Rayleigh anomalies and disorder-induced mixing of polarizations in amorphous solids

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Acoustic dynamics in disordered media present anomalous features in respect of the Debye’s theory. This is observed in attenuation, retardation and depolarization of acoustic excitations. Each of these phenomena have attracted a large interest but, even if they can originate from the same root, the disordered nature of the medium, they have been analyzed independently. Using a stochastic approach at wavelengths of the order of the characteristic length-scale of heterogeneities, allowed us to achieve a quantitative, experimentally corroborated and unified description of acoustic dynamics properties in a disordered medium. These include the so-called Rayleigh anomalies (attenuation and retardation) and, for the first time, the mixing of longitudinal and transverse polarizations occurring at higher wavevectors. Model predictions are compared to acoustic dynamics features and vibrational density of states experimentally determined in an ionic glass, obtaining an excellent agreement.

The reasearch on glasses in the last decades focused on the macroscopic anomalies which characterize as a whole these systems with respect to long-range ordered solids. In particular, a hump over the level predicted by the Debye theory is observed in the specific heat of glasses at about 10 K (1). It is related to an excess over the Debye level of the Vibrational Density of States (VDOS) at energies of few meV, called Boson Peak (BP) (2–8). The physical origin of this feature has been largely debated in the literature without merging into a unified theory. It is noteworthy that all existing theories beg the question of the effect of elastic disorder on acoustic waves, whether it is the disorder treated as only a small perturbation with respect to the ordered structure of the crystals (8, 9), modeled as defects embedded in an otherwise homogeneous medium (6, 10) or represented as spatial fluctuations of elastic moduli - local (3–7, 11) or with long-range correlations (12).

In this context large emphasis was given to the so-called Rayleigh anomalies (3–7, 11, 13–15). They consist in a strong increase of the acoustic wave attenuation and softening of the phase velocity with respect to the macroscopic value - the two quantities being related to each other by the Kramers-Kroning relations (7) - observed for wavelengths of few nanometers. The experimental study of acoustic excitations at such length-scales with the experimental techniques so far available is quite difficult (11, 13), thus obfuscating the experimental verification of the different theories.

An acoustic wave traveling in a three-dimensional material is characterized by its phase velocity, amplitude and polarization. It is known from elementary elasticity theory (16) that a purely longitudinal (or transverse) wave impinging on an interface between two different elastic media is transformed in waves with mixed polarization. Acoustic waves with mixed polarization have been observed by both Inelastic X-ray and Neutron Scattering (IXS, INS) techniques (17–20) as well as by MD simulations (21–23) in several amorphous solids and liquids in the first pseudo-Brillouin zone, typically at wavevectors equal to 4 – 5 nm⁻¹. This latter phenomenon, however, has never been related to Rayleigh anomalies and quantitatively described as a phenomenon also originating from the disordered nature of the medium. The authors believe that a coherent, full-blown, experimentally verifiable, mathematical description of all the phenomena arising from the elastic heterogeneous structure of an amorphous solid and affecting its acoustic excitations finally could trace the way towards the understanding of how microscopic disorder can be exhaustively described and how it can affects macroscopic properties in glasses. A stochastic approach developed in the framework of the Random Media Theory (RMT) (24), or Heterogeneous- Elasticity Theory (HET) (3–5) if one refers to the case of elastic constants inhomogeneity, would permit to reach this goal. However, the impossibility to solve of the Dyson equation, which describes the ensemble averaged elastodynamic response of the system to an impulsive force, leads to the introduction of scale-dependent approximations (4, 5, 24–27), thus hindering the unified description of these phenomena in turn occurring on different length scales. The Rayleigh anomalies appear at values of wavelength, λ, of elastic excitations lower than the average radius of inhomogeneity domains, a, whereas the coupling between longitudinal and transverse excitations (depolarization) is maximum when λ becomes comparable to a (28). On the other hand, the parameters of the theory, such as the average size of heterogeneity domains or the strength of spatial fluctuations, only in rare cases can be a priori determined by experiments (3–5, 24). They are usually chosen as ad hoc parameters to permit a correct description of the phenomena one aims to explain. This could give rise to tautologies. The goal of the present study is the building of a theoretical framework, able to make quantitative and verifiable predictions and allowing a unified description of the phenomena so far addressed. Within this aim we developed in the framework of the RMT, a simple enough, mathematically tractable approximation. It allows for a complete description of a real system, the ionic glass 1-Octyl-3-methylimidazolium chloride ([C8MIM][Cl]), whose heterogeneous structure can be well assessed and experimentally characterized. Furthermore, the acoustic dynamics of this system has been characterized by IXS with a detail never offered so far in the whole pseudo-Brillouin zone of a glass. Because of the analytical approach, a
certain level of generality of the proposed model is guaranteed. It can be thought as a starting point for describing acoustic dynamics in different kind of glasses, composites, ceramics, geophysical systems, or propagation of different kind of waves in disordered media.

Results

Experimental characterization of longitudinal acoustic waves in 1-Octyl-3-methylimidazolium chloride ionic glass. We report the experimental characterization of longitudinal dynamics and Vibrational Density of States (VDOS) of [C8MIM][Cl] at \( T = 176 \) K \( (T_0 = 214 \) K) obtained respectively by IXS and INS. Fig. 1 Panels I-IV shows IXS spectra for selected wavevector (Q)-values *.

* In the present text capital Q states for wavevectors experimentally determined (thus affected by undeterminancy), non-capital q states for a wavevector introduced inside the theory. If experimental and theoretical outputs are put in contrast, this latter notation will be preferred.

The measured signal reproduces the longitudinal dynamical structure factor, \( S_L(Q, E) \). The measured signal is modeled in turn with a sum of a delta function for the elastic component and a damped harmonic oscillator (DHO) function for the inelastic component. Such a protocol provides the characteristic energy and broadening (attenuation) of the inelastic excitation, respectively \( \Omega \) and \( \Gamma \). In the high-wavevectors region \( (Q > 5 \text{ nm}^{-1}) \) an additional feature on the inelastic component is observed. Similar features were so far observed in several glasses, see e.g. Refs. (17–19), and commonly related to 'projection' of transverse into longitudinal dynamics. To account for the presence of the extra feature, two DHO contributions can be inserted in the fitting model (see Fig. 1 Panel VI). Within the aim to enforce the reliability of the comparison with theoretical outcomes, these latter will be compared with experimental results obtained both by using in the fitting model a single-DHO function in the whole explored Q-range or by using two DHO-functions in the high-Q region. Experimentally determined \( \Omega \) and \( \Gamma \) are displayed in Fig. 2 Panels I and III. The figure, furthermore, shows the measured VDOS normalized to the square of the exchanged energy, \( g(E)/E^2 \), (Panel II) and the static structure factor, \( S(Q) \), measured by X-ray scattering (Panel III). The reduced VDOS presents (i) a peak around 2 meV, referred to be the Boson Peak (BP) (30); (ii) a broad feature at higher frequencies, in the region between 7 and 12 meV, related to librational modes of the imidazolium ring (30) - referred hereafter as Intermolecular Vibrational Modes (IVMs). The static structure factor shows - as it is the case for most ionic liquids (ILs) - a First Sharp Diffraction Peak (FSDP) at the exchanged energy between the probe and the sample, \( h \) is the Planck constant, \( \omega \) is the exchanged frequency. \( S_L(Q, E) \) is modeled in turn with a sum of a delta function for the elastic component and a damped harmonic oscillator (DHO) function for the inelastic component. Such a protocol provides the characteristic energy and broadening (attenuation) of the inelastic excitation, respectively \( \Omega \) and \( \Gamma \). In the high-wavevectors region \( (Q > 5 \text{ nm}^{-1}) \) an additional feature on the inelastic component is observed. Similar features were so far observed in several glasses, see e.g. Refs. (17–19), and commonly related to 'projection' of transverse into longitudinal dynamics. To account for the presence of the extra feature, two DHO contributions can be inserted in the fitting model (see Fig. 1 Panel VI). 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The static structure factor shows - as it is the case for most ionic liquids (ILs) - a First Sharp Diffraction Peak (FSDP) at
where summation over repeated indices is assumed, $s$ is a wavevector and the integral extends to $\mathbb{R}^3$. The function $R_{a\beta;j}^{\gamma\delta jk}(q)$ is the Fourier transform of the covariance of the elastic tensor fluctuations, $R_{a\beta;j}^{\gamma\delta jk}(r = r_1 - r_2) \sim \delta C_{a\beta;j}^{\gamma\delta jk}(r_1) \delta C_{jkl}(r_2)$. Thus, the self-energy in the Fourier space can be written as a convolution between the bare Green's dyadic and the Fourier transform of the covariance of the elastic tensor fluctuations. It is possible to show that the Bourret approximation holds for small fluctuations intensity, wavevectors and frequencies (25). The so-called Self-consistent Born Approximation (SCBA) (3, 5) or Kraichnan model (27) is a generalization of the Bourret (Born) approximation, which states

$$\Sigma_{ij}(q, \omega) = \tilde{L}_{1gjk} < G_{jk}(q, \omega) > ,$$

Eq. 3 together with Eq. 1 corresponds to successive self-consistent approximations for $\Sigma(q, \omega)$ and $< G(q, \omega) >$. At the zero-th step it is $< G(q, \omega) > = G^0(q, \omega)$. The first step of the iteration procedure thus corresponds to the Born approximation. The SCBA holds under the hypothesis of small fluctuations (3, 5) but it is not affected by the limitation in frequencies and wavevectors of the Born approximation. Analytical solutions of the self-consistent set of equations (Eqs. 1 and 3) while preserving their wavevector-dependence involves computational drawbacks, thus approximations or numerical approaches are required. Analytical expressions have been obtained in Rayleigh Region (RR) by assuming $q = 0$ in the expression of $\Sigma(q, \omega)$ at each step of the self-consistent procedure (SCBA) (3, 7, 11), allowing a description of the Rayleigh anomalies (7, 11). Outcomes from this procedure are discussed in Supporting Information.

We propose a novel approximate method for the calculation of $\Sigma(q, \omega)$. It introduces corrective terms to the Born approximation in the framework of the SCBA. We will thus refer to it as a Generalized Born Approximation. We report here the final expression for $\Sigma(q, \omega)$, whereas we outline the passages which lead to it in Sec. Method. Under the hypothesis of local isotropy \footnote{Pure transverse modes do not contribute to the measured IXS signal in the first Brillouin zone. In solids with local anisotropy quasi-longitudinal or quasi-transverse modes (i.e. modes composed by different polarizations with a dominant polarization component) exist in the mesoscopic region. They can contribute to the measured IXS signal giving rise to the shoulder at a frequency near the zero characteristic of the transverse excitation. It is possible, in the frame of RMT, to account both for local and statistical anisotropy (A. J. Turner, Elastic wave propagation and scattering in heterogeneous, anisotropic media: Textured polycrystalline materials., J. Acoust. Soc. Am. 106, 541 (1999)). We neglected such effects because our aim is to point out that, even in a isotropic medium, the disorder can generate the mixing of polarizations.}, we introduce the orthonormal vector base defined by the direction of wave propagation and the two orthogonal directions (26). On this basis all the ‘bare’ and average Green dyadics and the self-energy are diagonal. We introduce their longitudinal and transverse components, respectively $g_{L(T)}$, $\Sigma_{L(T)}$. These latter, in turn, contain two terms accounting for the coupling with longitudinal and transverse dynamics respectively, i.e. $\Sigma_{L(T)} = \Sigma_{L(T)}^{LL} + \Sigma_{LT(TL)}$. In the Generalized Born Approximation we find

$$\Sigma_{ij}(q, \omega) \sim \tilde{L}_{1gjk} \left\{ \frac{1}{\eta_{pk}} + \frac{1}{\eta_{pk}^2 - \eta_{qk}^2} \right\} ^2 \Delta \Sigma_{ij}(0, \omega).$$

where $\Delta \Sigma_{ij}(q, \omega) = [\Sigma_{ij}^{LL}(q, \omega) - \Sigma_{ij}^{LT}(q = 0, \omega = 0)]^2$ and $[\Sigma_{ij}^{LL}(q = 0, \omega = 0)]^{1/2}$ is the macroscopic velocity of the first step perturbed medium, $c_i^0$ is the phase velocity of the ‘bare’ medium along a given direction (longitudinal or transverse), $\epsilon$ is the square of the intensity of spatial fluctuations per density. The suffix 1 marks a quantity calculated to the first step of the self-consistent procedure. Generalizations of the Born approximation have attracted interest in several fields of physics (34, 35). The validity of the present approximation can be demonstrated up to wavelengths of the order of the average frequency $q_{FSDP} = 2.8 \text{ nm}^{-1}$. It is related to nanoscale segregation of the cations alkyl chains (31, 32), which in turn results in a local structure formed by an alternation of polar and nonpolar domains (33). The characteristic length scale defined by the FSDP, $2\pi/q_{FSDP}$, gives thus a rough estimation of the diameter of nonpolar domains (31), i.e. $2\pi \sim 2\pi/q_{FSDP}$ (see Fig. 4). Since, furthermore, intermolecular forces acting in polar and apolar regions are of different nature, it is possible to assume that (i) to the heterogeneous local structure corresponds an elastic heterogeneous structure (23); (ii) the elastic constants difference between the two kind of domains is quite large. The information that we can extract from inspection of Fig. 2 are in summary: (i) the existence of a crossover in the $\Gamma$ trend at $Q_c = 4.8nm^{-1}$; (ii) a related kink at $Q_c$ in the $\Omega$ dispersion; (iii) for $Q > Q_c$ the existence of two inelastic features in $S_L(Q, E)$. The crossover in the $\Gamma$ trend has been observed in several other glasses, see e.g. Refs. (13, 15). It usually falls at frequency in the region of the BP, i.e. at slightly lower frequency than in the present case. A power law of 4-th and 2-th order can describe the experimental trend of $\Gamma$ for $Q$ respectively lower or higher than $Q_c$. We observe that $Q_c \sim 2Q_{FSDP} \sim 2\pi/a$, i.e. the crossover appears at wavevectors related to the typical size of elastic heterogeneity domains. We notice, on the other hand, that $Q_c$ correspond as well to the energy cross point between longitudinal acoustic mode dispersion and the energy position of the barycenter of the broad high-frequency feature in VDOS at $\sim 9 \text{ meV}$ related to IVMs (see Fig. 2). The possible coupling between acoustic waves and IVMs is discussed in Supporting Information.
The main insights which can be gained from our theoretical
waves dispersions calculated in the framework of the RMT
and accounted for by a scalar model or by a dynamic structure factor presents features, which cannot be
attenuation show respectively the softening and the crossover
transition (Fig. 3). Its characteristic phase velocity and
representation of crystals (8, 9) with the consequent occurrence of the
the present case can be fixed by the
an exponentially decaying function with correlation length
a. The input parameters of the theory are α, whose value in the
present case can be fixed by the a priori (experimental)
knowledge of the local structure a = {D_{DD/DF}}, ε^2 and c^0(T).

It has been shown that the short-range structure of glasses
preserves residual order that characterizes the long-range structure
of crystals (8, 9) with the consequent occurrence of the pseudo-Brillouin zone and the related bending of the acoustic
waves dispersion. We empirically superimpose it to acoustic waves dispersion calculated in the framework of the RMT (see Fig. 3) by a suitable normalization of the frequency of
G^0(q, ω) in Eq. 1 as described in Sec. Method.

Discussion

The main insights which can be gained from our theoretical
approach are enclosed in Figs. 3 and 4 and outlined in the
following. (1) In the Rayleigh region (qa < 1) the dynamic structure factor is characterized by one only inelastic excitation (Fig. 3 Panel III). Its characteristic phase velocity and
attenuation show respectively the softening and the crossover in the q trend, as it is possible to infer from Fig. 4 Panels I and III, low q points. (2) In the region qa ≈ 1 the
dynamic structure factor presents features, which cannot be
accounted for by a scalar model or by SCBA RR. It is observed in S_r(q, ω) a shoulder at a frequency close to the characteristic frequency of the transverse excitation (Fig. 3 Panels II and IV). This originates from the mixing of transverse and longitudinal dynamics. The endorsement comes from the fact
that the shoulder disappears when the transverse contribution (Σ_{LT}(q, ω)) is removed from the longitudinal self-consistent
energy. Furthermore, depending on the disorder parameter
and transverse to longitudinal phase velocity ratio, a hump in
the wavevector trend (the stronger as the bigger it is the
former and the smaller the latter) is observed. It can be seen
to be a prolongation of the q^4 behavior observed in the Rayleigh
regime. The hump corresponds to a rapid increase of the
phase velocity and has been reported in theoretical charac-
terization of elastic waves in polycrystalline aggregates (28).
This latter fact emphasizes how in the case of longitudinal acoustic dynamics the coupling of polarizations contributes to the attenuation increase observed at the edge of the Rayleigh
region (qa ≈ 1). A scalar model thus can underestimate the
attenuation observed in this wavectors region in real systems.
(3) In the high wavevectors region, qa > 1, the co-existence of
two excitations can be observed (10, 28, 37).

This general picture is coherent with the experimental
characterization of real systems found in the literature
(11, 13, 17, 18). The contrast between theoretical and experimental results obtained for the [C8MIM]Cl glass shows
an excellent agreement in the whole measured wavectors.
range, as attested in Fig. 4 Panels I-IV. In particular the following experimental features are quantitatively reproduced by the theory: (i) the Rayleigh anomalies; (ii) the increase of attenuation and phase velocity beyond the Rayleigh region (i.e. at frequencies higher than BP), which can be attributed to the strong coupling between transverse and longitudinal dynamics (see point (2)), in turn related to the strong intensity of elastic fluctuations; (ii) the presence of the low-frequency shoulder in $S_L(q, E)$, related to the mixing of longitudinal and transverse polarization; (iii) the position of the BP. Such a quantitative agreement cannot be achieved by using the Born approximation. In Fig. 5 we report the features of transverse dynamics obtained for the same input parameters of the longitudinal dynamics. In the low-wavevector region we observe a single excitation, showing: i) a crossover in the $q$-trend of the attenuation at frequency where the BP shows up; ii) a softening of the phase velocity in the Reileigh regime; iii) the Ioffe-Regel crossover at wavevector and frequency point where $\pi \Gamma$ becomes larger than $\Omega$, occurring near the BP-frequency. At higher wavevector a high-frequency shoulder appears in the calculated dynamic structure factors. This is not entirely related to the mixing of polarizations. It is indeed partially preserved when the term $\Sigma_T L$ is fixed to zero. These results are in qualitative agreement with results from simulations in glasses (7, 14) or liquids at high-wavevectors (22, 23).

Our results shows that the mixing of polarization can be generated by disorder. Beyond Rayleigh anomalies an approximate solution of the Dyson equation can quantitatively account also for this latter phenomenon.

Materials and Methods

The Inelastic X-ray and Neutron Scattering experiments. The IXS experiment was carried out at the ID28 beamline of ESRF. The data were collected in the $Q$-range [1 - 15] nm$^{-1}$ with a (quite small) $Q$-step of about 0.4 nm$^{-1}$. The experiment was performed at 23.725 keV, using the silicon (12, 12, 12) reflection providing an overall energy resolution of about 1.4 meV (FWHM), which was determined from a plexiglass slab at $T=10$ K. The $Q$-resolution was set to 0.18 nm$^{-1}$ for the first 5 points (i.e. 1.0 nm$^{-1}$, 1.4 nm$^{-1}$, 1.8 nm$^{-1}$) and fixed to 0.37 nm$^{-1}$ for the remaining $Q$'s. The energy spectra were collected in the $\sim$30 to 30 meV $E$-range. The sample cell was an Al tube of 5 mm length capped with two oriented monocrystal diamond windows (0.5 mm thick). The [C8MIM][Cl] sample was loaded in the sample cell in an Ar-filled glove box to avoid water contamination. A cryostat was used to cool the sample to $T = 176.4$ K, down to the glassy state ($T_g = 214$ K). The background contribution from the sample environment was measured in the same experimental configuration as the sample and subtracted from the data after normalization for the [C8MIM][Cl] X-ray absorption coefficient. The IXS signal reproduce quantitatively the experimental signal, $I_{EXS}(E) = A(Q)E^\delta I_{DHO}(E)$, where $\delta$ is the Bose factor and $A(Q)$ is an overall intensity factor. $I_{EXS}(E)$ is convoluted with experimental energy resolution function prior to comparison with data. The $S_L(q, \omega)$ has been modeled with the expression $S_L(q, \omega) = \sum_{\eta=1,2} I_{DHO}(\eta)^{\frac{1}{2}}(\eta)^{\frac{1}{2}} + \eta \delta(\omega)$. The INS experiment was performed with the MARI instrument at the ISIS spallation neutron source. The sample was loaded in an Al made cylindrical annular can with a thickness corresponding to sample transmission of 70%. It was cooled down to $T=725$ K by using a cryostat. The incident neutron energy was 15 meV. The empty cell contribution was measured and subtracted from the data after normalization for the sample neutron absorption coefficient. In order to remove the Q-dependence of the measured data, they were integrated over $Q$ in the largest range available from the experimental configuration, i.e. $20 < Q < 40$ nm$^{-1}$. From the integrated measured signal, $I_{INS}(E)$, we obtain $\frac{E}{I_{INS}(E)} = A(QE)(\pi^2/2)$, being $A$ an intensity constant.

The Generalized Born Approximation and analysis of theoretical outputs. The bare Green propagator can be formally written as $G^0_k(q, \omega) = \lim_{\nu \rightarrow 0^+} \frac{1}{2\pi} \int_{-\infty}^{+\infty} dw e^{iq\cdot w} p.v.\left\{\frac{1}{w^2-(\nu\eta^2)q^2}\right\} - i\eta \frac{\delta(\nu\eta^2)}{(\nu\eta^2)^{\frac{1}{2}}} - i\eta \frac{\delta(q)}{\eta^2} \theta(q^2 - (\nu\eta^2)^2)$, where $\eta$ is a positive real variable, the symbol p.v. states for the Cauchy principal value and $\theta$ is the sign function. The self-consistent set of equations defining $G(q, \omega)$ and $\Sigma(q, \omega)$, Eqs. 1 and 3, is equivalent to a continued-fraction representation, e.g. $\Sigma_j(q, \omega) = \lim_{\nu \rightarrow 0^+} L_{1,j} q^2 \sum_{n=1}^{\infty} \left\{\frac{1}{\nu^2 - \nu^2 \omega^2 - \nu^2 \Delta q^4 \omega^4(q) - \nu^2 \Delta q^4 \omega^4(q)}\right\}$ [5].

The complex variable $\omega_0$ is equal to $\omega + i\eta$. In the following, for sake of clarity, it will be shortly marked as $\omega$ and the limit operation will be only implicitly assumed. By relying on Eq. 5 we find an approximate expression for $\Sigma_j(q, \omega)$ by including corrective terms to the Born approximation, which constitutes the first step of the SCBA. To this end the continued fraction representation is truncated to the second order, $\Sigma_j(q, \omega) = L_{1,j} q^2 \sum_{n=0}^{\infty} \left\{\frac{\nu^2 \Delta q^4 \omega^4(q)}{\nu^2 q^2 + \nu^2 \Delta q^4 \omega^4(q)}\right\}$

The expression in curly bracket, $<G_0^0(q, \omega)>, 1$, is then formally expanded in Taylor series with respect to $\frac{\Delta q^4 \omega^4(q)}{\nu^2 q^2}$ in a bounded domain of the wavevector-frequency plane, $(0, \omega_{Max}) \times (0, q_{Max})$. In this domain the power series $\sum_{n=0}^{\infty} \left\{\frac{\nu^2 \Delta q^4 \omega^4(q)}{\nu^2 q^2 + \nu^2 \Delta q^4 \omega^4(q)}\right\}$ converges almost everywhere (a.e.) to $<G_0^0(q, \omega)>(36)$. By grounding on this result, it can be demonstrated that the second-step self-energy, i.e. $L_{1,j} <\Delta q^4 \omega^4(q)>, 1$, can be approximated by the series $\sum_{n=0}^{\infty} \left\{\frac{\nu^2 \Delta q^4 \omega^4(q)}{\nu^2 q^2 + \nu^2 \Delta q^4 \omega^4(q)}\right\}$ within a negligible error (36) for frequencies inside and wavevectors well inside such a domain. It is $\Delta q^4 \omega^4(q) = \Delta q^4 \omega^4(q)(\theta(q - q_{Max}), 0)\cdot \theta(q - q_{Max})$, where $\theta(q)$ is...
the Heaviside function. For these wavevectors and frequencies, the iterative sequence, Eq. 5, is convergent (36). This ensures that the truncation to the second order of the iteration procedure provides an approximate expression for the self-consistently defined mass operator. We truncate the Taylor series to the first order, obtaining

\[ \Sigma_j(q, \omega) \simeq \delta_{1jk} \left\{ \frac{1}{\epsilon^2 k^2} + \frac{1}{\epsilon^2 k^2} \frac{q^2}{2} \Delta \Sigma^{(1)}_k(q, \omega) \right\} \]

where \( \delta_{1jk} \) is the Kronecker delta. Assuming \( \eta \) is relevant to our interest, \( q = O(a^{-1}) \), does not significantly enlarge the error done in the approximation described in Eq. 4, while facilitating the analytical calculation (36). Specifics of calculations are reported in Supporting Information.

Characteristic frequencies, broadenings and intensities are derived from the calculated \( S_L(T)(q, \omega) \) and \( S_D(T)(q, \omega) \) using the VDOS is obtained through the expression:

\[ \rho(E) = \frac{2}{\pi^2 k} \int_0^{2\pi} dq |\Sigma_L(q, \omega) + 2G_T(q, \omega)|, \]

where \( q = \delta D \) is the Debye wavevector (we assume \( Q_0 = \Omega_0 \)). The value of the edge of the pseudo-Brillouin zone, \( Q_0 = 17.9 \text{ nm}^{-1} \), is experimentally determined trough the longitudinal acoustic dynamics dispersion.

In Eq. 1 the frequency of the bare Green dyadic \( G^0_k(q, \omega) \) is normalized via the expression \( \omega = \epsilon^2 \frac{q}{2\pi} \sin(\frac{\pi q}{\Omega_0})^{-1} \) to account for the bending of acoustic dispersion related to the occurrence of the pseudo-Brillouin zone (29). The possible broadening of the acoustic excitations due to the existence of a distribution of nearest-neighbor values is taken under account, similarly to what is done in Ref. (9). Detailed procedure is specified in Supporting Information.

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Appendix A

Expressions of the longitudinal and transverse self-energies in the Generalized Born Approximation. We derive the expressions of the longitudinal and transverse dynamic structure factors in the Generalized Born Approximation, by exploiting Eq. 4 in the main text. We re-write it here for sake of clarity,

\[ \Sigma_j(q, \omega) \equiv \lim_{\eta \to 0^+} \delta_{1jk} \left\{ \frac{1}{(q^2 + \epsilon^2 k^2)\eta} + \frac{1}{(q^2 + \epsilon^2 k^2)\eta} \right\} \Delta \Sigma^{(1)}_k(q, \omega) \]

where \( \eta \) and \( \tilde{\eta} \) are related to each other by the relationship \( \tilde{\eta} = \epsilon \eta \).

We recall that \( L_1 = \epsilon^2 q^2 L_1, \Delta \Sigma^{(1)}_k(q, \omega) = \Sigma^{(1)}_k(q, \omega) - \Sigma^{(1)}_k(q, 0) = 0, \) \( \Sigma^{(1)}_k(q, 0) = L_0 G^0(q, \omega), \) \( q_0k = \tilde{\epsilon} \) and \( \tilde{c}_k = (\epsilon^2 + \epsilon^2 \hat{\Sigma}^{(1)}_k(q = 0, \omega = 0))^{1/2} \) is the macroscopic velocity of the (first step) perturbed medium. Under the hypothesis of local isopycnic, the elastic tensor, \( C_{ijkl}(r) \), vary in the space by following the expressions

\[ C_{ijkl}(r) = \tilde{C}_{ijkl}(1 + \delta \tilde{C}_{ijkl}(r)) = \]

\[ = \lambda(1 + \delta \lambda(r))\delta_{ij}\delta_{kl} + \mu(1 + \delta \mu(r))\delta_{ik}\delta_{jl} + \delta \sigma_{ijkl}, \]

where \( \lambda, \mu, \) and \( \tilde{\lambda}, \tilde{\mu}, \tilde{\sigma} \) are the shear modulus and Lamé parameter of the bare medium and \( \delta \lambda = \frac{\Lambda}{\mu}, \delta \mu = \frac{\Lambda}{\mu}, \delta \sigma \). The operator \( L_1 \) in the Fourier space is defined as

\[ \Sigma^{(1)}_j(q, \omega) = L_1 \delta_{1jk} C^{(1)}_j(q, \omega) = \]

\[ = \int d^3 q s q_0 q_0 s_R \tilde{C}_{ijkl}(q, s) G^0_k(s, \omega) \]

is a wavevector and the integral extends to \( \mathfrak{M} \). The function \( R_{\alpha\beta\gamma\delta}(q) \) is the Fourier transform of the covariance of the elastic tensor fluctuations, \( R_{\alpha\beta\gamma\delta}(r) = \phi_1(r_1 - r_2) \Rightarrow \)

\[ R_{\mu\nu}(q) = \epsilon^2 \frac{q^2}{2} \Delta \Sigma^{(1)}_k(q, \omega) \]

where \( q = |q| \) and \( \int d^3 q R_{\mu\nu}(q) = 1 \).

The bare Green’s dyadic, ensemble average Green’s dyadic and self-energy in the orthonormal basis defined by the direction of wave propagation, \( i \), and the two orthogonal ones, can be written as

\[ G_i^0(q, \omega) = \eta_{ii}(q, \omega) + \eta_{ij}(q, \omega)(I - \eta_{ii}); \]

\[ \Sigma_i(q, \omega) = \eta_{ii}(q, \omega) + \eta_{ij}(q, \omega)(I - \eta_{ii}); \]

\[ < G_i(q, \omega) > = \eta_{ii}(q, \omega) + \eta_{ij}(q, \omega)(I - \eta_{ii}); \]

\[ \eta_{ii}(q, \omega) = \eta_{ii}(q, \omega) + \eta_{ij}(q, \omega)(I - \eta_{ii}); \]

\[ < g_L(q, \omega) > = \frac{1}{g_L(T)(q, \omega)} - \Sigma_{LL}(T)(q, \omega). \]

The longitudinal and transverse average wave speeds are defined respectively by the relations:

\[ c_L^2 = \frac{\lambda + 2\mu}{\mu}; \quad c_T^2 = \frac{\lambda}{\mu} \]

where \( \mu \) is the shear modulus, \( \lambda = \frac{\mu}{\lambda}(\lambda + 2\mu), \)

\[ \Sigma_{LL}(T)(q, \omega) + \Sigma_{LT}(T)(q, \omega); \]

\[ \Sigma_{LL}(T)(q, \omega) + \Sigma_{LT}(T)(q, \omega). \]

Each partial term of the self-energies, \( \Sigma_{LL}(T)(q, \omega) \) and \( \Sigma_{LT}(T)(q, \omega) \) in the Generalized Born Approximation is composed of two terms,

\[ \Sigma_{LL}(T)(q, \omega) = \Sigma_{LL}(T)(q, \omega) + \Sigma_{LT}(T)(q, \omega); \]

\[ \Sigma_{LT}(T)(q, \omega) + \Sigma_{LT}(T)(q, \omega). \]

We outline in the following the respective expressions for each polarizations coupling,

\[ \frac{1}{\epsilon^2 \rho} \frac{1}{(q^2 + \epsilon^2 k^2)\eta} \]

\[ \frac{1}{\epsilon^2 \rho} \frac{1}{(q^2 + \epsilon^2 k^2)\eta} \]

\[ \frac{1}{\epsilon^2 \rho} \frac{1}{(q^2 + \epsilon^2 k^2)\eta} \]

\[ \frac{1}{\epsilon^2 \rho} \frac{1}{(q^2 + \epsilon^2 k^2)\eta} \]
By using spherical coordinates in Eqs. A8-A15, it is obtained
\[ \Sigma_{L}(q,\omega) = e^{2q^2} \int_{-\frac{1}{2}}^{\frac{1}{2}} dx 4\pi a^{-1} I_{L}(0)(q,\omega, x), \]  
[A16]

\[ \Sigma_{L}(q,\omega) = e^{2q^2} \int_{-\frac{1}{2}}^{\frac{1}{2}} dx 4(1-x^2)x^2 \frac{1}{\pi} a^{-1} I_{L}(0)(q,\omega, x), \]  
[A17]

\[ \Sigma_{L}(q,\omega) = e^{2q^2} \int_{-\frac{1}{2}}^{\frac{1}{2}} dx (1-3x^2+4x^4) \frac{1}{\pi} a^{-1} I_{L}(0)(q,\omega, x), \]  
[A18]

\[ I_{k}^{(n)}(q,\omega, x) = \lim_{\eta \to 0} \int_{0}^{\infty} ds s^2 \tilde{\Phi}_{\mu}(q, s, x) \left[ \frac{s^2 \Delta \Sigma_{1}^1(0,\omega+i\eta)}{(\eta+h+i\eta)^2 - s^2)^{n+1}} \right] = \# \int_{0}^{\infty} ds s^2 \tilde{\Phi}_{\mu}(q, s, x) \left[ \frac{s^2 \Delta \Sigma_{1}^1(0,\omega)}{(\eta+h+s)^2 - s^2)^{n+1}} \right] + \]  
[A20]

The symbol \# states for the Hadamard finite part integral (equal to the Cauchy principal value when \( n = 0 \)). The quantities \( \Delta \Sigma_{1}^1(T)(q,\omega) = \Sigma_{1}(T)(q,\omega) - \Sigma_{1}(T)(0, 0) \), which appear in the expressions of \( \Sigma_{L}(L,T) \) and \( \Sigma_{L}(T,L) \) are related to the first step self-energies, coincident with the self-energies obtained through

\[ \Sigma_{L}(q,\omega) = \lim_{\eta \to 0} \int q^2 \delta d^3 s \tilde{\Phi}_{\mu}(q, s, x) \left[ \frac{1}{(\eta+h+i\eta)^2 - s^2)^{n+1}} \right] = \int_{-\frac{1}{2}}^{\frac{1}{2}} dx 4\pi a^{-1} \frac{1}{\pi} \left[ \frac{\delta}{(\eta+h+i\eta)^2 - s^2)^{n+1}} \right] \cdot 3/2 + \frac{(qz+z)\hat{z}}{\eta_0 L - (qz+z)\hat{z}} \]  
[A23]

By comparing Eqs. A21 and A22 with Eqs. A8, A10, A12, A14 it is immediate to verify that \( \Sigma_{L}^1 \) is equivalent to \( \Sigma_{L}^0 \) under the transformation \( \ell_{L}(T) \to c_{L}(T) \).

\[ \# \int_{0}^{\infty} ds s^2 \tilde{\Phi}_{\mu}(q, s, x) \left[ \frac{s^2 \Delta \Sigma_{1}^1(0,\omega)}{(\eta+h+s)^2 - s^2)^{n+1}} \right] = p.v. \int_{0}^{\infty} ds \frac{1}{n!} (-1)^{n+1} \frac{1}{(\eta-h-s)^{n+1}} \left[ s^2 \tilde{\Phi}_{\mu}(q, s, x) \right], \]  
[A24]

The Hadamard finite part integral exists because it exists the Cauchy principal value of the integral on the right side of Eq. A24, since it is possible to demonstrate (36) that the integrand satisfy the Lipschitz property. The integral in Eq. A24 can be calculated by exploiting the Residue Theorem because the function

\[ \Sigma_{L}(q,\omega) = e^{2q^2} \int_{-\frac{1}{2}}^{\frac{1}{2}} dx 4(1-x^2)x^2 \frac{1}{\pi} a^{-1} I_{L}(0)(1)(q,\omega, x), \]  
[A19]

where \( x = \cos(\theta) \) and \( \theta \) is the \( \hat{r} \) angle \( \hat{r}_{L}^{(n)}(q,\omega, x) \), where \( n = 0, 1, ... \), is an integral which remains implicitly defined in the previous equation. By exploiting the Sokhotski-Plemelj-Fox theorem (39), integration by part and the Cauchy’s Residue Theorem the integral \( I_{k}^{(n)}(q,\omega, x) \) for a generic polarization \( k \), is given by (36)

**Footnote:** The x-integration is performed numerically.
We also exploit the fact that the integrand function is even with respect to \( x \), thus

\[
\frac{1}{\pi} \int_{-\infty}^{\infty} dx \ldots = \frac{1}{\pi} \int_{0}^{\infty} ds = \ldots
\]

with \( a(q,x) = \sqrt{q^2(1 - x^2)} + a^2 \), \( R_{\lambda q}(n) (p) \) is the residue of the integrand of \( I^{(n)} \) around the pole \( p \) of order \( m \). The first term in Eq. A25 is related to the Hadamard finite part integral in Eq. A20, whereas the second term to the second term in Eq. A20.

Appendix B

Bending of acoustic wave dispersion and definition of the edge of the pseudo-Brillouin zone

It has been shown that the short-range structure of glasses preserves residual order that characterizes the long-range structure of crystals (8). The most evident consequence is the occurrence in glasses of a pseudo-Brillouin zone whose extension depends on nearest-neighbour average distance. In the present model, the occurrence of the pseudo-Brillouin zone with the consequent bending of the acoustic waves dispersion is empirically accounted by superimposing it to longitudinal and transverse acoustic dispersions calculated in the framework of HET. The value of the edge of the first pseudo-Brillouin zone, \( Q_0 = 17.9 \, \text{nm}^{-1} \), is determined by the experimental longitudinal acoustic dispersion. The frequency of the bare Green dyadic \( G^0(q,\omega) \) in the Dyson equation is thus normalized, as specified in the main text, i.e.

\[
\bar{\omega} = \frac{Q_0}{\pi} \sin \left( \frac{\pi}{Q_0} \right)^{-1}.
\]

It is possible to introduce a distribution function for the values of the pseudo-Brillouin edge related to the nearest-neighbor values distribution function. This latter can be empirically extrapolated from the atomic-form-factors-weighted static structure factor measured by X-ray Scattering, similarly to what is done in Ref. (9). The main peak in \( S(Q) \) is first shaped by a Lorentz function centered in \( Q = 17.9 \, \text{nm}^{-1} \), i.e. at the edge of the first pseudo-Brillouin zone experimentally determined. The FWHM of the Lorentz function is fixed such that the area of the main peak, i.e. the area of the static structure factor in between 13 and 24 \( \text{nm}^{-1} \), is equal to the total area of the Lorentz function, each function being normalized to their respective maxima. By using the Lorentzian shaped nearest-neighbour values distribution function and the dispersion relation given in Eq. B1 the distribution function of the values of the pseudo-Brillouin zone edge can be given in turn by a Lorentz function. At each wavevector the calculated dynamic static structure factor is normalized simultaneously, the VDOS in the whole measured energy range. The presence of IVMs also influences the broadening of the longitudinal and transverse current at frequencies higher than BP frequency, as well as the position of BP in VDOS. It cannot, however, account for the mixing of polarization, which can be instead described by the RMT. We furthermore stress that taking under consideration only the effect of the coupling of acoustic modes with IVMs (completely neglecting the effect of the heterogeneous elastic structure on the acoustic dynamics described in the framework of RMT) permits to achieve a reliable description of the experimentally observed feature in \( S_L(Q,E) \) only at those frequencies of acoustic excitations near the characteristic frequency of IVMs. By looking at Fig. 5 in the main text, one can think that the broadening of the transverse current could be underestimated in the present approximation at high wavevectors. This can depend on the fact that we neglected there a possible coupling with IVMs. Actually this latter effect, as it is possible to deduce by the observation of Fig. 6, leads to larger values of the transverse broadenings.

Appendix C

Coupling of acoustic waves with intermolecular vibrational modes

It is possible to introduce intermolecular vibrational modes (IVMs) coupled to acoustic waves. The self-energy operator in this case becomes \( \Sigma = \Sigma_{\text{MRT}} + \Sigma_{\text{IVM}} \), where it is added to the self-energy calculated by using the Random Media Theory in the Generalized Born Approximation, \( \Sigma_{\text{MRT}} \), the term \( \Sigma_{\text{IVM}} = \sum_{q} \frac{q^2 A^2_{\text{IVM}}(q)}{\omega^2 - \omega^2_{\text{IVM}} + i \omega \Gamma_{\text{IVM}}} \). The coupling of IVM’s with acoustic waves is treated to lowest \( T^0 \) (5). The dynamic structure factor of IVM (taking into account also its coupling with the acoustic modes) and the characteristic frequency and attenuation \( \omega_{\text{IVM}}(Q) \) are given by:

\[
\omega_{\text{IVM}}(Q) = \sqrt{\omega_{\text{IVM}}^2(Q) + \Sigma_{\text{IVM}}(Q,E)} - \omega_{\text{IVM}}(Q) = \sqrt{\omega^2_{\text{IVM}}(Q) + \Sigma_{\text{IVM}}(Q,E)} - \omega_{\text{IVM}}(Q) \]

\[
g_{\text{IVM}}(E) = \sum_{q} \frac{2}{\pi D} \int_{D} dq S_{\text{IVM}}(q,\omega),
\]

where \( P = L, T_1, T_2 \) is the polarization VDOS (longitudinal and transverse degenerate).

The high-frequency region of the measured VDOS can be described only after the introduction of IVM. Their occurrence has been observed by Raman scattering (30). To cope with literature data we introduce two IVM with respectively characteristic frequency and attenuation \( \omega^2_{\text{IVM}}(Q) = 7.2(10) \, \text{meV} \) and \( \Sigma_{\text{IVM}}(Q,E) = 4.0(3.8) \, \text{meV} \). In Fig. 6 the results are compared with experimental findings. By taking into account also for the presence of IVM it is possible to quantitatively reproduce the feature related to measured \( S_L(Q,E) \) in the first pseudo-Brillouin zone and, simultaneously, the VDOS in the whole measured energy range. The presence of IVMs also influences the broadening of the longitudinal and transverse current at frequencies higher than BP frequency, as well as the position of BP in VDOS. It cannot, however, account for the mixing of polarization, which can be instead described by the RMT. We furthermore stress that taking under consideration only the effect of the coupling of acoustic modes with IVMs (completely neglecting the effect of the heterogeneous elastic structure on the acoustic dynamics described in the framework of RMT) permits to achieve a reliable description of the experimentally observed feature in \( S_L(Q,E) \) only at those frequencies of acoustic excitations near the characteristic frequency of IVMs. By looking at Fig. 5 in the main text, one can think that the broadening of the transverse current could be underestimated in the present approximation at high wavevectors. This can depend on the fact that we neglected there a possible coupling with IVMs. Actually this latter effect, as it is possible to deduce by the observation of Fig. 6, leads to larger values of the transverse broadenings.

\[a(q,x) = \sqrt{q^2(1 - x^2)} + a^2, R_{\lambda q}(n) (p)\]

\[\frac{1}{\pi} \int_{-\infty}^{\infty} dx \ldots = \frac{1}{\pi} \int_{0}^{\infty} ds = \ldots\]

We also exploit the fact that the integrand function is even with respect to \( x \), thus

\[
\int_{-\infty}^{\infty} ds = \ldots = \frac{1}{2} \int_{0}^{\infty} ds = \ldots
\]

\[\Sigma_{\text{IVM}} = \sum_{q} \frac{q^2 A^2_{\text{IVM}}(q)}{\omega^2 - \omega^2_{\text{IVM}} + i \omega \Gamma_{\text{IVM}}}.
\]

\[g_{\text{IVM}}(E) = \sum_{q} \frac{2}{\pi D} \int_{D} dq S_{\text{IVM}}(q,\omega),
\]

\[\omega_{\text{IVM}}(Q) = \sqrt{\omega_{\text{IVM}}^2(Q) + \Sigma_{\text{IVM}}(Q,E)} - \omega_{\text{IVM}}(Q) = \sqrt{\omega^2_{\text{IVM}}(Q) + \Sigma_{\text{IVM}}(Q,E)} - \omega_{\text{IVM}}(Q) \]

\[\omega_{\text{IVM}}(Q) = \sqrt{\omega_{\text{IVM}}^2(Q) + \Sigma_{\text{IVM}}(Q,E)} - \omega_{\text{IVM}}(Q) = \sqrt{\omega^2_{\text{IVM}}(Q) + \Sigma_{\text{IVM}}(Q,E)} - \omega_{\text{IVM}}(Q) \]

\[g_{\text{IVM}}(E) = \sum_{q} \frac{2}{\pi D} \int_{D} dq S_{\text{IVM}}(q,\omega),
\]

\[\omega_{\text{IVM}}(Q) = \sqrt{\omega_{\text{IVM}}^2(Q) + \Sigma_{\text{IVM}}(Q,E)} - \omega_{\text{IVM}}(Q) = \sqrt{\omega^2_{\text{IVM}}(Q) + \Sigma_{\text{IVM}}(Q,E)} - \omega_{\text{IVM}}(Q) \]

\[g_{\text{IVM}}(E) = \sum_{q} \frac{2}{\pi D} \int_{D} dq S_{\text{IVM}}(q,\omega),
\]
Fig. 6. Characteristic features of longitudinal and transverse dynamics obtained by taking into account both the heterogeneous elastic structure of the medium by exploiting the Generalized Born Approximation described in the text and the occurrence of IVMs. LONGITUDINAL DYNAMICS. Panel I. Phase velocity of inelastic excitations observed in experimental (circles with error bars) and calculated (stars) longitudinal dynamic structure factors. Panel II. Ratio of intensities of the DHO functions modeling the two high-q excitations obtained from experimental (circles with error bars) and calculated (stars) longitudinal dynamic structure factors. Panel III. Broadening of inelastic excitations observed in experimental (circles with error bars) and calculated (stars) longitudinal dynamic structure factors. Panel IV. Experimental (black lines) and calculated (red stars) reduced VDOS, $g(E)/E^2$. Full line shows the components of the reduced VDOS related to acoustic modes and the dashed lines to the two IVMs. TRANSVERSE DYNAMICS. Panel a) Broadening, $\Gamma$, multiplied by $\pi$ (open circles) and characteristic frequency, $\Omega$, of the transverse low-frequency inelastic excitation obtained from the calculated transverse dynamic structure factor. Panel b) Phase velocity of the two inelastic excitations observed in the calculated $S_{2\Omega}(q, \omega)$. 

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