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

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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.

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## 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 $\Omega_k$ 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 $\Gamma_k$ 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 $\omega$ 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]:

$$\underbrace{\left(-\omega^2 + K_0 k^2\right)}_{G_0^{-1}(\mathbf{k},\omega)} G(\mathbf{k},\mathbf{k}',\omega)\delta_{\mathbf{k},\mathbf{k}'} = \delta_{\mathbf{k},\mathbf{k}'} - \sum_{\mathbf{q}} \mathbf{k}\cdot\mathbf{q}\Delta K(\mathbf{k}-\mathbf{q})G(\mathbf{q},\mathbf{k}'\omega) \tag{6}$$

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:

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

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 $\Delta K$ 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:

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

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 $\Delta K$ 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:

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

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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