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Some comments on fluctuating-elasticity and local oscillator models for anomalous vibrational excitations in glasses☆

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ABSTRACT

An overview is given on the present status of the theoretical description of vibrational spectra of glasses, as seen by inelastic neutron, X-ray and light (Raman) scattering. Using the language of Green's/response functions the merits and shortcomings of a local oscillator and a generalized elasticity-theory point of view are discussed. It is pointed out that in both cases the interaction of phonons with disorder-induced irregularities leads to Rayleigh scattering (mean free path $\ell \propto \omega^{-4}$) at low enough frequencies and temperatures. In disordered solids at ambient temperature the Rayleigh scattering is usually masqued by Akhiezer-like anharmonic scattering $\ell \propto \omega^{-2}$, but it can be made visible by lowering the temperature. Using a combination of fluctuating-elasticity theory with an incoherent spectrum of local oscillators a fair description of the vibrational spectrum of glassy SiO_2 can be achieved.

1. Introduction

Since the appearance of the seminal paper of Uli Buchenau et al. [1] on neutron scattering from glassy SiO_2 a wealth of publications on the rather anomalous vibrational properties of glasses has been published [2–9]. But, in fact, this discussion started already some 50 years ago [10] with the observation of a low-frequency Raman band that is not present in crystalline Raman spectra and which has been called "boson peak" [11,12]. According to a suggestion of Shuker and Gammon [13] these spectra were assumed to be proportional to the vibrational density of states (DOS), so the excess over the Debye DOS, observed in SiO_2 [1] and many other glasses [4–8] inherited also the name "boson peak" [14]. Among the vibrational anomalies observed in disordered solids as compared to crystals this feature is the most striking one. It also shows up as a characteristic peak in the temperature-dependent specific heat, plotted as $C(T)/T^3$. Near this peak the thermal conductivity $\kappa(t)$ shows a characteristic shoulder or "dip", which can be shown [15] to be intimately related to the boson peak. Below the "boson peak temperature" (mostly ~10 K) $C(T)$ varies almost linearly with T and $\kappa(T)$ almost quadratically, which can be explained by the two-level model [2].

The boson peak shows up in a frequency range where the broadening of the acoustic excitations becomes of the same order of magnitude of the Brillouin resonance frequency ("Ioffe-regel limit" [16,17]). This observation led different authors to hypothesize a relationship between the appearance of the boson peak and the existence of localized vibrations [18,19]. Acoustic waves that become Anderson-localized, it was argued, could produce the plateau in the thermal conductivity. Following this idea, investigations of (Anderson) localization properties of waves in disordered systems based on simulations [20], model calculations [21,22] and field-theoretical techniques [23] have shown that Anderson-localized states in disordered media do actually occur, but in a much higher frequency range (near the upper band edge) than the boson peak frequency.

So the question is: what is the very nature of the states near and above the boson peak frequency? As these states are neither really propagating nor localized, Fabian et al. [24] suggested to call them "diffusons": They behave like diffusing light in milky glass. In this regime, however, the Brillouin resonance frequency Ω_k as measured by inelastic X-ray scattering still exhibits a linear dispersion $\Omega_k = v_L k$ with the wave number k. In this frequency range the width Γ_k of the excitations appears to acquire a k^2 dependence [25]. As in this regime the "would-be" mean-free path $\ell = 2v_L/\Gamma_k$ is of the same order of magnitude as the wavelength of the sound-like vibrational excitations the wave vector loses its property of labeling the vibrational mode. In quantum theory (we are discussing classical vibrational excitations) one would say, k is no more a good quantum number. Also perturbation theory with respect to $(k\ell)^{-1}$ breaks down, and one has to find a non-perturbative description of the observed spectra. Such a description – in terms of elasticity theory with fluctuating elastic constants – is nowadays available [12,15,22,26–29] (fluctuating-elasticity concept, FE), and we shall give an overview in the next sections and compare it with the soft-potential/local oscillator (LO) model.

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Beforehand we briefly summarize the previous efforts to understand or explain the boson-peak anomaly. In fact, an enormous number of possible explanations have been published in the literature [4], which can roughly be grouped into three classes: *i*) Models with spatially fluctuating elastic constants. *ii*) Models associated with the glass transition and *iii*) defect models.

i) In models with quenched disorder of elastic constants [15,22,26–34] the boson peak marks the lower frequency bound of a band of irregular delocalized states with random mutual hybridization. These states are neither propagating nor localized [22]. The models have been solved with the help of numerical simulations as well as effective-medium theories.
ii) In theories of the glass transition [35–39] the boson peak arises as a benchmark of the frozen glassy state.
iii) Defects with a heavy mass can produce resonant quasi-local resonant states within the DOS [40–42] and be thus the reason for the boson peak and the reduction of the thermal conductivity. Similarly defects with very small elastic constants, near which anharmonic interactions are important (soft potentials), can produce quasi-local states, which, if hybridized with acoustic excitations may produce a boson peak [8,43] and a plateau in the thermal conductivity [44,45]. Inhomongeneities may also be the source of local vibrational excitations that contribute to the excess DOS [46]. Specifically in network glasses bond-angle distortions can also contribute to the boson-peak anomaly [1,7]. All these models essentially assume that the boson peak arises from the coupling of sound waves to local oscillators. In a recent study [26] the predictions of a LO model has been compared with those of a FE model [15,26–29]. This will also be done in the present contribution.

2. Rayleigh scattering, fluctuating elastic constants and local oscillators

Before we go into the details of the FE and LO theory we introduce some general concepts, which are helpful for discussing the matter.

We start with a simple wave model, in which waves are described by a scalar amplitude $u(\mathbf{r},t)$, which is supposed to obey a wave equation:

$$\frac{\partial^2}{\partial t^2}u(\mathbf{r},t) = K_0\nabla^2 u(\mathbf{r},t) \tag{1}$$

Here $K_0 = v_0^2$ is an elastic constant, divided by the mass density and v_0 is the sound velocity. In frequency space we have:

$$-\omega^2 u(\mathbf{r},\omega) = K_0\nabla^2 u(\mathbf{r},\omega) \tag{2}$$

The Green's function obeys:

$$-\omega^2 G_0(\mathbf{r},\mathbf{r}',\omega) - K_0\nabla^2 G_0(\mathbf{r},\mathbf{r}',\omega) = \delta(\mathbf{r}-\mathbf{r}') \tag{3}$$

Here ω must contain an infinitesimal imaginary part for mathematical reasons [40]. As is well known the Green's function is very helpful for describing the presence of inhomogeneities (in the physical *and* mathematical sense).

The first inhomogeneity one can study is a spatial variation of the elastic constant:

$$\mathrm{K}(\mathbf{r}) = \mathrm{K}_0 + \Delta K(\mathbf{r}), \tag{4}$$

which leads to an equation of motion:

$$-\omega^2 G(\mathbf{r},\mathbf{r}',\omega) - \nabla(K_0 + \Delta K(\mathbf{r}))\nabla G(\mathbf{r},\mathbf{r}',\omega) = \delta(\mathbf{r}-\mathbf{r}') \tag{5}$$

or in k space [47]:

$$\underbrace{\left(-\omega^2 + K_0k^2\right)}_{G_0^{-1}(\mathbf{k},\omega)} G(\mathbf{k},\mathbf{k}',\omega)\delta_{\mathbf{k},\mathbf{k}'} = \delta_{\mathbf{k},\mathbf{k}'} - \sum_{\mathbf{q}} \mathbf{k}\cdot\mathbf{q}\Delta K(\mathbf{k}-\mathbf{q})G(\mathbf{q},\mathbf{k}'\omega) \tag{6}$$

As the macroscopic (averaged) Green's function depends only on the difference of $\mathbf{r}$ and $\mathbf{r}'$ we perform the Fourier transform [47] with respect to this difference and write:

$$G(\mathbf{k},\omega) = \langle G(\mathbf{k},\mathbf{k}',\omega)\rangle\delta_{\mathbf{k},\mathbf{k}'} = \frac{1}{-\omega^2 + K_0k^2 - \Sigma(\mathbf{k},\omega)} \tag{7}$$

Here $\Sigma(\mathbf{k},\omega)$ is an unknown function, which describes in an average way the influence of the disorder.One can define a complex, frequency-dependent sound velocity, in analogy to optics:

$$v^2(\omega) = v_0^2 - \lim_{\mathrm{k}\to 0}\Sigma(\mathbf{k},\omega)/k^2 \equiv v_0^2 - \Sigma(\omega) \tag{8}$$

where we have defined a $\mathbf{q}$ independent low-wave vector self energy $\Sigma(\omega)$. The real part of the complex sound velocity is the "real" (disorder-modified) sound velocity, the imaginary part gives rise to a finite mean-free path $\ell(\omega)$:

$$v''(\omega) = \frac{1}{2\omega}\frac{|v(\omega)|^2}{\ell(\omega)} \tag{9}$$

from which follows:

$$\frac{1}{\ell(\omega)} = \frac{\omega}{v^3}\Sigma''(\omega) \tag{10}$$

We now solve Eq. (7) for $\Sigma(\mathbf{k},\omega)$ and expand the resulting expression to second order in ΔK to obtain:

$$\Sigma(\mathbf{k},\omega) = \sum_{\mathbf{q}} (\mathbf{k}\cdot\mathbf{q})^2 C(\mathbf{k}-\mathbf{q})G_0(\mathbf{k},\mathbf{q}) \tag{11}$$

Where:

$$C(\mathbf{q}) = \frac{1}{V}\mathrm{d}^3\mathbf{r}e^{i\mathbf{q}\mathbf{r}}\langle\Delta K(\mathbf{r}+\mathbf{r}_0)\Delta K(\mathbf{r}_0)\rangle \tag{12}$$

is the Fourier transform of the spatial correlation function of the fluctuating elastic constant. In deriving Eq. (11) we have used the fact that the average of ΔK is zero.

In the low-wave number limit we have for the correlation function:

$$C(\mathbf{q}\to 0) = \langle\Delta K^2\rangle\frac{\xi^3}{V} \tag{13}$$

and we obtain:

$$\sum(\mathbf{k},w) = k^2\frac{\xi^3}{3}\langle\Delta K^2\rangle\frac{1}{(2\pi)^3}\int\mathrm{d}^3\mathbf{q}\frac{q^2}{-w^2 + v_0^2q^2} \tag{14}$$

we now use the identity:

$$\mathrm{Im}\left\{\frac{1}{q^2 - \omega^2/v_0^2}\right\} = \pi\delta\left(q^2 - \omega^2/v_0^2\right) = v_0\frac{\pi}{2\omega}\delta(q-\omega/v_0) \tag{15}$$

to obtain:

$$\boxed{\frac{1}{\ell(\omega)} = \frac{\omega}{v^3}\Sigma''(\omega) = \frac{\xi^3}{3\pi}\frac{<\Delta K^2>}{v_0^8}\omega^4} \tag{16}$$

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which is the famous Rayleigh law, which gives rise to the blue color of the sky. In light scattering off the inhomogeneities of the admosphere ΔK represents the fluctuations of the dielectric constant and/or the density of air.

We now turn to the alternative model, namely to the possibility of coupling the waves with local oscillators, described by a configuration coordinate x_i, which could be, for example the libration angle of SiO_2 tetrahedra [1]. We assume that they couple to the strain in the immediate vicinity, so that they obey an equation of motion [26]:

$$\ddot{x}_i(t) = -w_0^2 x_i(t) - \frac{1}{M}\int \mathrm{d}^3\mathbf{r} V(\mathbf{r}-\mathbf{r}_i)\nabla u(\mathbf{r},t) \tag{17}$$

Here M is the effective mass, e.g. the moment of inertia of the Buchenau-tetrahedra. $V(\mathrm{r})$ is the coupling potential. (We still treat the phonon amplitudes as scalar entities). For the phonons, we have [26]:

$$\ddot{u}(\mathbf{r},t) - v_0^2\nabla^2 u(\mathbf{r},t) = \frac{1}{\rho_m}\sum_i \nabla V(\mathbf{r}-\mathbf{r}_i)x_i \tag{18}$$

where ρ_m is the mass density. The inhomogeneous Eq. (17) can be solved with the Green's function of the homogeneous local oscillator equation:

$$G_i^{(0)}(\omega) = \frac{1}{-\omega^2 + \omega_i} \tag{19}$$

Assuming a very short-ranged potential in comparison with the length scale of the phonon wave lengths:

$$V(\mathbf{r}-\mathbf{r}_i) = V_0\delta(\mathbf{r}-\mathbf{r}_i) \tag{20}$$

we obtain an effective equation for the phonons:

$$\ddot{u}(\mathbf{r},t) - v_0^2\nabla^2 u(\mathbf{r},t) - \nabla K_{\mathrm{eff}}(\mathbf{r},\omega)\nabla u(\mathbf{r},t) \tag{21}$$

with an "effective fluctuating elastic constant":

$$K_{\mathrm{eff}}(\mathbf{r},\omega) = \lambda\sum_i G_i^{(0)}(\omega)\delta(\mathbf{r}-\mathbf{r}_i) \tag{22}$$

with the coupling constant $\lambda = \frac{V_0^2}{M\rho_m}$. We now can perform the same steps, which led to the Rayleigh law (16), but we have to be careful, because the quantities $\Delta K(\mathbf{r})$ the variance of which appears in its prefactor have (by definition) zero average. In the present case the average of $K_{\mathrm{eff}}(\mathrm{r},\omega)$ is non-zero, and we obtain for the local oscillator-induced self energy of the phonons to second order in the small parameter V_0^2:

$$\Sigma(\mathbf{k},\omega) = k^2\langle K_{\mathrm{eff}}(\omega)\rangle + \langle \Delta K_{\mathrm{eff}}(\omega)^2\rangle \int \frac{\mathrm{d}^3\mathbf{q}}{(2\pi)^3}(\mathbf{k}\cdot\mathbf{q})^2 S(\mathbf{k}-\mathbf{q})\frac{1}{-\omega^2 + v_0^2 q^2} \tag{23}$$

where $S(\mathrm{k}) = \frac{1}{N}\sum_{ij} e^{i\mathbf{k}(\mathrm{r}_i-\mathrm{r}_j)}$ is the "structure factor" of the local oscillators. The first term is identical to that obtained in Ref. [26], the second one was missing in this treatment, because only the lowest-order term was discussed. In the regime $\omega \ll \omega_i$ (for all i), i.e. below the resonances, the second-order term obviously gives rise to a *Rayleigh law* like the non-frequency-dependent fluctuating elastic constants. I believe this result is quite important as it shows that the Rayleigh law is a very general one and applies also to the LO model. Important for the derivation is, however, that the underlying Lagrangian [26] is invariant under translative coordinate transformations (global translation invariance), which was achieved by assuming that the local oscillators couple not to the phonon displacements, but to the strain.

2.1. A comment on the local oscillator DOS

There is an extended literature on soft potentials and the corresponding local oscillators [8,43,48] in which the relaxational reconstruction of an initially arbitrary (and therefore assumedly smooth) LO DOS in the presence of interactions is discussed. These interactions are the anharmonic interaction and the indirect interaction via static elasticity. Whereas the latter is shown to shift the local squared frequencies ω_0^2 downwards, thus inducing an instability, the former stabilize the frequencies. The result is a uniform distribution of the renormalized ω^2, resulting in a linear $g(\omega) = 2\omega g(\omega^2)$. However, at very small frequencies, these authors argue, local forces f_i become distinct, which balance the anharmonic oscillator forces:

$$f_i = M\omega_i^2 x_0 + Ax_0^3 \tag{24}$$

where A is the strength of the anharmonicity and x_0 is the new equilibrium position of the local configuration coordinate. The local forces shift the original (already elasticity-renormalized) frequencies ω_i:

$$\omega_{\mathrm{new}} = \frac{1}{M}\frac{\mathrm{d}f_i}{\mathrm{d}x_0} = \omega_i^2 + \frac{3A}{M}x_0^2 \tag{25}$$

which results in a renormalized low-frequency local DOS g:

$$\begin{aligned} g_{\mathrm{new}}(\omega) &\propto \int \mathrm{d}\omega_i^2 \underbrace{g\left(\omega_i^2\right)}_{\text{const.}} \int \mathrm{d}f_i \underbrace{g\left(f_i^2\right)}_{\text{const.}} \delta(\omega-\omega_{\mathrm{new}}) \\ &= 2M\int \mathrm{d}\omega_i\omega_i \int \mathrm{d}x_0 \underbrace{\left(\omega_i^2 + 3\frac{A}{M}x_0^2\right)}_{\frac{1}{M}\partial f_i/\partial x_0} \times \delta\left(\omega - \sqrt{\omega_i^2 + 3\frac{A}{M}x_0^2}\right) \end{aligned} \tag{26}$$

This contribution is proportional to ω^4.

The authors claim that the cross-over between the ω^4 behavior and the linear ω behavior of the local DOS produces the boson peak.

We now want to make clear that there is also another reason for a low-frequency contribution $\propto\omega^4$. The local oscillators not only couple statically to the strains but also dynamically by the interaction (2) [26,44]. This introduces the *local strain susceptibility* as a self energy into the local DOS:

$$G(\omega) = \frac{1}{\left[G_i^{(0)}(\omega)\right]^{-1} - \lambda\chi_i(\omega)} \tag{27}$$

Where

$$x_i(w) \propto \int \mathrm{d}^3\mathbf{k}\frac{k^2}{-w^2 + v_0^2k^2} \tag{28}$$

Taking the imaginary part and using (15) we have:

$$g_{\mathrm{loc}}(w) = 2w\mathrm{Im}\{G(w)\} \propto \mathrm{Im}\{x_i(w)\} \propto \int \mathrm{d}^3\mathrm{k}k^2\delta(w-vk) \propto w^4 \tag{29}$$

This relation holds for frequencies below the minimal resonance frequency. On the other hand, using cylinder coordinates ($k_\rho = \sqrt{k_x^2 + k_y^2}, k_z$):

$$\begin{aligned} \chi_i(\omega)'' &\propto \mathrm{d}^3\mathbf{k}k^2\delta(\omega-vk) \\ &= \int \mathrm{d}k_x k_\rho \int \mathrm{d}k_\rho \left(k_\rho^2 + k_z^2\right)\delta\left(\omega - v\sqrt{k_\rho^2 + k_z^2}\right) \end{aligned} \tag{30}$$

we see that this contribution has the same mathematical structure as that of the local forces (26). However, the physical meaning of Eqs.

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(26,30) is very different. The latter is an entirely harmonic contribution and generalizes to ω^{d+1} in d dimensions. The former is of anharmonic origin and is not d dependent.

The situation within the LO picture is now the following: Below a characteristic frequency ω_c the linear $g(\omega_i^2)$ (corresponding to a constant $g(\omega^2)$ of LO's) is – according to LO theory – *reconstructed* by local relaxation towards an ω^4 law. So this mechanism creates a "soft gap" within an initially structureless level spectrum. ω_c is interpreted as boson-peak frequency. On top of that, we argue, is *another* ω^4 contribution from the dynamic oscillator-phonon coupling, which will modify any estimates of the prefactors of the ω^4 and ω^1 contributions to the local DOS. Furthermore, the *measured* DOS will not only include the Debye DOS and the LO DOS, but also the contribution from fluctuating elasticity ΔK, be it static or from randomly distributed LO's.

3. Model with fluctuating shear modulus

We now work in terms of elasticity theory with the standard Lagrangian:

$$\mathcal{L} = \frac{1}{2}\rho_m\left[\dot{\mathbf{u}}(\mathbf{r},t)\right]^2 + \frac{1}{2}\lambda\left(\sum_{i=1}^{3} u_{ii}\right)^2 + \mu\sum_{i,j=1}^{3} u_{ij}^2, \tag{31}$$

where $\mathbf{u}(\mathbf{r},t)$ is the displacement vector, $u_{ij} = \frac{1}{2}[\partial_i u_j + \partial_j u_i]$ is the strain tensor and λ and μ are Lamé's elastic constants. $\mu \equiv G$ is the shear modulus and λ is related to the bulk modulus K by $K = \lambda + \frac{2}{3}\mu$. We now assume that the shear modulus μ fluctuates in space around its mean value μ_0 (the non-shear part of the bulk modulus $\lambda \equiv \lambda_o$ is supposed to be constant): $\mu(\mathrm{r}) = \mu_0 + \Delta\mu(\mathbf{r})$. This leads to the following equation of motion for the components of the displacement vector:

$$\ddot{u}_i(\mathbf{r},t) = \sum_j \left(\lambda_0\partial_i\partial_j + \partial_j\mu(\mathbf{r})\partial_i + \delta_{ij}\sum_l \partial_l\mu(\mathbf{r})\partial_l\right)u_j(\mathbf{r},t) \tag{32}$$

We assume that the correlation function of $\Delta\mu(\mathbf{r})$, $C(\mathbf{r}) = \langle\Delta\mu(\mathbf{r}+\mathbf{r}_0)\Delta\mu(\mathbf{r}_0)\rangle$ and its Fourier transform are of the forms:

$$\begin{aligned} C(\mathrm{r}) &= \langle\Delta\mu^2\rangle e^{-r/\xi} \\ C(\mathrm{k}) &= \langle\Delta\mu^2\rangle(8\pi/\xi)\left[k^2 + \xi^{-2}\right]^{-2} \end{aligned} \tag{33}$$

The corresponding mean-field equation for the low-wave number self energy $\Sigma(\omega) = \Sigma(q=0,\omega)$ (self-consistent Born approximation, SCBA) takes the form [12,23,27,50]:

$$\begin{aligned} \Sigma(\omega) &= \frac{\gamma}{2\varphi_3\langle\Delta\mu^2\rangle}\int_{|\mathbf{k}|<k_D}\left(\frac{\mathrm{d}\mathbf{k}}{2\pi}\right)^3 C(k)[\chi_L(k,\omega) + \chi_T(k,\omega)] \\ \chi_L(k,\omega) &= k^2\left[-\omega^2 + k^2\left(v_{L,0}^2 - 2\Sigma(\omega)\right)\right]^{-1} \\ \chi_T(k,\omega) &= k^2\left[-\omega^2 + k^2\left(v_{T,0}^2 - \Sigma(\omega)\right)\right]^{-1} \end{aligned} \tag{34}$$

Here $\gamma = \langle\Delta\mu^2\rangle\varphi_3 / v_0^4$. is the "disorder parameter", $v_{L,0}$, $v_{T,0}$, are the (unrenormalized) sound velocities, and $\varphi_3 = \int_{|\mathbf{k}|<k_D}(\mathrm{d}\mathbf{k}/2\pi)^3 C(\mathbf{k})$ is a normalization constant.

The DOS is given by:

$$g(\omega) = \frac{2\omega}{3\pi}\int_{|\mathbf{k}|<k_D}\left(\frac{\mathrm{d}\mathbf{k}}{2\pi}\right)^3\frac{1}{k^2}\mathrm{Im}\{\chi_L(k,\omega) + 2\chi_T(k,\omega)\}, \tag{35}$$

In Fig. 1 we have plotted the "reduced DOS" $g(\omega)/g_D(\omega)$ (where g_D is Debye's DOS) for three values of γ and four values of ξ. First we notice that, similarly to the uncorrelated case ($\xi \to 0$) [15,26,27] there exists a critical amount of disorder γ_c, beyond which the system becomes unstable. This instability is due to a situation in which non-negligible configurations with negative shear moduli exist. This situation is comparable with that of

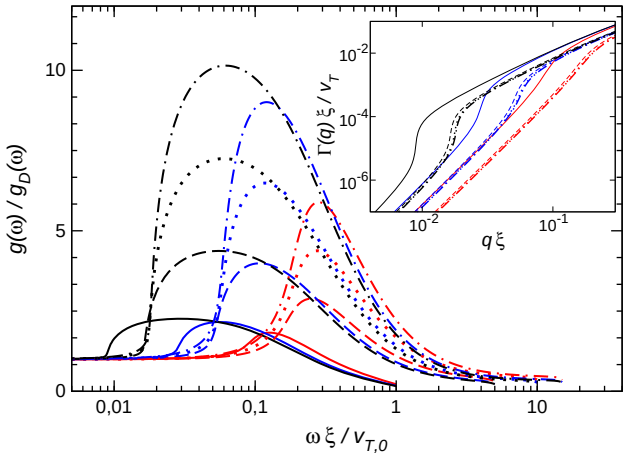

Fig. 1. Reduced DOS $g(\omega)/g_D(\omega)$ against scaled frequency $\omega\xi/v_{T,0}$ for $\xi = 1/k_D$ (full lines) $\xi = 5/k_D$ (dashed lines), $\xi = 10/k_D$ (dotted lines), $\xi = 15/k_D$ (dash–dotted lines), for three disorder parameters (from left to right) $\gamma - \gamma_c = 0.0001$ (black), 0.001 (blue) and 0.01 (red). Inset: Scaled Brillouin line width $\Gamma\xi/v_0$ against scaled wave number $q\xi$ for the same parameters and with the same line and color codes (figure taken from Ref. [29]). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

the LO model with interaction-induced instabilities. However, the philosophy behind the present model is that the input distribution of elastic constants is supposed to be the *relaxed* one, which should *not* include unstable configurations. It is, however, instructive to explore the origin of this instability, as it is related to that of the boson peak.

The quenched disorder induces an *effective interaction* between the vibrational modes, which leads to *level repulsion* between the eigenvalues. This occurs, where k is no more a "good quantum number". In the regime, where the modes are to good approximation plane waves, i.e. in the long-wavelength regime $k \to 0$, there is no level repulsion, and one obtains the Debye spectrum.

Level repulsion is a characteristic feature of *random matrices*. If we would discretize the equation of motion (32) the fluctuating part of the resulting dynamical matrix would be a random matrix, albeit a rather sparse one, compared with those dealt with in random matrix theory [49]. The characteristic level-distance statistics (GOE statistics) has been verified in a model with fluctuating force constants of a dynamical matrix [22]. The boson peak now turns out to be the lower edge of the "random matrix states", which are subject to level repulsion and which have roughly a semi-circular density of levels $g(\omega^2)$. The instability occurs, once this lower boundary becomes smaller than zero. Approaching the instability the boson peak can be interpreted as the "precursor" of the instability or the "memory of the instability" which might have occurred during the quenching process, followed by relaxation processes towards stable configurations. Operationally this random-matrix regime is described within the present model-description by the frequency-dependent self energy $\Sigma(\omega)$.

Returning to the discussion of Fig. 1 we notice that the boson peak frequency scales with the inverse correlation length, as we have plotted against $\omega\xi/v_{T,0}$. In contrast to the uncorrelated case ($C(\mathrm{r}) \propto \delta(\mathrm{r})$) the height of the boson peak is not bounded.

The inelastic coherent neutron and X-ray spectrum is given by the 1-phonon dynamical structure factor:

$$\begin{aligned} S(k,\omega) &= \frac{1}{\pi}(n(\omega)+1)\mathrm{Im}\{\chi_L(k,\omega)\} = \frac{1}{\pi}(n(\omega)+1) \\ &\quad \times 2k^4\Sigma''(\omega)\left\{\left[k^2 c_L(\omega)^2 - \omega^2\right]^2 + 4k^4\Sigma''(\omega)^2\right\}^{-1} \\ &\approx \frac{1}{\pi}[n(\omega)+1]\frac{k^2}{2\omega}\frac{k^2\Sigma''(\omega)/\omega}{[c_L(\omega)k-\omega]^2 + [k^2\Sigma''(\omega)/\omega]^2} \end{aligned} \tag{36}$$

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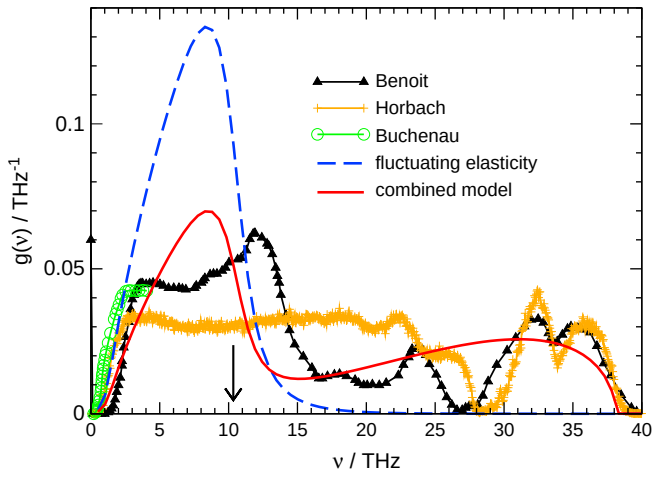

Fig. 2. Density of state of SiO_2. Benoit: first-principle simulation [51]; Horbach: MD simulation [6]; Buchenau: neutron data [1]; fluctuating-elasticity (FE): evaluation of Eqs. (34) and (35) with the parameters $\nu_D = 10.36$ (indicated by an arrow) $\gamma = 0.66$, $v_L/v_T = 1.5$, $\xi = 2k_D$; combined model: 50% of FE, 50% of a half-ellipse Eq. (3) with half width $B = 27$ THz.

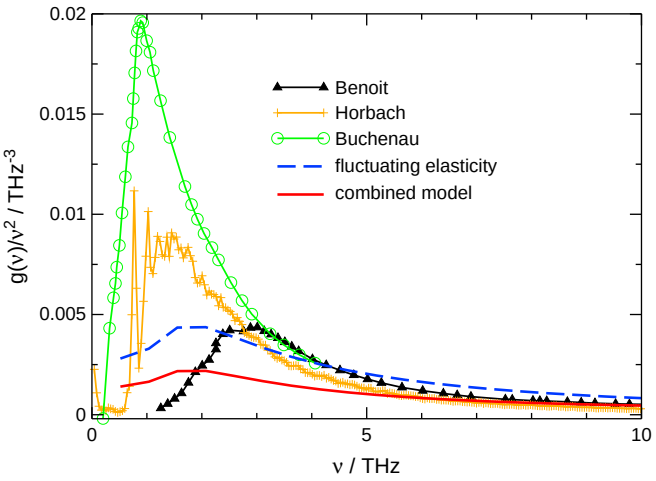

Fig. 3. Same data as in Fig. 2, divided by ν^2.

From this equation we can read off the width of the Brillouin line (full width at half maximum):

$$\Gamma(\omega) \approx 2k^2\Sigma''(\omega)/\omega \approx \frac{2}{c_L(\omega)^2}\omega\Sigma''(\omega) \tag{37}$$

In the insert of Fig. 1 we have plotted $\Gamma(\omega)$ against $\omega\xi$. We observe a remarkable scaling property: for $\xi \gg 1/k_D$ the quantity $\xi\Gamma$ is a universal function of $\omega\xi$ if the disorder parameter γ is fixed. This property can be verified by rescaling the self-consistency Eq. (34) [28]. Another interesting feature of $\Gamma(\omega)$ is its sudden increase just below the boson peak in the case where the disorder parameter γ is only slightly smaller than its critical value. For smaller γ this "step" becomes smeared out and eventually indistinguishable from the Rayleigh ω^4 behavior.

In the literature the possible presence or absence of Rayleigh scattering has been strongly debated [4]. From the experimental evidence it is clear that in the GHz regime and below the sound attenuation is proportional to ω^2. This behavior has been called "Akhiezer law" according to an old calculation of transverse sound damping in crystals [52], but has been shown by numerical simulations [53] and by a quantum-field-theoretical evaluation [54] to arise from anharmonic interactions. From the analytical theory [54] it emerges that the anharmonic sound attenuation is proportional to the temperature, so that by reducing the temperature one has the chance to observe Rayleigh scattering in disordered solids. This is a challenge for future experimental work. Whether there are traces of the Rayleigh scattering below the boson peak in SiO_2 is currently controversely discussed [55,56].

4. Comparison and tentative combination of the FE and LO models for glassy SiO_2

We turn now to a critical assessment of the fluctuating-elasticity (FE) and local oscillator (LO) approach towards the boson-peak anomaly. From the physics it must be clear that in an amorphous solid *both*, spatially fluctuating elasticity *and* local oscillatory motions are present. So attempts to find out "which model is better" [57] are doomed to failure. The *renormalized* resonance frequencies in the relaxed state will have a lower bound ω_c according to the soft-potential theory [8,43,48]. We convinced ourselves in Section 2 that *below resonance* the local oscillators can be subsumed into the fluctuating-elasticity framework. To say it clearly: The frozen-disorder aspect of the local motion is already contained in the fluctuating-elasticity picture. Only the *resonant frequencies* appear as an additional feature in the spectrum. This also answers the long-debated question: "Are there *optical modes* in an amorphous solid?" From a solid-state-textbook standpoint the answer is, of course, "No", because there is neither long-range topological nor long-range chemical order present in an amorphous material. What *is* present, is an interaction-renormalized spectrum of local oscillatory motions like the famous librations of tetrahedra [1].

That this is so can be clearly seen from the DOS of glassy SiO_2, depicted in Fig. 2. We show there the neutron data [1] together with a molecular-dynamics simulation [6] using the BKS pair potential and a more recent ab-initio calculation [51]. In fact the higher frequency part resembles very much that of crystalline cristobalite [58] with its characteristic "floppy" librational, bond-bending and stretching modes. All these modes can be viewed to form in the amorphous states local oscillators, which are coupled to the low-frequency acoustic excitations.

Now we have argued that the disorder aspect of this coupling is already contained in the fluctuating-elasticity approach. The resonant aspect then can just be added to the spectrum. The simplest formula for a broad, inhomogeneous and bounded spectrum of levels ω^2 is a half-ellipse, normalized to one, which for $g_i(\omega)$ takes the form:

$$g_i(\omega) = 2\omega g_i\left(\omega^2\right) = 2\omega\frac{2}{\pi B^2}\sqrt{B^2-\omega^4} \tag{38}$$

B is the half width, i.e. one of the half axes of the ellipse (for the other one we have $A = 2/\pi B$).

In Fig. 3 we have added to the experimental and simulated DOS two calculated model functions. The first model function is the DOS resulting from the FE model, which is, by construction, limited to the Debye frequency $\omega_D = 2\pi\nu_D$. ω_D has been calculated from the measured longitudinal and transverse sound velocities and the averaged density of atoms [27]. Clearly this does not contain the would-be optical oscillations and librations. As the total DOS is normalized to unity the FE DOS is much higher than the simulated and measured one. In the second model calculation we just superimposed equally the FE DOS with a "local oscillator DOS" of the form (3) with $2B = 54$ THz, roughly the width of the total spectrum. In view of the differences of the simulated spectra this superposition does not do a bad job.

We now turn to the reduced DOS $g(\nu)/\nu^2$, featuring the boson peak. This representation of the data is presented in Fig. 3. We see that the boson peaks of the experiment and the two simulations are quite different. From this one concludes that the boson peak is more sensitive to disordered-sample details than the overall DOS. If the

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boson peak would result from a resonant spectrum this could not be understood. On the other hand, if it stems from the mean-square fluctuations of the quenched disorder it is easily understood, because both in real materials as well as in simulations the statistics of the quenched disorder is strongly dependent on the preparation and annealing history. We did not attempt to obtain a "best fit" to any of the data, but emphasize that the present approach allows for a qualitative understanding of the underlying physics.

In passing we also mention that the neutron scattering DOS is obtained from averaging over a high-wave number part of $S(q,\omega)$, measured with *coherently* scattering nuclei. While it is widely believed that this procedure (incoherent approximation [59]) gives a good account of the DOS, it still might over-estimate the boson peak.

5. Conclusions

We have tried to obtain a unified view of possible spectral mechanisms which may be responsible for forming the boson-peak anomaly in the THz regime of amorphous materials. The fluctuating-elasticity model has been compared with the local oscillator model developed in the framework of soft-potential theory. The fluctuating-elasticity model enables to classify the disorder-induced vibrational modes as "random-matrix" modes. These modes have the property of spectral level repulsion, which becomes distinct at the Ioffe–Regel point, where the wave vector ceases to be a good "quantum number" for labeling the modes. We have found that local oscillators, if treated on the same footing as fluctuating elasticity, can contribute to this fluctuating elasticity away from (i.e. below) the oscillator resonances. This opens the possibility to subsume the disorder-related aspects of the local excitations in the fluctuating-elasticity formalism and treat the resonant spectrum separately. This procedure gives a qualitative understanding of the vibrational spectrum of a network glass like SiO_2.

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References

[1] U. Buchenau, N. Nücker, A.J. Dianoux, Phys. Rev. Lett. 24 (1984) 2316.
[2] W.A. Philips (Ed.), Amorphous Solids — Low Temperature Properties, Springer, Berlin, 1981.
[3] S.R. Elliott, Physics of amorphous materials, 2nd ed.Longman, New York, 1990.
[4] For extensive literature and recent discussions of the boson peak see e.g [5, 6, 7, 8, 9].
[5] K. Binder, W. Kob, Glassy materials and disordered solids, World Scientific, Singapore, 2005.
[6] J. Horbach, W. Kob, K. Binder, Eur. Phys. J. B 19 (2001) 531.
[7] T. Nakayama, Rep. Prog. Phys. 65 (2002) 1195.
[8] V.L. Gurevich, D.A. Parshin, H.R. Schober, Phys. Rev. B 67 (2003) 094203 71, 014209 (2005).
[9] G. Baldi, V.M. Giordano, G. Monaco, F. Sette, E. Fabini, A. Fontana, G. Ruocco, Phys. Rev. B 77 (2008) 214309.
[10] Flubacher, Leadbetter, Morrison,, Stoicheff, J. Phys. Chem. Sol. 12 (1959) 53.
[11] See J. Jäckle in Ref. [2]. For an extended literature concerning Raman scattering on glasses see [12].
[12] B. Schmid, W. Schirmacher, Phys. Rev. Lett. 100, 137402 (2008); 103, 169702 (2010); see also T. Unruh, A. Schulte, B. Schmid, and W. Schirmacher, this volume.
[13] R. Shuker, R. Gammon, Phys. Rev. Lett. 25 (1970) 222.
[14] The Raman intensity, in fact is not proportional to the vibrational DOS, see [12] and the discussion below.
[15] W. Schirmacher, Europhys. Lett. 73 (2006) 892.
[16] In their compilation of electronic transport properties of semiconductors Ioffe and Regel' [17] emphasize that one should not apply the conductivity formula derived from lowest-order scattering theory (Drude's formula), once the mean-free path entering the formula becomes comparable with (or smaller than) the De-Broglie wavelength. In this paper nothing about wave localization can be found.
[17] A.F. Ioffe, A.R. Regel, Prog. Semicond. 4 (1960) 237.
[18] J.E. Graebner, B. Golding, L.C. Allen, Phys. Rev. B 43 (1986) 5696.
[19] S.R. Elliott, Europhys. Lett. 19 (1992) 201.
[20] P.B. Allen, J.L. Feldman, Phys. Rev. Lett. 62 (1989) 645; P.B. Allen, J.L. Feldman, Phys. Rev. B: 48 (1993) 12581.J.L. Feldman et al., ibid., p. 12589.
[21] W. Schirmacher, M. Wagener, Solid State Commun. 86 (1993) 597.
[22] W. Schirmacher, G. Diezemann, C. Ganter, Phys. Rev. Lett. 81 (1998) 136.
[23] S. John, H. Sompolinsky, M.J. Stephen, Phys. Rev. B 27 (1983) 5592; S. John, M.J. Stephen, Phys. Rev. B 28 (1983) 6358.
[24] J. Fabian, P.B. Allen, Phys. Rev. Lett. 77 (1996) 3839.
[25] G. Ruocco, F. Sette, J. Phys. Condens. Matter 13 (2001) 9141.
[26] E. Maurer, W. Schirmacher, J. Low Temp. Phys. 137 (2004) 453.
[27] W. Schirmacher, G. Ruocco, T. Scopigno, Phys. Rev. Lett. 98 (2007) 025501.
[28] W. Schirmacher, B. Schmid, C. Tomaras, G. Viliani, G. Baldi, G. Ruocco, T. Scopigno, Phys. Status Solidi A c) 5 (2008) 862 a.
[29] W. Schirmacher, C. Tomaras, B. Schmid, G. Baldi, G. Viliani, G. Ruocco, T. Scopigno, submitted to J. Condensed Matter (Ukraine).
[30] S.N. Taraskin, Y.H. Loh, G. Natarajan, S.R. Elliott, Phys. Rev. Lett. 86 (2001) 1255.
[31] V. Martin-Mayor, G. Parisi, P. Verroccio, Phys. Rev. E 62 (2000) 2373.
[32] T.S. Grigera, V. Martin-Mayor, G. Parisi, P. Verroccio, Phys. Rev. Lett. 87 (2001) 085502; T.S. Grigera, V. Martin-Mayor, G. Parisi, P. Verroccio, Nature 422 (2003) 289.
[33] J.W. Kantelhardt, S. Russ, A. Bunde, Phys. Rev. B 63 (2001) 064302.
[34] R. Kühn, U. Horstmann, Phys. Rev. Lett. 78 (1997) 4067.
[35] W. Götze, M.R. Mayr, Phys. Rev. E 61 (2000) 587.
[36] S.-H. Chong, Phys. Rev. E 74 (2006) 031205.
[37] N. Xu, M. Wyart, A.J. Liu, S.R. Nagel, Phys. Rev. Lett. 98 (2007) 175502.
[38] V. Lubchenko, P.G. Wolynes, PNAS 100 (4) (2003) 1515.
[39] C.A. Angell, Y. Yue, L.-M. Wang, J.R.D. Copley, S. Brick, S. Mossa, J. Phys. Condens. Matter 15 (2003) S1051.
[40] E. N. Economou, Green's Functions in Quantum Physics, 2nd Edition, Springer-Verlag Heidelberg. See $ 6.3.4 on p. 122.
[41] A.L. Burin, L.A. Maksimov, I.Ya. Polishchuk, Physica B 210 (1995) 49.
[42] I.Ya. Polishchuk, L.A. Maksimov, A.L. Burin, Phys. Rep. 288 (1997) 205.
[43] V.G. Karpov, M.I. Klinger, F.N. Ignatev, Zh. Eksp. Teor. Fiz 84 (1983) 760; U. Buchenau, Yu.M. Galperin, V.L. Gurevich, H.R. Schober, Phys. Rev. B 43 (1991) 5039 ibid, 46, 2798 (1992).
[44] U. Buchenau, Yu.M. Galperin, V.L. Gurevich, D.A. Parshin, M.A. Ramos, H.R. Schober, Phys. Rev. B 46 (1992) 2798.
[45] L. Gil, M.A. Ramos, A. Bringer, U. Buchenau, Phys. Rev. Lett. 70 (1993) 182.
[46] E. Duval, A. Boukenter, T. Archibat, J. Phys. Condens. Matter 2 (1990) 10227; E. Duval, A. Mermet, L. Saviot, Phys. Rev. Lett. 75 (2007) 024201.
[47] For the waves we work with periodic boundary conditions w. r. to the sample volume V, which discretizes the components of $\mathbf{k}$, and we have $f(\mathbf{r}) = \sum_{\mathbf{k}} e^{-i\mathbf{kr}} f(\mathbf{k})$ with $\sum_{\mathbf{k}} = \frac{V}{(2\pi)^3} \mathrm{d}^3\mathbf{k}$ and we have $f(\mathbf{k}) = \frac{1}{V}\int_{\mathbf{r}\in V} e^{i\mathbf{kr}} f(\mathbf{r})$.
[48] V.G. Karpov, D.A. Parshin, JETP Lett. 38 (1983) 648; V.G. Karpov, D.A. Parshin, Sov. Phys. JETP 61 (1985) 1308; V.L. Gurevich, D.A. Parshin, H.R. Schober, JETP Lett. 76 (2002) 553.
[49] M.L. Mehta, Random matrices, 3 rd ed.Elsevier/Academic Press, Amsterdam, 2004.
[50] V.A. Ignatchenko, V.A. Felk, Phys. Rev. B 74 (2006) 174415.
[51] M. Benoit, W. Kob, Europhys. Lett. 60 (2002) 269.
[52] A. Akhiezer, J. Phys. 1 (1939) 277 Moscow.
[53] P. B. Allen, J.L. Feldmann, Phys. Rev. B 48, 12581J.L. Feldman, M.D. Kluge, P.B. Allen, F. Wooten, Phys. Rev. B 48 (1993) 12589.
[54] C. Tomaras, B. Schmid, W. Schirmacher, Phys. Rev. B 81 (2010) 104206.see also C. Tomaras, B. Schmid and W. Schirmacher in this volume.
[55] C. Masciovecchio, G. Baldi, S. Caponi, L. Comez, S. Di Fonzo, D. Fioretto, A. Fontana, A. Gessini, S.C. Santucci, F. Sette, G. Viliani, P. VIlmercati, G. Ruocco, Phys. Rev. Lett. 97 (2006) 035501.
[56] A. Devos, M. Foret, S. Ayrinhac, P. Emery, B. Rufflé, Phys. Rev. B 77 (2008) 100201.
[57] B. Rufflé, D.A. Parshin, E. Courtens, R. Vacher, Phys. Rev. Lett. 100 (2008) 015501.
[58] I.P. Swainson, M.T. Dove, Phys. Rev. Lett. 71 (1993) 193; M.T. Dove, M.J. Harris, A.C. Hannon, J.M. Parker, I.P. Swainson, M. Gambhir, Phys. Rev. Lett. 78 (1997) 1070.
[59] U. Buchenau, Z. Phys. B: Condens. Matter 58 (1985) 181 Condensed M.

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