Fluctuating Elasticity Fails to Capture Anomalous Sound Scattering in Amorphous Solids

Christiane Caroli[1] and Anaël Lemaître[2]
[1]Sorbonne Universités, UPMC Université Paris 06, CNRS-UMR 7588, Institut des NanoSciences de Paris, 4 place Jussieu, 75005 Paris, France
[2]Laboratoire Navier, UMR 8205, École des Ponts, IFSTTAR, CNRS, UPE, Champs-sur-Marne, France

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The fluctuating elasticity (FE) model, introduced phenomenologically and developed by Schirmacher [J. Non-Cryst. Solids **357**, 518 (2011)], is today the only theoretical framework available to analyze low-temperature elastic acoustic scattering in glasses. Its existing formulations, which neglect the tensorial nature of elasticity and exclude long-range disorder correlations, predict that the acoustic damping coefficients obey the standard Rayleigh scaling law: $\Gamma \sim k^{d+1}$, with k the acoustic wave vector, in dimension d. However, recent numerical data, supported by the analysis of existing experimental results, show that Γ does not obey this scaling law but $\Gamma \sim -k^{d+1} \ln k$. Here we analyze in detail how a fully tensorial FE model can be constructed as a long wavelength approximation of the elastic response of the discrete, atomistic, problem. We show that, although it incorporates all long-range correlations, it fails to capture the observed damping in two respects: (i) it misses the anomalous scaling, and predicts the standard Rayleigh law; (ii) it grossly underestimates the amplitude of scattering by about 2 orders of magnitude. This brings clear evidence that the small scale nonaffine displacement fields, although not simply reducible to local defects, play a crucial role in acoustic wave scattering and hence cannot be ignored.

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Starting with the work by Zeller and Pohl [1] it has become increasingly clear that the low-temperature thermal and acoustic properties of glasses present universal features that differ markedly from those of the corresponding crystals. Below ~1 K, the specific heat $C(T)$ grows linearly with temperature T, a feature that has been interpreted as resulting from the presence of tunneling two-level systems (TLSs). Around 10 K, which corresponds to THz frequencies, $C(T)/T^3$ presents a hump, which signals an excess of the density of vibrational states with respect to the Debye prediction—the so-called Boson peak, which was proposed to result from the existence of localized soft modes akin to the TLS [2–4]. In the 1 to 10 K range, Zeller and Pohl showed that the phonon mean-free path decays roughly as T^{-4} with temperature, which is the signature of Rayleigh-like scattering. This strongly suggests that the glass anomalies in this range are determined by elastic acoustic phonon scattering, i.e., by the linear vibrational response. This view has motivated a line of theoretical work, which represents glasses as elastic continua with short-range correlated fluctuating elastic moduli [5,6], and accounts both for Rayleigh scattering and the presence of a hump in the reduced density of states.

It is only fairly recently, however, that thanks to the work of Monaco and co-workers it has become possible to directly experimentally probe elastic phonon scattering [7–10]. At the same time, computational advances have also permitted us to access elastic phonon scattering in the relevant frequency range [11–14]. In particular, a recent numerical study [14] in dimension $d = 2$ has shown, quite surprisingly, that the acoustic damping coefficient Γ does not obey the usual Rayleigh law, but that it should scale as $\Gamma \sim -k^{d+1} \ln k$, with k the wave vector, in the long wavelength limit. The analysis [14] of all currently available experimental and numerical data [7–13] supports very convincingly the validity of this prediction.

Since the Rayleigh law is the signature of scattering by local heterogeneities with finite-range correlations, its prediction by existing formulations of fluctuating elasticity (FE) [5,6] results from their assumption that the fluctuations of elastic constants decorrelate exponentially in space. Therefore, the observation of a different scaling is, in all likelihood, assignable to the presence of long-range correlations in the microstructural elastic features that cause scattering. And indeed the local elasticity tensor, as defined in Ref. [14], was found to present correlation tails decreasing with distance r as $1/r^d$. Since, moreover, John and Stephen [15] have shown that, in a scalar wave problem with $1/r^d$-correlated mass disorder, damping obeys the $\Gamma \sim -k^{d+1} \ln k$ scaling, it is natural to ask, and crucial to assess, whether or not a FE theory incorporating long-range correlations of elastic moduli captures the observed damping law.

In this perspective, it is important to note that these long-range correlations were found in Ref. [14] to disappear when the full elasticity tensor is reduced to its Lamé constants, as done in previous FE models—which thereby neglect the breaking of local symmetry in amorphous media [5,6]. The question is therefore to construct a, necessarily fully tensorial, formulation of FE that takes into account all self- and cross-correlations between elastic coefficients. For this purpose, one must proceed from the atomic scale equation of motion which, in the linear response approximation, is the discrete wave equation, and analyze how a Navier-like continuum elasticity formulation can be inferred from it as a large wavelength approximation.

Here, building upon the suggestion of Ref. [14], we formulate a long-wavelength approximation for the microscopic vibrational dynamics, which we refer to as FE-m, that enables us to compute all the coefficients of the Navier-like wave equation associated with a given atomic configuration. This permits us to directly compute the acoustic linear response, for a given system size, as a configurational average, and thus obtain sound damping coefficients, which we compare with the values obtained by simulating directly the discrete wave equation.

We find that the FE-m approximation grossly fails to capture the results of the full simulation. Namely, (i) it barely corrects the Born estimate of sound velocities; meanwhile, (ii) it underestimates by more than a decade the order of magnitude of the damping coefficients, and (iii) predicts them to obey the standard Rayleigh scaling. This latter result is very striking since the model does incorporate elastic coefficients that are correlated at long range. This will lead us to conclude that FE-m is insufficient to describe sound damping in amorphous solids, which, we think, stems from the fact that it misrepresents small scale atomic dynamics and thus grossly underestimates its contribution to elastic scattering.

The question of defining a coarse-grained elasticity for amorphous solids is delicate, due primarily to the ambiguities in the definition of strain at the mesoscopic scale [16,17]. Many authors have focused on defining local elastic moduli from the linear response of small regions isolated from the embedding medium [12,18,19]. These studies have shed light on the fluctuations of such local elastic properties. However, in this approach, it remains unclear how these small regions could be recoupled into an elastic continuum compatible with the microscopic dynamics, which is indispensable to tackle the problem of sound propagation. Here, we follow a different route, namely, we start from the discrete equations that describe the atomic dynamics linearized around an inherent state, i.e., a mechanically balanced amorphous configuration. For a system of volume V, composed of atoms of mass m, labeled as $i = 1, \ldots, N$, interacting via the pair potential U_{ij}, they read

$$\frac{\partial^2 u_i^\alpha}{\partial t^2} = \mathscr{D}_{ij}^{\alpha\kappa} u_j^\kappa. \tag{1}$$

Here, summation on repeated indices is implied; Greek subscripts denote Cartesian coordinates; $\underline{u}_i = \{u_i^\alpha\}$ is the displacement of atom i from its inherent state, mechanical equilibrium, position $\underline{r}_i$; the dynamic matrix $\{\mathscr{D}_{ij}^{\alpha\kappa}\}$ is defined by $\mathscr{D}_{ii}^{\alpha\kappa} = -(1/m)\sum_{i\neq j} M_{ij}^{\alpha\kappa}$ and $\mathscr{D}_{ij}^{\alpha\kappa} = (1/m)M_{ij}^{\alpha\kappa}$ if $i \neq j$, with

$$M_{ij}^{\alpha\kappa} = \left(U_{ij}''(r_{ij}) - \frac{U_{ij}{}'(r_{ij})}{r_{ij}}\right) n_{ij}^\alpha n_{ij}^\kappa + \frac{U_{ij}{}'(r_{ij})}{r_{ij}} \delta^{\alpha\kappa} \tag{2}$$

where $\underline{r}_{ij} = \underline{r}_j - \underline{r}_i$, $\underline{n}_{ij} = \underline{r}_{ij}/r_{ij}$, and $\delta^{\alpha\kappa}$ denotes the Kronecker delta.

We want to know whether, in the large wavelength limit, this equation can be reduced to the FE form, in the sense of classical continuum elasticity, that is without higher-order or higher-gradient contributions. It should be noted that the discrete problem involves displacements from the mechanical equilibrium, reference, state, and therefore must be compared with the Lagrangian formulation of FE, which reads [20]

$$\frac{\partial^2 u^\alpha(\mathring{\underline{r}})}{\partial t^2} = \frac{1}{\rho}\frac{\partial}{\partial \mathring{r}^\beta}\left(S^{\alpha\beta\kappa\chi}(\mathring{\underline{r}})\frac{\partial u^\kappa(\mathring{\underline{r}})}{\partial \mathring{r}^\chi}\right) \equiv \mathbf{D}^{\alpha\kappa}[u^\kappa](\mathring{\underline{r}}), \tag{3}$$

with $\rho = Nm/V$ the average mass density, and $\mathring{\underline{r}}$ the coordinates in the reference, equilibrium, state; $\underline{u}(\mathring{\underline{r}})$ is the continuous Lagrangian displacement field, and the elasticity coefficients are

$$S^{\alpha\beta\kappa\chi}(\mathring{\underline{r}}) = C^{\alpha\beta\kappa\chi}(\mathring{\underline{r}}) + \delta^{\alpha\kappa}\sigma^{\beta\chi}(\mathring{\underline{r}}), \tag{4}$$

where $C^{\alpha\beta\kappa\chi}$ and $\sigma^{\beta\chi}$ are, respectively, the elastic constants and the Cauchy stress in the reference configuration. Since plane waves form a complete, orthogonal basis for the continuum, the problem is equivalently written in Fourier space as $\partial^2\hat{u}^\alpha(\underline{k})/\partial t^2 = \int d\underline{k}' \hat{D}^{\alpha\kappa}(\underline{k}, \underline{k}')\hat{u}^\kappa(\underline{k}')$ where

$$\begin{aligned}\hat{D}^{\alpha\kappa}(\underline{k}, \underline{k}') &\equiv \frac{1}{V}\int d\mathring{\underline{r}} d\mathring{\underline{r}}' e^{-i\underline{k}\cdot\mathring{\underline{r}}} \mathbf{D}^{\alpha\kappa}[e^{i\underline{k}'\cdot\mathring{\underline{r}}'}] \\ &= -\frac{1}{\rho V} k^\beta k'^\chi \hat{S}^{\alpha\beta\kappa\chi}(\underline{k} - \underline{k}')\end{aligned} \tag{5}$$

defines the coupling between any pair of plane waves. Let us emphasize that any linear problem is of the FE type if and only if its kernel is of the above form, featuring in particular the $k^\beta k'^\chi$ factor.

Let us now return to the discrete problem and consider discrete plane waves, $\{(1/\sqrt{N})e^{i\underline{k}\cdot\underline{r}_i}\}$. Although they are not orthogonal for the discrete scalar product $(1/N)\sum_i e^{i(\underline{k}-\underline{k}')\cdot\underline{r}_i}$, a number $N \times d$ of them may generically be used as a basis to decompose displacement fields. So, writing $u_i^\alpha = (1/\sqrt{N})\sum_{\underline{k}} \hat{u}_{\underline{k}}^\alpha e^{i\underline{k}\cdot\underline{r}_i}$, where the sum involves $N \times d$ terms, defines unambiguously a set of coefficients $\hat{u}_{\underline{k}}^\alpha$ using which the discrete vibrational response writes

$$\frac{\partial^2 \hat{u}^\alpha_{\underline{k}}}{\partial t^2} = \mathscr{D}^{\alpha\kappa}_{\underline{k},\underline{k}'} \hat{u}^\kappa_{\underline{k}'}. \tag{6}$$

In the large wave limit of our interest, we neglect the lack of orthogonality between discrete plane waves, and thus approximate the coupling coefficients by

$$\begin{aligned}\mathscr{D}^{\alpha\kappa}_{\underline{k},\underline{k}'} &\simeq \frac{1}{N}\sum_i\sum_j e^{-i\underline{k}\cdot\underline{r}_i}\mathscr{D}^{\alpha\kappa}_{ij}e^{i\underline{k}'\cdot\underline{r}_j} \\ &= -\frac{4}{Nm}\sum_i\sum_{j>i} M^{\alpha\kappa}_{ij}\sin\left(\frac{\underline{k}'\cdot\underline{r}_{ij}}{2}\right) \\ &\quad\times\sin\left(\frac{\underline{k}\cdot\underline{r}_{ij}}{2}\right)e^{i(\underline{k}'-\underline{k})\cdot\bar{\underline{r}}_{ij}}. \end{aligned}\tag{7}$$

where $\bar{\underline{r}}_{ij} = (\underline{r}_i + \underline{r}_j)/2$. In this limit, furthermore, since $\underline{k}\cdot\underline{r}_{ij}$, $\underline{k}'\cdot\underline{r}_{ij} \ll 1$, the coefficients $\mathscr{D}^{\alpha\kappa}_{\underline{k},\underline{k}'}$ asymptotically reduce to the functional form Eq. (5) characteristic of FE, with the coefficients

$$\widehat{\mathscr{S}^{\alpha\beta\kappa\chi}}(\underline{q}) = \sum_i\sum_{j>i}\mathscr{S}^{\alpha\beta\kappa\chi}_{ij}e^{i\underline{q}\cdot\bar{\underline{r}}_{ij}} \tag{8}$$

where the pair contributions $\mathscr{S}^{\alpha\beta\kappa\chi}_{ij} = \mathscr{C}^{\alpha\beta\kappa\chi}_{ij} + \delta^{\alpha\kappa}\sigma^{\beta\chi}_{ij}$ with

$$\begin{aligned}\mathscr{C}^{\alpha\beta\kappa\chi}_{ij} &= [r_{ij}U''_{ij}(r_{ij}) - U_{ij}{}'(r_{ij})]r_{ij}n^\alpha_{ij}n^\beta_{ij}n^\kappa_{ij}n^\chi_{ij}, \\ \sigma^{\beta\chi}_{ij} &= U'_{ij}(r_{ij})r_{ij}n^\beta_{ij}n^\chi_{ij}.\end{aligned}\tag{9}$$

Let us emphasize that any continuum problem is by nature defined for arbitrarily large wave vectors. Furthermore, any FE model is necessarily specified by an operator $\hat{D}^{\alpha\kappa}(\underline{k},\underline{k}')$ of the form given by Eq. (5). To construct a FE approximation we must replace the discrete operator $\mathscr{D}^{\alpha\kappa}_{\underline{k},\underline{k}'}$, which is only of the form [Eq. (5)] asymptotically in the low $\underline{k}$, $\underline{k}'$ limit, by a continuum one which is necessarily of the form [Eq. (5)] at all $\underline{k}$ and $\underline{k}'$. Therefore, since Eq. (5) is not verified by the discrete operator at all wave vectors, FE is perforce an ansatz. That is, there is an unavoidable arbitrariness in the specification of the elastic coefficients $\hat{S}^{\alpha\beta\kappa\chi}$ for the continuum problem. Taking these remarks into account, we construct the FE-m approximation by requiring that its asymptotic form matches that of the discrete operator in the large wavelength limit, and assume for simplicity that

$$\hat{S}^{\alpha\beta\kappa\chi}(\underline{q}) = \begin{cases}\widehat{\mathscr{S}^{\alpha\beta\kappa\chi}}(\underline{q}) & \text{if} \quad q < q_{\max} \\ 0 & \text{if} \quad q > q_{\max}\end{cases} \tag{10}$$

where we have introduced a cutoff $q_{\max}$, which we will show not to affect attenuation in the small wave vector, acoustic, domain.

Now we test our continuum elasticity model using the 2D binary soft sphere (power law repulsive) system, the acoustic properties of which have been extensively studied in Ref. [14]. As in that work, the configuration ensemble we use consists of inherent states produced by energy minimization from liquid configurations in equilibrium at temperature $T = 0.32$, in $L \times L$ square cells with periodic boundary conditions. Lennard-Jones units are used throughout.

To measure attenuation in our FE approximation, we follow the protocol of Ref. [14]. Namely, for each inherent state, we compute the elasticity coefficients $\widehat{\mathscr{S}^{\alpha\beta\kappa\chi}}(\underline{q})$, which defines the associated continuum problem via Eqs. (8) and (10) (see Supplemental Material [21]). We then solve numerically the corresponding wave equation, Eq. (3), using a Verlet-like integration, starting from an initial condition of zero displacement, and either a transverse (T) or a longitudinal (L) plane wave velocity field of wave vector $\underline{k} = (0, k)$ with $k = 2\pi n/L$, and measure the correlation $C(t)$ of velocity between initial and running time t. We only consider $n > 6$ since, for lower n values, attenuation is strongly affected by finite size effects. We finally average the time evolution of the correlation function $C(t)$ for each system size, each $\underline{k}$, and each polarization, and different values of the cutoff $q_{\max}$. The ensemble-averaged $C(t)$'s fit very nicely the expression $\cos(\Omega t)\exp(-\Gamma t/2)$, which provides the sound speeds $c_{L,T}$ as well as the attenuation coefficients $\Gamma_{L,T}$.

We report in Fig. 1, as a function of k, the values of c_L (a), c_T (b), Γ_L (c), and Γ_T (d) measured in both the full discrete problem (solid black line) and in the FE-m approximation (symbols) for system size $L = 673$ and different values of the cutoff $q_{\max}$. We first observe that the FE-m approximation predicts sound speed values that are significantly larger than the full problem ones, and quite closely approach the elastic Born approximation of their $k \to 0$ limit (red dashed lines). This immediately suggests that the FE-m approximation significantly underestimates nonaffine effects.

Moreover, for each polarization, the attenuation coefficients produced by the FE-m approximation at different $q_{\max}$ values collapse on a single curve for $k \lesssim q_{\max}$; at higher k's, as can be expected, attenuation increases with $q_{\max}$. This demonstrates the irrelevance of the precise shape of the ultraviolet cutoff on $\widehat{S^{\alpha\beta\kappa\chi}}(\underline{q})$, and therefore the robustness of the FE-m predictions in the relevant, acoustic, domain.

Beyond this, two striking features emerge. On the one hand, FE-m underestimates attenuation by about 2 orders of magnitude. On the other hand, the low-k FE-m master curves appear consistent with the standard Rayleigh scaling $\Gamma \propto k^{d+1}$ in dimension $d = 2$, in clear contrast with the measured attenuation in the full discrete problem.

In Fig. 2, we present, for $q_{\max} = 0.8$ and various system sizes, log-lin plots of Γ_T/k^3 or Γ_L/k^3 vs k, which permit us to discriminate between Rayleigh scaling and the

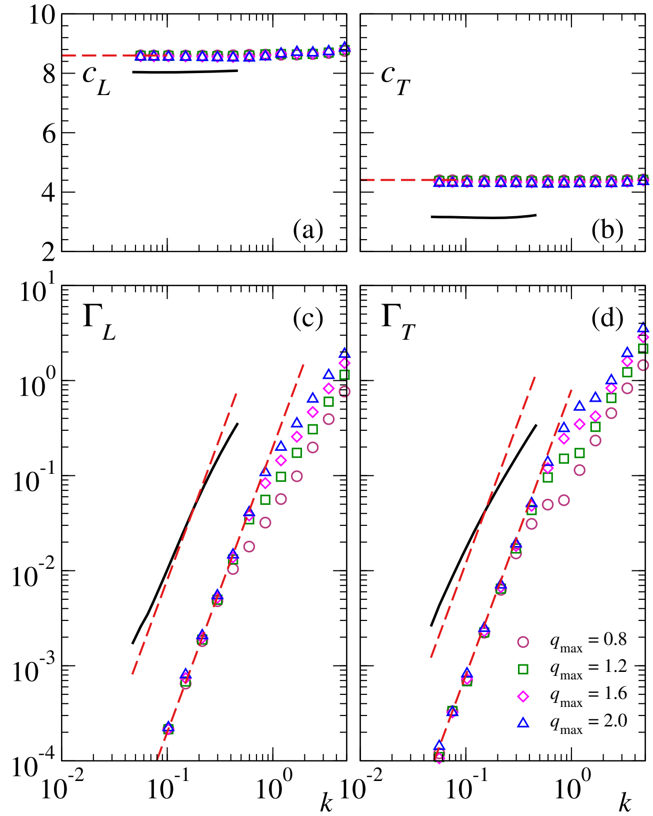

FIG. 1. Top: Longitudinal (a) and transverse (b) sound velocities $c_{L,T}$ vs k, with the same symbols. Red dashed lines correspond to their values in the elastic Born approximation. Bottom: Longitudinal (c) and transverse (d) attenuation coefficents $\Gamma_{L,T}$ vs wave vector k, for the full problem (solid black line) and for the fluctuating elasticity approximation model (symbols) for different values of the cutoff $q_{\max}$. System size $L \simeq 673$. Red dashed guidelines indicate the standard 2D Rayleigh behavior $\Gamma \sim k^3$.

anomalous $-k^3 \ln k$ behavior of the real discrete system [14]. The discrete data (full symbols) clearly show the anomalous scaling up to an upper cutoff that roughly corresponds to the Boson peak ($\omega_{\mathrm{BP}} \simeq 1.3$). In contrast, for increasing L's, the FE-m data (empty symbols, see also inset) collapse onto a master curve which plateaus at low k thus demonstrating that the continuum FE-m problem does obey Rayleigh scaling in the large system size limit. We have checked that, as already seen on Fig 1, while its upper cutoff increases with $q_{\max}$, the height of the Rayleigh plateau is $q_{\max}$ independent.

This result is quite surprising since, in dimension $d = 2$, the FE-m elasticity coefficients $\mathscr{C}^{\alpha\beta\kappa\chi}$ [see Eq. (9)] as well as the stress field $\sigma^{\alpha\beta}$ have been shown to present long-range, $1/r^2$-decaying, correlations [14,22,23]. Indeed, Rayleigh scaling is usually associated with scattering from short-range correlated heterogeneities; moreover, John and Stephen have shown that $1/r^d$-correlated mass disorder results in $-k^{d+1} \ln k$ attenuation [15].

We think the solution of this paradox lies in the tensorial nature of elasticity. Indeed, the autocorrelation tensors of $\mathscr{C}^{\alpha\beta\kappa\chi}$ and $\sigma^{\alpha\beta}$ comprise spatially isotropic and anisotropic (i.e., having zero angular average) terms, and it was shown [14,23] that only the anisotropic ones present long-range decay (see also Ref. [21]). Moreover, a calculation of the FE-m damping coefficients within the scattering Born approximation [21] shows that the contributions of the various long-range, anisotropic correlation components, which scale individually as $-k^3 \ln k$ at leading order, cancel out exactly for reasons of symmetry, so that the only remaining contributions to damping come from isotropic, short-ranged, contributions to the autocorrelation of elastic coefficients. The standard Rayleigh scaling thus appears to be a consequence of symmetry properties of the elastic tensor in the small k asymptotic limit. The above result suggests that any FE ansatz that matches asymptotically the microscopic elasticity tensor in the large wavelength limit will lead to the same Rayleigh prediction.

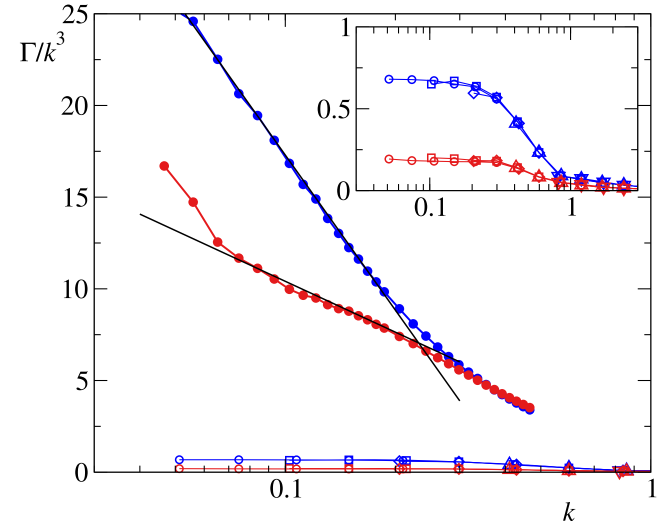

FIG. 2. Log-lin plot of Γ_L/k^3 (blue) and Γ_T/k^3 (red) vs wave vector k, for the $L = 1348$ full problem (data taken from Ref. [14], filled circles), and for the FE-m (open symbols) with system sizes: $L = 84$ (down triangles), $L = 168$ (up triangles), $L = 336$ (diamonds), $L = 673$ (squares), and $L = 1348$ (circles). Inset shows a zoom on FE-m data points.

In summary, we have shown here how a FE approximation can be constructed as a small k expansion of the full discrete problem, and conclude to its inability to capture acoustic properties: it hardly corrects the elastic Born prediction to sound speeds, fails to capture the scaling behavior of acoustic attenuation, and underestimates the magnitude of attenuation in the whole accessible k range, up to the upper end of the $-k^3 \ln k$ scaling regime.

The incorrect prediction of sound speeds points to a deficiency in the representation of nonaffine effects, since the latter are completely neglected in the Born approximation [24,25]. In this regard, it must be stressed that (i) the respective importance of the contributions of different spatial scales of the nonaffine response has remained unexplored until now, while (ii) it remains unclear whether sound speed values, on the one hand, and the magnitude and wavelength dependence of attenuation, on the other, are controlled by the same features of the small scale nonaffine response [26]. Our FE-m approximation does incorporate large scale nonaffinity, since it correctly captures the direct couplings between large wavelength plane waves; its identified failures therefore demonstrate the crucial role of the nonaffine contributions originating from small scale heterogeneities. This is illustrated by a movie of the particle velocity field (in response to a transverse $k \simeq 0.22$ wave, for $L = 168$) [21], which shows very clearly that an initial small-k wave excites, in the full discrete problem (left), small scale nonaffine motions of rapidly growing amplitude, while no such effect is discernible for the FE-m model (right).

An attempt at taking into account small scale nonaffinity, the phenomenological model of Maurer and Schirmacher [27,28], inspired from the soft potential model [3,28,29], consists in augmenting the Schirmacher FE model with the introduction of local oscillators. However, it remains insufficient, since, once more, it predicts the Rayleigh scaling. We believe that any scattering theory should comprise as its zeroth order the FE approximation we have constructed here, so as to correctly incorporate the direct couplings between long-wavelength plane waves, and the associated long-range correlations of elasticity coefficients. But it should, most probably, also take into account local torques, as micropolar formulations of elasticity do. Yet, it must derive from a controlled coarse-graining procedure of the microscopic system, which incorporates more realistically how plane waves couple with small scale heterogeneities.

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