Anharmonic elasticity theory for sound attenuation in disordered solids with fluctuating elastic constants

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Using anharmonic elasticity theory in the presence of spatially fluctuating elastic constants we derive a self-consistent theory for sound attenuation in disordered solids. In the low-frequency regime (below the boson peak frequency) we obtain a sound attenuation law proportional to $T\omega^2$, where T is the temperature and ω is the frequency. Together with the usual Rayleigh scattering mechanism this yields a crossover of the Brillouin linewidth from a ω^2 to a ω^4 regime. The cross-over frequency is fully determined by the boson peak frequency and the temperature. For network glasses like SiO_2 at room temperature this crossover is predicted to be situated one order of magnitude below the boson peak frequency.

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I. INTRODUCTION

The role of the phonon-phonon interaction in crystalline solids is very well understood. It accounts for a finite thermal conductivity, thermal expansion, as well as transverse ultrasound attenuation.[1,2] Landau and Rumer[3] realized that, due to energy and momentum conservation, a decay of the low-frequency density waves (longitudinal phonons) would be phase-space suppressed. In their theory only transverse phonons acquire a lifetime $\tau_T \propto \omega^{-1}$ at GHz frequencies. Akhiezer[4] solved the corresponding Boltzmann equation which results in a $\tau_T \propto 1/\omega^2 T$ behavior at substantially lower frequencies.[5]

The hydrodynamic modes are at the focus of interest in glass physics, because their physical properties appear to be independent of the specific underlying nonequilibrium static structure. Recent experiments showed the presence of an Akhiezer-like sound attenuation regime in the longitudinal channel,[6,7] however these authors claim different values of the cross-over frequency from the Akhiezer-like to the Rayleigh law. The authors refer to the old and frequently cited theory of Akhiezer.[4] However neither his approach nor that of Rumer/Landau[3] do apply, as they are only valid for the *low-frequency transverse* modes of *crystalline* solids. Fabian and Allen[8] performed simulations on a disordered lattice in the kinetic regime, they established the validity of an Akhiezer-like law for the longitudinal modes as well. In our analytical approach, it turns out that this observation is only possible in presence of disorder. In addition, our calculation is not limited to the kinetic regime. A treatment of anharmonic damping in disordered solids has been formulated some time ago,[9] but the focus was on weak and strong localization, so the applicability range is at very high frequencies above the boson peak.

We present an extension of the "fluctuating elastic constants" model,[11–14] to account for anharmonic sound damping of disordered solids at "low" frequencies (i.e., in the GHz-THz regime). This is achieved, using the fact, that the disorder-averaged spectral function $\chi''(\mathbf{q},\omega)$—which describes the vibrational spectrum of the disordered material—does not reflect momentum conservation anymore and exhibits a characteristic rapid increase in the THz frequency regime.[11–14] This phenomenon is frequently called "boson peak" (BP). The interaction of the irregular states near the BP produces, as we shall show an Akhiezer-like $T\omega^2$ law for the anharmonic sound attenuation. The prefactor is intimately related to the disorder-induced elasticity fluctuations, which produce the BP.

This paper is organized as follows: first we briefly review the theory for the spectrum of a harmonic disordered solid based on disorder-modified elasticity theory.[11–14] Then we introduce anharmonic interactions. Using standard quantum field-theoretical techniques we derive self-consistent mode-coupling equations, obtained from a saddle-point treatment of the disorder-averaged replica field theory,[10,15,16] which determine the phonon spectrum. For low enough temperatures and frequencies the nonself-consistent mode-decay approximation is sufficient, which results in the Akhiezer-like $\Gamma \propto T\omega^2$ law.

II. ANHARMONIC ELASTICITY THEORY WITH DISORDER

We start by reviewing the elasticity theory for a disordered solid. The starting point is Lamé's elasticity theory,[17] but with spatially fluctuating elastic constants. The dynamics of the displacement field $\mathbf{u}(\mathbf{x},t)$ is determined by the Lagrange density

$$\mathcal{L} = \frac{\rho}{2}\dot{\mathbf{u}}(\mathbf{x},\mathbf{t})^2 - \frac{1}{2}\lambda(\mathbf{x})\left(\sum_i u_{ii}(\mathbf{x},t)\right)^2 - \mu(\mathbf{x})\sum_{ij} u_{ij}(\mathbf{x},t)^2, \tag{1}$$

in which $u_{ij}=(\partial_i u_j+\partial_j u_i)/2$ is the usual strain tensor. Here λ and μ are Lamé's elastic constants and ρ is the mass density. As in our previous work we assume that the disorder affects the elastic properties only via μ:

$$\mu(\mathbf{x}) = \mu_0 + \delta\mu(\mathbf{x}) \quad \mu_0 = \langle\mu(\mathbf{x})\rangle,$$

$$\lambda(\mathbf{x}) = \lambda_0 = \langle\lambda(\mathbf{x})\rangle, \tag{2}$$

i.e., the spatial fluctuations of λ are neglected. The fluctuations $\delta\mu(\mathbf{x})$ are characterized by a correlation function $\langle\delta\mu(\mathbf{x})\delta\mu(\mathbf{x}')\rangle=K_\mu(|\mathbf{x}-\mathbf{x}'|)$; Higher moments of $\delta\mu(\mathbf{x})$ are discarded. The correlation function is given in terms of the variance of the shear modulus Δ^2 and the correlation length ξ (Ref. 18)

$$K_\mu(|\mathbf{x}-\mathbf{x}'|) = \Delta^2 \exp\left(-\frac{|\mathbf{x}-\mathbf{x}'|}{\xi}\right), \tag{3}$$

and has the Fourier transform

$$K_\mu(\mathbf{k}) = \int \mathrm{d}^3\tilde{\mathbf{x}} e^{-i\mathbf{k}\tilde{\mathbf{x}}} K_\mu(\tilde{\mathbf{x}}) = \frac{8\pi\xi^3\Delta^2}{[1+(k\xi)^2]^2}. \tag{4}$$

The quantities Δ^2 and ξ are the two phenomenological parameters, which characterize our model. As noted in,[11–14] there exists a critical amount of disorder Δ_c^2, depending only on the ratio of the squared sound velocities $(v_L/v_T)^2$, beyond which the system becomes unstable. This criticality arises, because a too large variance Δ^2 allows for negative values of the shear modulus $\mu(\mathbf{x})$, which is unphysical.

Performing standard field theoretical methods[10,16] the following set of self-consistent equations for the disorder-averaged longitudinal ($i=L$) and transverse ($i=T$) dynamical susceptibilities has been derived:[11–14]

$$\chi_i(q,z) = \frac{q^2}{-z^2 + q^2[v_{i,0}^2 - \Sigma_{i,\mathrm{dis}}(z)]}, \tag{5}$$

where $z=\omega+i\epsilon$ and $\epsilon\to+0$. Because only fluctuations of μ have been considered, we have $\Sigma_{L,\mathrm{dis}}=2\Sigma_{T,\mathrm{dis}}$. $\Sigma_{T,\mathrm{dis}}(z)$ is given by

$$\Sigma_{T,\mathrm{dis}}(z) = \frac{1}{V}\sum_{\mathbf{k}} \frac{K_\mu(\mathbf{k})}{\rho^2}[\chi_L(k,z) + \chi_T(k,z)]. \tag{6}$$

These mean-field equations arise from a saddle point within an effective replica field theory. The self-energy is evaluated in the long wavelength limit $\Sigma_{T,\mathrm{dis}}(z)=\Sigma_{T,\mathrm{dis}}(\mathbf{q}\to 0,z)$, in agreement with similar approximation schemes, e.g., the coherent potential approximation or dynamical mean field theory (DMFT).[19] The dynamical structure factor then follows from the fluctuation-dissipation theorem

$$S(q,\omega) = \frac{1}{\pi}\frac{1}{1-e^{\beta\omega}}\mathrm{Im}\{\chi_L(q,z)\}. \tag{7}$$

with $\beta=\hbar/k_BT$. The real part of the self energies $\Sigma_{i,\mathrm{dis}}$ renormalizes the sound velocities $v_i^2=v_{i,0}^2-\Sigma'_{i,\mathrm{dis}}(\omega)$ and their imaginary parts give the (harmonic) sound attenuation. In particular, the Brillouin linewidth of $S(q,\omega)$ is given by Ref. 12,

$$\Gamma(k) = \frac{\omega_k}{v_L^2}\Sigma''_{L,\mathrm{dis}}(\omega=\omega_k) \quad \omega_k = v_L k. \tag{8}$$

In the case of a short correlation length the function $K_\mu(k)$ is constant in the relevant wave-number range and one recovers the *uncorrelated* version of the self-consistent Born approximation (SCBA),[11,12] which has been shown to explain the excess density of states (DOS) [boson peak (BP)] and the dip in the temperature dependence of the thermal conductivity as a consequence of strong disorder-induced scattering. The Brillouin linewidth has been shown to vary as ω^2 in the BP regime and to be related to the excess DOS.[12] In the case of a *finite* correlation length ξ the ratio between DOS and Debye DOS is not limited to a factor 2 (see Ref. 12) but increases indefinitely with ξ.[13,14] Within this description (uncorrelated *and* correlated) the BP marks the crossover of the vibrational spectrum from a weakly scattered plane-wave regime to a disorder-dominated regime, where k does no more label the modes.

We turn now to the consideration of anharmonic elasticity. In the general theory of elasticity[17,20] the strain tensor contains nonlinear terms. Inserting the full nonlinear strain tensor into the elastic Lagrangian yields an additional anharmonic interaction

$$\mathcal{L}_{\mathrm{an}} = \lambda_0\left(\sum_i u_{ii}\right)\sum_{ij} v_{ij}v_{ij} + \mu(\mathbf{x})\sum_{ij\ell} u_{ij}v_{i\ell}v_{\ell j}, \tag{9}$$

where $v_{ij}=\frac{1}{2}(\partial_j u_i-\partial_i u_j)$ is the rotation tensor.

Further anharmonic terms would be due to anharmonic potential contributions yielding mode-Grüneisen-type couplings (cf., e.g. Refs. 17 and 21). In a system without spatial fluctuations of elastic constants translational invariance leads to a phase-space suppression of the anharmonic damping channel via Eq. (9) due to momentum conservation and a linear sound dispersion.[3,22] In the older treatments of anharmonic sound damping,[3,4] therefore, the mode-Grüneisen terms were taken as dominant anharmonic coupling which couple one transverse with two longitudinal phonons. As momentum conservation does not apply in the present situation, a finite phonon lifetime $\Gamma\{\mu(\mathbf{x})\}$ can already be calculated for a given configuration $\{\mu(\mathbf{x})\}$ from the interaction (9). The configurationally averaged lifetime $\langle\Gamma\rangle_{\{\mu(\mathbf{x})\}}$ would be nonzero in general. However it is more useful to work out the effect of the anharmonic interaction in terms of the effective fields χ_i,Σ_i, as they are important entities describing harmonic vibrations in glasses.

Using the standard replica field theory of disordered solids[15,16] we are able to show that the interaction (9) induces a third-order term in the effective theory for the fields representing the dynamic susceptibilities. A replica diagonal,

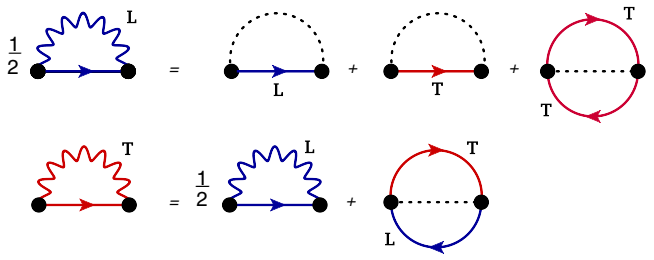

FIG. 1. (Color online) Diagrammatic representation of the anharmonic mean-field equations: blue=L, red=T; full lines are susceptibilities, dashed lines represent the correlation function; winding lines are the full self energies.

translation, and rotational invariant saddle point has been chosen, which yields the following self-consistent equations for the full self energies:

$$\Sigma_L(\omega_n) = \Sigma_{L,\mathrm{dis}}(z = i\omega_n) + 2\Sigma_{T,T,\mathrm{an}}(\omega_n), \tag{10}$$

$$\Sigma_T(\omega_n) = \frac{\Sigma_L(\omega_n)}{2} + \Sigma_{L,T,\mathrm{an}}(\omega_n), \tag{11}$$

$$\Sigma_{ij,\mathrm{an}}(\omega_n) = \frac{k_B T}{6V^2\rho^3}\sum_{\mathbf{k},\mathbf{q},m} \chi_i(\mathbf{k}, i\omega_{n-m}) K_\mu(\mathbf{k}-\mathbf{q})\chi_j(\mathbf{q}, i\omega_m). \tag{12}$$

Such equations have been shown[21] to be mathematically equivalent to mode-coupling equations.[23,24]

Indeed, these saddle-point equations resemble the structure of a calculation of the self-energy to first order in the anharmonic and disorder-induced interaction. The corresponding diagrams are shown in Fig. 1. The saddle-point equations contain self-energy dressed propagators instead of free ones.

III. MODE-DECAY APPROXIMATION AND RESULTS

In the following, we exploit this analogy for justifying the most obvious approximation to Eqs. (10)–(12), the *mode-decay approximation*. Our interest is the anharmonic effect on the dynamical structure factor and hence we are investigating the anharmonic effect on the longitudinal self-energy. We treat the anharmonic interaction as small perturbation and split the self-energy into harmonic and anharmonic parts $\Sigma_L = \Sigma_{\mathrm{dis}} + \Sigma_{\mathrm{an}}$. The propagators χ_i are approximated by the solution of the linearized equations, which are just the harmonic SCBA Eqs. (5) and (6).

Σ_{an} is then represented by the last self-energy diagram of the longitudinal channel in Fig. 1, in which the full transverse susceptibilities are replaced by the disorder-dressed ones.

We now represent $\mathbf{k}$ sums by

$$\sum_{\mathbf{k}} = \frac{V}{(2\pi)^3}\int_{|\mathbf{k}|<k_D} \mathrm{d}^3\mathbf{k},$$

and Matsubara sums by[25]

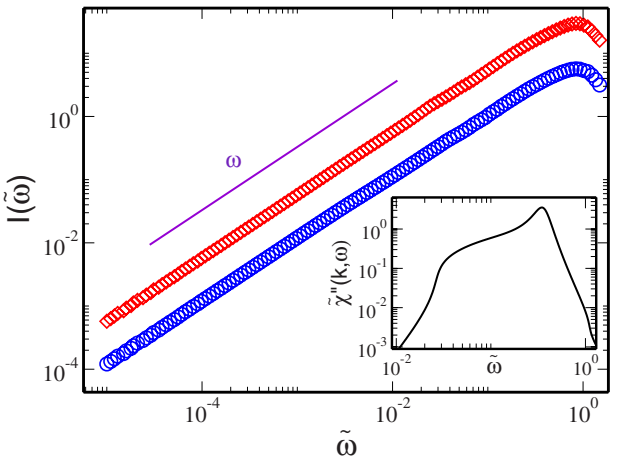

FIG. 2. (Color online) Frequency dependence of the anharmonic self-energy correction, for $\Delta^2 = 0.99\Delta_c^2$, $\xi = 2/k_D$ and $\Delta^2 = 0.9999\Delta_c^2$, $\xi = 1.5/k_D$. Inset: the function $\chi''(k=0.5k_D, \omega)$.

$$\sum_{\omega_m} A(i\omega_m) = \frac{\beta}{2\pi}\int_{-\infty}^{\infty} \mathrm{d}\omega \coth\frac{\beta\omega}{2}\mathrm{Im}\{A(z=\omega+i\epsilon)\}.$$

The anharmonic contribution to the Brillouin linewidth is then given by:

$$\Gamma_{\mathrm{an}}(k) = \frac{\omega_k}{v_L^2}\Sigma''_{\mathrm{an}}(\omega=\omega_k) \quad \omega_k = v_L k,$$

$$\Sigma''_{\mathrm{an}}(\omega) = \frac{\hbar}{6(2\pi)^7\rho^3}\int_{k_D} \mathrm{d}^3q\,\mathrm{d}^3k \int_0^\infty \mathrm{d}\bar{\omega}\coth\left(\frac{\beta\bar{\omega}}{2}\right) \times \chi''_T(\mathbf{k},\bar{\omega})K(\mathbf{q}-\mathbf{k})[\chi''_T(\mathbf{q},\bar{\omega}+\omega) - \chi''_T(\mathbf{q},\bar{\omega}-\omega)]. \tag{13}$$

The behavior of the susceptibility can be estimated from the inset of Fig. 2, it resembles the shape of the momentum independent self-energy, only the position of Brillouin peak is momentum dependent. We replace the correlation function $K_\mu(\mathbf{k})$ by its maximal value $8\pi\xi^3\Delta^2$. This approximation is robust, as it reflects the anharmonic expression we would have obtained in an uncorrelated framework. In addition we apply the classical (high temperature) limit in which we can replace the hyperbolic cotangent by its inverse argument. Then the self-energy is given by

$$\tilde{\Sigma}''_{\mathrm{an}}(\tilde{\omega}) = g(\Delta^2,\xi,k_BT)I(\Delta^2,\xi,\tilde{\omega}), \tag{14}$$

$$g(\Delta^2,\xi,k_BT) = \frac{\Delta^2 k_B T}{3\xi^3\pi^4\rho^3 v_T^4}, \tag{15}$$

$$I(\Delta^2,\xi,\tilde{\omega}) = \int_0^\infty \int_0^{\xi k_D} \mathrm{d}\tilde{k}\tilde{k}^2 \int_0^{\xi k_D} \mathrm{d}\tilde{q}\tilde{q}^2 \frac{\mathrm{d}\tilde{\bar{\omega}}}{\tilde{\bar{\omega}}} \times \tilde{\chi}''_T(\tilde{\mathbf{q}},\tilde{\bar{\omega}})[\tilde{\chi}''_T(\tilde{\mathbf{k}},\tilde{\bar{\omega}}+\tilde{\omega}) - \tilde{\chi}''_T(\tilde{\mathbf{k}},\tilde{\bar{\omega}}-\tilde{\omega})], \tag{16}$$

with g the interaction parameter in units of a squared sound

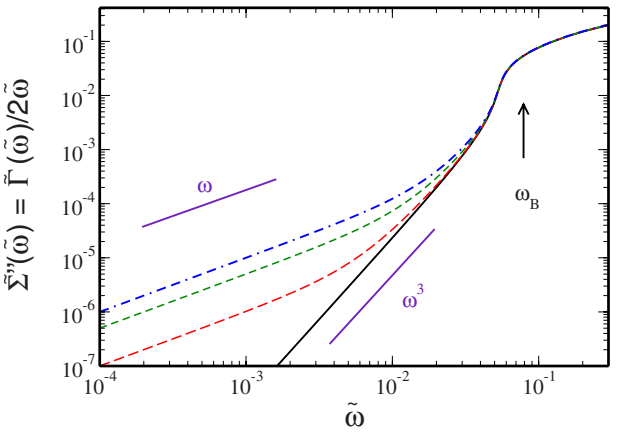

FIG. 3. (Color online) Brillouin linewidth, divided by frequency $\tilde{\Sigma}'' \cdot \alpha$ including disorder and anharmonicity for $\xi = 2/k_D$, $\Delta^2/\Delta_c^2 = 0.99$, $\alpha = (v_L/v_T)^2 = 2.52$ for different temperatures $T/T_0 = 0$ (full, black), 0.001 (long dashes, red), 0.005 (short dashes, green), 0.01 (dash-dots, blue). T_0 is defined by $\tilde{\Sigma}''_{\mathrm{an}}(\tilde{\omega}) = (T/T_0)\tilde{\omega}$ in the linear region.

velocity and a dimensionless integral I.[26] For small external frequencies, the integral kernel scales such as

$$\chi''_T(\mathbf{k}, \bar{\omega} + \omega) - \chi''_T(\mathbf{k}, \bar{\omega} - \omega) = \omega \partial_{\bar{\omega}} \chi''_T(\mathbf{k}, \bar{\omega}), \tag{17}$$

the Brillouin linewidth satisfies an Akhiezer-like sound attenuation law

$$\Sigma''_{\mathrm{an}}(\omega) \propto \omega T \Rightarrow \Gamma_{\mathrm{an}}(k) \propto \omega_k^2 T. \tag{18}$$

The validity of this ω^2 behavior has been tested by evaluating the full integral (16) instead of using Eq. (17). The result is shown in Fig. 3.

The self-energy behaves linearly in the whole frequency range, and not just at the lower band-edge $\omega \to 0$, as one would naively expect. This happens because the main contribution to the integral I, for frequencies below the BP, comes from the band of irregular delocalized high-frequency modes; these states contribute a wide $\chi'' \propto \omega$ regime, situated above the BP, to the disordered spectral density (see the inset of Fig. 2), for which the approximation (16) becomes exact.

Further, we estimate the amount of the attenuation induced by anharmonicity as compared with the disorder-induced one. The ratio $R(\omega) = \Gamma_{\mathrm{an}}(\omega)/\Gamma_{\mathrm{dis}}(\omega) \propto I(\tilde{\omega})/\tilde{\omega}$, which becomes frequency-independent beyond the BP, determines the crossover frequency. This ratio is fixed by the parameters Δ^2 and ξ, which set up the BP position, and the temperature. For example SiO_2 requires $\Delta^2 = 0.99\Delta_c^2 = 0.401\rho^2 v_T^4$ and $\xi = 2/k_D$. The ratio of the squared sound velocities is $(v_L/v_T)^2 = 2.52$ and the Debye cutoff $1.6 \times 10^{10}/m$.[12] At room temperature $k_B T k_D^3/\rho v_T^2 = 0.6$ and the ratio $I(\tilde{\omega})/\tilde{\omega} \approx 50$ is evaluated numerically for these parameters. We obtain $\Sigma''_{\mathrm{an}}(\tilde{\omega}) \approx 0.005\tilde{\omega}$, i.e., $T_0 = 7.3 \cdot 10^5$ K. Figure 3 shows the full Brillouin linewidth due to disorder and anharmonicity. The anharmonic corrections already lead to deviations from the disordered contribution slightly below the shoulder of the BP, the highest frequency, where the Akhiezer-like behavior is present in SiO_2 is therefore predicted to be one order of magnitude below the BP, i.e., in the 100 GHz regime. In general, for observing the Rayleigh law one needs to go to low enough temperature, which shifts the crossover toward lower frequencies.

IV. DISCUSSION AND CONCLUSIONS

Let us now discuss the possible role of potential-induced anharmonicity. There is no problem to introduce interactions involving longitudinal and transverse mode-Grüneisen parameters $g_{L,T}$ into the theory. In the longitudinal case one just has to replace Δ^2 by $(1+g_L^2)\Delta^2$ in the saddle point Eqs. (10)–(12). This will shift the crossover from Akhiezer-like to Rayleigh scattering upwards and reduce the "Rayleigh window" between the Akhiezer-Rayleigh crossover and the BP. Within our continuum description it is not possible to judge the importance of the mode-Grüneisen parameters $g_{L,T}$, because they are effective constants which have to be calculated from a certain microscopic theory, taking into account the material-specific details of the interaction. It has been pointed out by Fabian *et al.*,[8] that the Grüneisen parameters which they have extracted from their simulations are unusually strong, compared to the bare crystalline couplings. However, a suitable choice of the parameters for the Weber-Stillinger potential[27] used in their simulations[28] should account for the nonlinear part of the strain tensor as well. Hence, their strong Grüneisen parameters γ_i, should not be confused with our nonlinear couplings $g_{L,T}$, which may as a first guess be identified with their rather weak crystalline counterparts,[1,2] as long as one deals with weak disorder. However it cannot be excluded, that strong disorder or impurities drive these constants toward a strong-coupling regime within a renormalization-group approach. Therefore, for a particular material it has to be determined by experiment, whether our "minimal" description is sufficient, or one has to deal with mode-Grüneisen parameters. In any case our estimate of the Akhiezer-Rayleigh crossover serves as a lower bound.

The measurements on vitreous SiO_2, performed by Masciovecchio *et al.*,[6] exhibits the Akhiezer-Rayleigh crossover around 100 GHz, which fits our estimates quite well. In contradiction Devos *et al.*[7] claim the crossover to take place near 400 GHz. However they performed their measurements on amorphous thin films of SiO_2, prepared by chemical vapor deposition. Such materials—in comparison to glasses quenched from the liquid state—are known to have a wealth of additional defects such as voids and dangling bonds. It is at the heart of impurity physics that defects give rise to additional effective interactions, i.e., anharmonicities. This can lead to enhanced mode-Grüneisen parameters $g_{L,T}$, which then have to be taken into account.

In several experiments dealing with vibrational spectra near and below the glass transition[29,30] one observes an increase of the DOS in the BP regime. Within the mode-decay approximation one can account for the trends but not numerically reproduce such spectra.[31] We believe that one has to solve the full mode-coupling Eqs. (10)–(12) in order to be able to do so.

In conclusion, we developed a consistent perturbative treatment of the anharmonic contribution to the Brillouin

linewidth in disordered solids. Our treatment solely in terms of elasticity parameters, which enter into the mean values and correlation functions, suggests a correlation between the boson peak position and the Akhiezer-Rayleigh crossover at temperatures scaled with the Debye temperature. Further developments in experimental techniques are required to explore this extremely interesting frequency window in the upper GHz regime.

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