation ratio $p(\omega_\lambda)$ was calculated as follows:

$$p(\omega_\lambda) = \frac{[\sum_j |\mathbf{e}_\lambda(\mathbf{r}_j)|^2]^2}{N \sum_j |\mathbf{e}_\lambda(\mathbf{r}_j)|^4}, \quad (2)$$

where $\mathbf{e}_\lambda(\mathbf{r}_j)$ represents the normalized eigenvector of each atom j in the mode ω_λ . $p \approx 1/N$ corresponds to the localized modes, while $p \approx 1$ corresponds to the fully extended modes. Figure 3(b) shows the p of all vibrational modes. A p value of $\sim 50\%$ for modes between 10 and 25 meV demonstrates that these vibrational modes are spatially extended [39]. They penetrate through the entire material, close to the nature of acoustic waves in solids. For the modes of 32 to 50 meV, which are solely due to Al atoms, the p values ($< 10^{-3}$) are very low and system size dependent, retained indicating that these modes are indeed localized [9,40,41]. Thus, the mode of $E \sim 17 \text{ meV}$ is spatially extended, which is a necessary condition for propagating modes. To further elucidate the plane-wave feature of this mode at $E \sim 17 \text{ meV}$ and $Q \sim 3.8 \text{ \AA}^{-1}$, we analyzed the projection of the eigenstates on the plane waves over the whole Q range. As shown in Fig. 3(c), there exhibits strong Q dependence in both longitudinal and transverse VDOS $D_\alpha(Q, E)$ ($\alpha \in L, T$) at $E = 17 \text{ meV}$, indicating strong spatial correlation of this mode. The peaks of the Q dependent $D_T(Q, E)$ indicate that there exist some characteristic Q values in the transverse part of this mode, and one is located at $Q \sim 3.8 \text{ \AA}^{-1}$. This demonstrates the plane-wave character of the transverse phonon at $Q \sim 3.8 \text{ \AA}^{-1}$. In other words, the traditional transverse phonons still have its trace in high-frequency modes in disordered materials. We also compared $D_T(Q, E)$ for the modes of $E = 10$ and 23 meV whose p values are also about 0.5. As shown in Fig. 3(d), in contrast to the mode of $E = 17 \text{ meV}$, the modes of $E = 10$ and 23 meV show much weaker Q dependence. This contrast reinforces our conclusion that the observed high-frequency transverse phonon at $E = 17 \text{ meV}$ is indeed a spatially extended mode.

Discussions.—As disorder-induced broadening is a signature of the phonon spectrum in disordered materials [7,14,15], we have further studied the phonon width, or the lifetime, using MD simulations. The dispersion curves and peak widths were estimated by fitting the VDOS calculated by DM [Figs. 2(e) and 2(f)] with a Gaussian function,

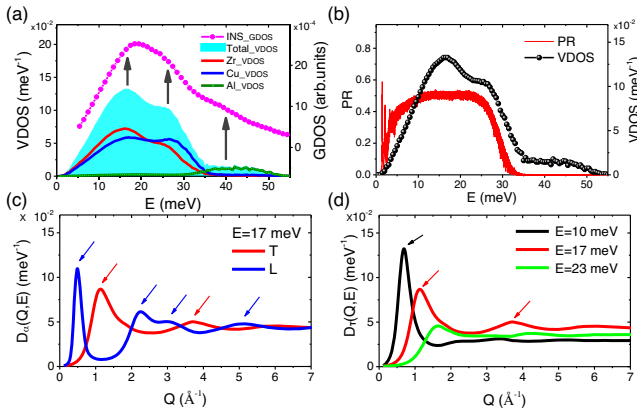


FIG. 3. MD simulation results based on the DM method. (a) The total VDOS, and partial VDOS for each element, and (b) the participation ratio (PR) for each mode. For comparison, the experimental GDOS is superimposed in (a), which was obtained by averaging a Q range of 2.5–9.0 $\text{\AA}^{-1}$ for the INS data. (c) Q dependence of the transverse and longitudinal VDOS $D_\alpha(Q, E)$ ($\alpha \in L, T$) at $E = 17 \text{ meV}$. These peaks indicate the plane-wave characteristics in this mode. (d) Comparison of $D_T(Q, E)$ at $E = 10, 17$, and 23 meV . The mode of $E = 17 \text{ meV}$ shows stronger transverse wave characters around $Q = 3.8 \text{ \AA}^{-1}$.

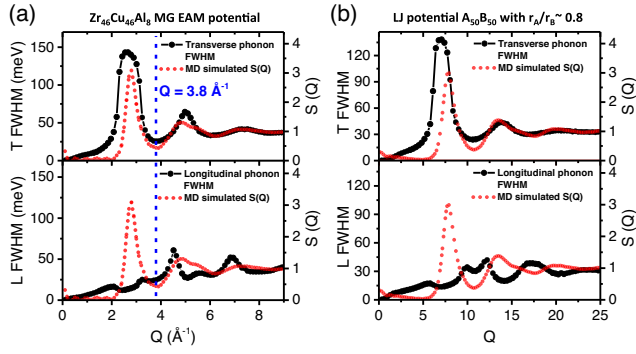


FIG. 4. Peak widths of the longitudinal and transverse phonon spectra vs $S(Q)$ obtained by MD simulations based on the DM method. The widths were obtained by fitting the MD spectra with a Gaussian function [20,27]. (a) Empirical potential for $\text{Zr}_{46}\text{Cu}_{46}\text{Al}_8$ MG. (b) Lennard-Jones (LJ) potential for a model glass. A one-to-one correlation can be seen between the apparent peak width and $S(Q)$ for the transverse phonon mode. These results suggest a universal correlation between the transverse phonon dynamics and the underlying disordered structure. No correlation was found for the longitudinal phonons. These LJ results are in scaled units.

following Arai *et al.* [20]. Fitting with a Lorentzian function gave similar results. Details of fitting are given in Supplemental Material [27]. We have found that for transverse phonons, the phonon width, measured by the FWHM, follows the $S(Q)$, as illustrated in Fig. 4(a). Like the $S(Q)$, the FWHM reaches a minimum at the boundary of the second QBZ. This is where the spectrum for the transverse phonons is the sharpest, and hence the reason that the transverse phonons were observed at this Q value rather than elsewhere. That being said, our study does not contradict the findings from Ref. [7], which is limited to small Q , where the phonon peak width increases sharply with Q . Instead, our study points out that transverse phonons survive at larger Q . For the longitudinal mode, the phonon width also varies with Q , but there seems to be no correlation with $S(Q)$.

The correlation between the FWHM of transverse phonons and $S(Q)$ suggests that in amorphous materials, the phonon dynamics and static structure are intimately related. To check on whether this observation is specific to MGs, we have carried out a simulation with a generic Lennard-Jones potential, and the results are shown in Fig. 4(b) and Supplemental Material [27]. The remarkable one-to-one correspondence between the phonon FWHM and $S(Q)$ demonstrated here strongly suggests that the correlation is universal for amorphous materials. As in $\text{Zr}_{46}\text{Cu}_{46}\text{Al}_8$, the best point to observe the transverse phonons is also at the boundary of the second QBZ, or the valley of $S(Q)$ following the first sharp diffraction peak, where the transverse phonons reveal themselves with the sharpest spectrum.

Conclusion.—Through high-quality INS measurements, we have established the existence of transverse acoustic

phonons in amorphous materials. The transverse phonon modes, although significantly damped or broadened, persist well into the high-frequency regime, and beyond the second QBZ. MD simulation provided the atomistic insights, showing that the transverse phonons are spatially extended, rather than localized, and involve only Zr and Cu atoms which form the short-range atomic clusters. Moreover, the simulations showed a universal one-to-one correspondence between the transverse phonon's width and the static structure factor, suggesting that in amorphous materials, the structure and dynamics are linked. It should be mentioned that the one-to-one correspondence seems to be unique to glass, as it is not found in crystalline materials. As transverse phonons are related to many fundamental phenomena in glass, such as the Boson peak and deformation behaviors, the findings of the present study should have broad implications in future study of the nature of glass and glass transitions.

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