Vibrational excitations in disordered solids

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The vibrational excitations of disordered solids differ appreciably from those of crystals. The reason for this anomalous behaviour can be traced to the absence of the lattice symmetry, i.e., to the structural disorder. We review the experimental findings and simulational results of the vibrational spectrum of disordered solids, in particular of glasses. We further give an overview of the existent pertinent models and theoretical treatments for explaining the vibrational anomalies, in particular the enhancement of the vibrational density of states with respect to the Debye law ("boson peak").

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I. INTRODUCTION

The vibrational excitations of disordered solids, in particular of glasses, have attracted the attention of experimental [21, 33, 35, 42, 67, 101, 124, 134, 169, 206, 215] and theoretical work [32, 53, 69, 72, 73, 79, 90, 93, 98, 104, 113, 116, 118, 120, 125, 136, 152, 153, 155–157, 171] (to cite only a few at the beginning) in the last threequarter century. In crystals the harmonic vibrational excitations – the phonons – can be satisfactorily described by the symmetry properties of the lattice [15]. In modern crystalline-lattice dynamics investigations [123], therefore, the emphasis is on anharmonic effects. On the other hand, in disordered solids, due to the absence of the lattice symmetries, even the theoretical descriptions of the harmonic vibrational excitations poses a challenge. Scientific efforts facing this challenge are still a very active part of condensed-matter research.

1. Density of states (DOS)

In a solid the harmonic part of the potential energy, i.e. the part, which is bilinear in the displacements $\mathbf{u}_i(\mathbf{r}, t)$ (where *i* denotes the center of mass of a molecule) can be written as $E_{pot} = \sum_{ij} \frac{1}{2} \mathbf{u}_i \stackrel{\leftrightarrow}{D}_{ij} \mathbf{u}_j$ with the dynamical (or Hessian) matrix

$$D_{ij}^{\alpha\beta} = \frac{\partial}{\partial r_i^{\alpha}} \frac{\partial}{\partial r_j^{\beta}} E_{pot}(\mathbf{r}_1, \dots, \mathbf{r}_N) \,. \tag{1}$$

The DOS is then given by

$$g(\omega) = \frac{1}{3N} \left\langle \sum_{\mu=1}^{3N} \delta(\omega_{-}\omega_{\mu}) \right\rangle , \qquad (2)$$

where the eigenfrequency ω_{μ} are the square-roots of the eigenvalues $\lambda_{\mu} = \omega_{\mu}^2$ of the dynamical matrix, and N is the number of atoms/molecules.

We have started our review of vibrational excitations in disordered solids by introducing the dynamical matrix and the corresponding DOS, because this concept remains valid going from crystalline to amorphous solids. Concepts like Bloch's theorem, leading to dispersion relations in reciprocal space are not valid in the absence of crystalline order, but the DOS, the normalized histogram of eigenfrequencies, can be considered for any type of arrangements of the atomic or molecular sites \mathbf{r}_i . This is the reason, why most of the experimental and simulational efforts dedicated to the vibrational spectra of glasses¹ are aimed at finding out the DOS, i.e. the vibrational spectrum of the material.

2. The boson peak

A paradigm for the anomalous vibrational features of glasses is the so-called boson peak (BP), which is an enhancement of the DOS with respect to Debye's $g(\omega) \propto \omega^2$

 $^{^1}$ We take "glasses" and "amorphous solids" as synonyms, independent of the preparation process.



FIG. 1. Top: Specific heat, divided by T^3 of several materials against temperature T (from [213]).

Bottom: Thermal conductivity of several glassy materials against temperature (from [70]). Note that, compared to glassy SiO₂, at T = 10 K, the thermal conductivity of α -quartz is 4 orders of magnitude larger [25, 36].

law. The latter transforms to a T^3 law for the temperature dependence of the specific heat C(T) [15]. In contrast to Debye's $C(T) \propto \hat{T}^3$ prediction, in glasses at low temperatures, C(T) behaves completely in a different way. At very low temperatures, in the range around ~ 1K, C(T) varies almost linearly with temperature [100, 215], and the thermal conductivity (which is orders of magnitude smaller than that of crystals) quadratically [25, 36, 100, 215]. This behaviour has been successfully ascribed to bistable states in the amorphous structure, between which quantum-mechanical tunneling becomes possible (tunneling states, two-level systems [10, 140, 141]). At slightly higher temperatures, the specific heat of glasses still does not follow Debye's T^3 law. If plotted as $C_V(T)/T^3$ one observes a broad maximum around 10 K, see the upper panel of Fig. 1. In the same temperature regime, the temperature variation of the thermal conductivity exhibits a pronounced shoulder, as is shown in the lower panel of Fig. 1. It seems likely that this deviation from Debye's $C(T) \propto T^3$ law must be due to a deviation of Debye's $q(\omega) \propto \omega^2$ law



FIG. 2. Raman spectrum of glassy and crystalline SiO_2 (from Shuker and Gammon [173]).

for the DOS. Indeed, in Raman spectra, which – in some way (see below) – represent the DOS, a broad continuum in the ~ 50 cm⁻¹ (\doteq 1.3 THz) range² was found, in a frequency region, where there is no intensity in the corresponding crystal, see Fig. 2. It was further found that the anomalous Raman spectrum of glasses in the frequency range below 100 cm⁻¹ precisely follows the temperature dependence of the boson occupation factor $n(\omega) + 1 = [1 - e^{-\hbar\omega/k_BT}]^{-1}$. For this reason the lowfrequency spectral anomaly in the 1 THz range was called "boson peak" [90]. Shuker and Gammon [172] argued, that in disordered solids the selection rules for Raman scattering [29] do not apply and came up with a formula for the (depolarized = VH) Raman spectrum

$$I_{VH}(\omega) \propto C(\omega)[n(\omega)+1]\frac{g(\omega)}{\omega}$$
. (3)

Here $C(\omega)$ is the so-called light-vibration coupling coefficient [90, 161, 162, 198, 199], which Shuker and Gammon [172] assumed to be frequency-independent. Now, if the temperature dependence of the Raman intensity is due to the boson occupation factor, the conclusion is that the spectrum $g(\omega)/\omega$ must be temperature independent, and that, consequently, the BP anomaly in the DOS must be a harmonic effect. This means that the boson peak must be a feature of the harmonic degrees of freedom³.

Further, in a seminal paper Buchenau *et al.* [33], using inelastic neutron scattering, showed a strong deviation of the DOS of SiO₂ glass from the ω^2 law, which, if

² The Raman intensity is usually reported as a function of $\nu/c = \omega/2\pi c$, which has the unit of an inverse length. 32 cm^{-1} (in the Raman slang "32 wavenumbers") correspond to $\nu = 1$ THz and to $h\nu = \hbar\omega = 4$ meV.

³ Recent theoretical efforts, claiming that the BP anomaly would be of anharmonic origin [17], turned out to be mathematically in error [174].

represented as $g(\omega)/\omega^2$ shows up as a maximum. They identified this with the Raman boson peak, although the Raman (would-be) DOS did not agree to that extracted from the neutron spectra. In order to cope with this discrepancy, one introduced the frequency dependence of the coefficient $C(\omega)$ [90, 161, 162, 198, 199], into the Shuker-Gammon formula (3). Because the deviation of the specific heat of glasses from the T^3 law is due to the boson peak in the DOS, the maximum in the temperature variation of $C(T)/T^3$ of glasses is also called boson peak [146].

The boson peak and related vibrational anomalies are observed in most glassy or disordered materials, and almost the entire scientific work on vibrational excitations in glasses has been devoted to uncover the nature of the (harmonic) vibrational wave functions associated with the BP. The present review simply cannot cover the overwhelmingly large amount of literature published on this subject. We just select some key results, indicating in some way or another the origin of the anomaly. In fact, as we shall state in the conclusion, the matter is still highly controversial.

We start (section II.) by enumerating (most of) the suggestions put forward in the past for explaining the boson peak. In sections III. and IV. we try to give an overview over the vast amount of experimental and simulational work on the present topic. In the final section V. we discuss the present status of the interpretation of the boson-peak-related vibrational anomalies of glasses.

II. MODELS AND SUGGESTIONS FOR EXPLAINING THE BP ANOMALIES

In contrast to the rather convincing explanation of the linear-T behaviour of the specific heat and the T^2 behaviour of the thermal conductivity in the ~ 1 K range in terms of bistable tunneling defects [10, 140] the boson peak (BP) was from the beginning subject to rather diverse modelling, which continues until now. We here give a brief historical overview and refer to the quoted original literature for more details.

• Karpov, Klinger and Ignat'ev [93] tried to formulate a classical version of the tunneling model, the softpotential model. This model features a set of structural defects with anharmonic potentials with very small positive and negative quadratic (stiffness) terms. The (renormalized) density of states of such defects can be shown [31, 32, 78, 80, 93] to vary as ω^4 , due to a swallowtail singularity in the classification of catastrophe theory [14, 191].

• Orbach [132] conjectured that glasses (in particular network glasses) might exhibit a self-similar (fractal) structure. Such structures, imbedded in a homogeneous medium at larger scales were known to exhibit a transition from a Debye-type spectrum $g(\omega) \propto \omega^2$ waves (phonons) at low frequencies (large wavelengths) to fractal-type behaviour $g(\omega) \propto \omega^{1/3}$ at higher frequen-



FIG. 3. Sketch of a typical radial distribution function g(r) of an amorphous (monatomic) structure, which approaches the decorrelated value $g(r) \rightarrow 1$.

cies ("phonon-fracton crossover", Alexander and Orbach [6], Nakayama *et al.* [129]). Simulations of percolating networks, which are examples of such imbedded fractal structures [129, 211] indeed showed such a crossover, but, unfortunately, no boson-peak-type enhancement. Furthermore, no traces of self-similar structure were found experimentally in glasses, except in loose structures such as aerogels [197] or biopolymers [126].

• According to an argument of Ioffe and Regel [87], (electronic) waves with a scattering mean-free path ℓ smaller than its wavelength cannot exist. This lead Mott [128] to the conclusion that waves in the presence of disorder, which would produce an attenuation with a mean-free path shorter than the wavelength, become localized by disorder (Anderson-localization, [1, 9]). This argument was taken over in papers discussing BP-related anomalies [4, 63, 66, 75], in which it was conjectured that the anomalies are due to phonon localization [91, 94].

• A step forward in the understanding of the BP anomalies was the investigation of disordered-force constant models by making contact with hopping transport in disordered systems. It was noted by Alexander *et al.* [5] that the excitation dynamics of a harmonic system of masses m connected with force constants K_{ij} , which obeys an equation of motion

$$m\frac{d^2}{dt^2}u_i = -\sum_j K_{ij}(u_i - u_j) \tag{4}$$

with scalar "displacements" $u_i = u(\mathbf{r}_i)$ can be mapped to a disordered random-walk (hopping) problem by replacing the double time derivative with a single one. Schirmacher and Wagener [158] noted that the cross-over of the AC conductivity $\sigma(\omega)$ from being frequency-independent to a strong disorder-induced frequency dependence [59] corresponds in the analogous harmonic force constant system to a cross-over from a frequency independent to a frequency dependent (squared) sound velocity (elastic constant). They realized that this crossover produces a boson peak and corroborated it with calculations by the coherent-potential approximation (CPA) [159, 160]. Using the self-consistent localization theory [61, 200], Schirmacher and Wagener [160] predicted phonon Anderson



FIG. 4. Reduced DOS $g(\omega)/\omega^2$ 2 versus frequency for Gaussian force-constant distributions with $\sigma/K_0 = 1$ and several lower cutoffs (in units of K_0). The symbols represent the numerical diagonalization, the full lines the CPA results. The agreement is achieved without any adjustable parameters. From [153].

localization to happen much above the boson peak, near the upper band edge.

• A pathfinding paper on the boson-peak anomaly was published 1992 by S. R. Elliott [63]. He argued that in an amorphous structure there exists a length scale at which the atomic structure is no more distinct. This length scale is the correlation length and describes the decay of the radial distribution function⁴ g(r) towards its decorrelated value 1 (s. Fig. 3). On a scale larger than ξ the material looks homogeneous and isotropic, therefore, so the argument of Elliott, waves with wavelength λ larger than ξ can be supported by the material. Waves with smaller λ become rather strongly scattered, and, with increasing frequency, the vibrational excitation loses gradually its wave character, as formulated by Ioffe and Regel [87]. Converting length scales to frequency scales by the transverse velocity v_T , which is roughly equal to the Deby velocity $v_D = \omega_D / k_D$ (Debye frequency ω_D , divided by the Debye wavenumber $k_D = \sqrt[3]{6\pi^2 N/V} = \sqrt[3]{6\pi^2}/a$, where a is an intermolecular spacing) Elliott [63] conjectured that the frequency at which disorder-induced deviations from the Debye wave physics should occur near $\omega_B = v_T / \xi \sim \omega_D a / \xi$, i.e. roughly 1/10 of the Debye frequency. This is precisely the frequency range of ~ 1 THz, around which the boson peak is observed in most materials [134]. A correlation between the boson-peak position and v_T/ξ is indeed observed in many glasses [55, 63, 134].



FIG. 5. Thermal conductivity calculated in self-consistent Born approximation (SCBA) [152] for three values of the disorder parameter $\gamma \propto \langle (\Delta G)^2 \rangle$, compared with the data of Fig. 1 [70], scaled with the Debye temperature Θ_D . The red arrows correspond both to the boson peak positions of the specific heat, calculated in SCBA as those of the experiments (from Schirmacher [152]).

• In a model calculation on a 3-dimensional lattice with randomly distribution of force constants Schirmacher et al. [153] considered a mass-spring system of the type (4) on a simple-cubic lattice with Gaussian forceconstant disorder, truncated from below was treated both by CPA and by numerical diagonalization. In Fig. 4 we show their result for the reduced DOS $q(\omega)/\omega^2$. First, the agreement of the CPA with the numerical data showed that the CPA is a reliable mean-field theory for disorder. In both calculations a pronounced BP is visible (Fig. 4). The inclusion of negative force constants demonstrated that the BP is strongly enhanced by the presence of negative force constants and is a precursor of an instability, which occurs in the presence of too many negative force constants. The authors further evaluated the mean-free path $\ell(\omega)$, and found by comparison with the wavelength $\lambda(\omega)$ that the BP position is near the Ioffe-Regel limit, where $\ell(\omega) \sim \lambda(\omega)$. The authors also evaluated the leveldistance statistics of the eigenvalues and demonstrated that the vibrational states near and above the BP are de*localized*, because the statistics follows that of the Gaussian Orthogonal Ensemble (GOU) of random matrices [119, 178]. A mobility edge, above which the states are localized, appears only very near the upper end of the band, thus corroborating the predictions of Schirmacher and Wagener [160]. It was proved later in numerical investigations of a similar force-constant model [142, 143] that these states obey the Porter-Thomas statistics [144], i.e., are of the same type as random-matrix eigenstates.

• That in a disordered harmonic system an extended frequency region exists, in which the vibrational states are neither propagating nor localized had already been found by Allen et al. [7, 8, 65] in a simulation of amorphous Si. Allen *et al.* [8] called the corresponding excitatins "diffusions", because the intensity of such waves obey a diffusion equation like light in milky glass [88]. They called the Debye waves below the Ioffe-Regel

⁴ g(r) - 1 is the Fourier transform of the structure factor S(k) minus 1 [62, 81] of the amorphous material, which can be measured by elastic X-ray or neutron diffraction. $4\pi r^2 g(r) N/V$ (particle number N volume V) gives the probability density for the presence of other atoms/molecules with distance r from a given one at the origin.

crossover "propagons" and the localized states near the Debye frequency "locons". They deliberately avoided the term "phonons", because phonons are by definition the eigenstates of a crystal.

• A model calculation similar to that of Schirmacher et al. [153] (a crystal with force-constant disorder) was presented later by Taraskin et al. [190], but with a somewhat different interpretation. They observed - similar to the findings in [153] that - with increasing disorder - the lowest van-Hove singularity⁵ of the lattice broadens, moves down in frequency, and then smoothly goes over to the low-frequency BP. They concluded that the BP is essentially due to the levelling off of the transverse phonon branch and not so much due to disorder. This view was corroborated later by experimentalists who compared silicate glassses with their crystalline counterparts [44, 45, 218] (see below).

• Grigera *et al.* [77] investigated a disordered massspring model (Euclidean random-matrix model, ERM) by a diagrammatic procedure. Inspired by the behaviour of a mean-field theory obtained from a class of diagrams, they obtained a BP-type anomaly, which – with increasing disorder – leads to an instability like the CPA of Schirmacher *et al.* [153] with too many negative force constants. This instability was interpreted as a transition from a mimima-dominated potential-energy surface (PES) to a saddle-dominated PES. The ERM model was shown to imply Rayleigh scattering, i.e. a sound attenuation $\Gamma(\omega) \propto \omega^4$ [72, 76, 154], in agreement to the disordered-lattice calculations [153, 190] and the HET theory (see below).

• Schirmacher [152] formulated a heterogeneouselasticity theory (HET), which is a theory of elasticity, in which the shear modulus G exhibits spatial fluctuations. This theory was solved for the averaged DOS by field-theoretical techniques for a mean-field theory (Selfconsistent Born approximation, SCBA), which predicts

- a disorder-induced boson peak independent of an underlying lattice;

- a disorder-induced Rayleigh-like sound attenuation $\Gamma(\omega) \propto \omega^4;$

- a minimum of the transverse phase velocity $v_T(\omega)$ near the BP. This theory conforms with the previous conclusions that the BP is a phenomenon produced by the structural disorder of the glass. Within the same theoretical framework a theory for the thermal diffusivity was formulated. Combined with the inelastic scattering from two-level system, he found an explanation for the characteristic shoulder in the temperature dependence of the thermal conductivity: It is an *upside-down boson peak* (see Fig. 5).

The anomalous increase of the DOS above the Debye DOS was subsequently shown [155] to be related to the



FIG. 6. Top: $C(T)/C_D(T) \propto C(T)/T^3$ against T/Θ_D for four different (glassy) alcohols, compared with glassy glycerol (from Ramos *et al.* [147]).

Bottom: $[C(T) - \gamma T]/T^3$ for three metallic glasses. γT is the Sommerfeld contribution of the electrons (from Li *et al.* [107]).

Rayleigh ω^4 behavior of the sound attenuation: It was shown that the excess DOS is just proportional to the sound attenuation $\Gamma(\omega)$.

The SCBA version of HET, which only applies to Gaussian distributions of elastic moduli and moderate disorder was generalized by means of an off-lattice version of the CPA, again by field-theoretical techniques [95]. This version served recently to obtain a disorder classification of glasses [134].

• An important aspect in the discussion about the boson peak has been contributed by the theoretial investigation of the jamming transition [51, 52, 69, 108, 130, 207– 210]. The disordered solid is considered as a random packing of soft spheres with a finite range of interactions. The latter induce a number of constraints, which lead to a

⁵ A Van-Hove singularity occurs in the DOS of a crystal at a frequency, at which the phonon dispersion $\omega(k)$ becomes constant at the Brillouin-zone boundary.

finite shear elasticity. The instability of the solid – where the shear stiffness becomes zero – is reached at the isostatic point, where the number of constraints equal the number of degrees of freedom [117]. It was shown by simulations and effective-medium calculations that in such random packings near the jamming instability a boson peak appears in the vibrational spectrum. This was rationalized by a rather simple mean-field jamming model, the so-called perceptron [68, 69], which was introduced in connection with neural networks. The generic frequency dependence in the mean-field limit (dimension $d \to \infty$) is given by a modification of the the Marchenko-Pastur law of rectangular random matrices [112]

$$g(\omega) \propto \omega \frac{\sqrt{(\omega^2 - \omega_0^2)(\omega_{\max}^2 - \omega^2)}}{\omega^2 + \omega_*^2} \,. \tag{5}$$

In finite dimensions the gap below ω_0 is filled with Debyetype waves, so that one has in general [51, 69]

$$g(\omega) \propto \begin{cases} \omega^{d-1} & \omega \ll \omega_0 \\ \omega^2/\omega_0^2 & \omega_0 \ll \omega \ll \omega_* \\ \text{const.} & \omega_* \ll \omega \ll \omega_{\max} . \end{cases}$$
(6)

The resulting boson peak at ω_0 marks – as in the heterogeneous-elasticity theory – the crossover between the Debye wave regime and the random-matrix regime. The eigenvectors of random matrices [119] are known to be delocalized.

III. EXPERIMENTAL INVESTIGATIONS

1. Specific heat and thermal conductivity

The specific-heat BP, i.e. a maximum in the temperature variation of $C(T)/T^3$, see the top panel of Fig. 1, is observed in practically all glassy materials. As further examples we display boson peaks of a number of alcohols [147] and metallic glasses [107]. In the latter the Sommerfeld contribution due to the free electrons γT , with $\gamma = \frac{1}{3}\pi^2 k_B^2 N(E_F)$ ($N(E_F)$ is the electronic DOS at the Fermi level), must be subtracted, in order to obtain the vibrational contribution. It is interesting to note that in many cases the BP temperature T_{max} is proportional to the Debye temperature Θ_D .

We mentioned in the introduction (bottom panel of Fig. 1), that in the temperature variation of the thermal conductivity a characteristic shoulder (plateau) is observed near $T_{\rm max}$. Similar features are observed in other complex solids like oriental glasses [212], and other disordered crystalline materials [3, 16, 47, 180, 187, 188]. In all cases the temperature in the middle of the plateau of the thermal conductivity coincides with $T_{\rm max}$ of the boson peak [96], as displayed in Fig. 7, where thermal-conductivity data are displayed with a temperature scale normalized with $T_{\rm max}$.

As mentioned also in the introduction, there is a correspondence between of the vibrational part of the specific



FIG. 7. Temperature dependence of the thermal conductivity of several materials plotted vs. $T/T_{\rm max}$, where $T_{\rm max}$ is the maximum of the reduced specific heat $C(T)/T^3$: SiO₂ [37, 49], PMMA and PS [177], glassy C₂D₂OD [97], oriental glass (KBr)_{0.75}(CN)_{0.25} [212], disordered ferro-electric perovskite crystals [187, 188], clathrate semiconductors [16, 47, 180] and disordered Y₂O₃ZrO₂ [3]. The dashed line indicated the (boson) peak of $C(T)/T^3$ (from Krivchikov and Jezowski [96]).

heat and the vibrational DOS $g(\omega)$. This relationship is given by [15]

$$C(T) = \frac{\hbar^2 \omega^2}{k_B T^2} \int_0^\infty d\omega g(\omega) \frac{e^{\hbar\omega/k_B T}}{\left[e^{\hbar\omega/k_B T} - 1\right]^2} \,.$$
(7)

Buchenau *et al.* [35] showed for the example of glassy SiO_2 , by comparing with inelastic neutron-scattering data, that the DOS obtaining from the neutrons reproduces the temperature variation of the specific heat using (7).

Inverting Eq. (7) for $g(\omega)$ is a mathematically ill-posed problem, but, using methods explained in Tikhonov and Arsenin [192] (Tikhonov regularization) it can be still achieved, for obtaining the DOS, as shown by Surovtsev [181]. This has been widely used in the meantime [11, 134].

2. Spectroscopic Methods

A. Early investigations

Other than by the temperature dependence of the specific heat, information about the vibrational DOS of a solid can be obtained by several spectroscopic methods: Raman spectroscopy [29, 90], neutron [164], X-ray [169], and nuclear [43] inelastic scattering as well as Terahertz spectroscopy [131, 163, 189]. In the early times vibrational spectroscopy in glasses was focussed on comparing



FIG. 8. a) HH and VH Raman intensity of vitreous silica, b) vibrational density of states $g(\omega)$ of vitreous silica, and c) $g(\omega)$ of polycrystalline silica, both obtained by inelastic neutron scattering, from Galeener *et al.* [71].

the obtained spectra with those of their crystalline counterparts. In Fig. 8 we show an investigation the paradigmal glass silica (SiO₂). The Raman and neutron spectrum of glassy silica is compared with the spectrum of polycrystalline quartz. It is observed that in the regime above ~ 300 cm⁻¹ ~ 10 THz the spectra exhibit essentially the same features, which correspond to the typical vibrational excitations of a covalent network [71].

B. Inelastic Neutron and Raman scattering

For incoherent neutron scattering the scattering intensity is given by the incoherent one-phonon neutron scattering law [164]

$$S_{\rm incoh}(k,\omega) \propto k^2 \left[n(\omega,T) + 1 \right] \frac{g(\omega)}{\omega}$$
. (8)

Here $\hbar \mathbf{k}$ is the momentum transfer and $\hbar \omega$ the energy transfer experienced by the neutrons during the scattering process.



FIG. 9. Density of states of glasses as evaluated by inelastic neutron scattering. Top: Glycerol (from Wuttke *et al.* [206]), Bottom: SiO₂ (from Wischnewski *et al.* [203]). It can be seen that in both cases in the low-temperature region (below ~ 170 K) the boson-peak spectrum is temperature independent. The arrows (top panel) and straight lines (bottom panel) correspond to the Debye $g(\omega) \propto \omega^2$ prediction [15].

The thermal prefactor $n(\omega, T)$ is due to the condition of detailed balance $S(k, -\omega) = S(k, \omega)e^{-\hbar\omega/k_BT}$. In principle, (8) applies only to *incoherent* neutron scattering, which would limit the investigations mainly to hydrogencontaining materials like glycerol [206] or water [111]. However, it was demonstrated, that Eq. (8) may also be used for coherent scatterers, which comprise most nuclei in the investigated materials. It turned out, that expression (8) may be used for coherent spectra, averaged over all experimentally available momenta $\hbar \mathbf{k}$ ("incoherent approximation" [40, 64, 133, 164]).

Let us have a look at the DOS of glassy glycerol and silica, obtained [203, 206] by inelastic neutron scattering, shown in Fig. 9. The DOS is plotted as $g(\omega)/\omega^2$, which should be frequency independent if Debye's law





FIG. 10. Reduced DOS of collective motions in toluene, ethylbenzene dibutylphthalate, and glycerol glasses. Arrows indicate the energy of the boson peak estimated from the data at lowest temperature (from Chumakov *et al.* [42]).

would apply. We see that the strong enhancement with respect to the Debye expectation (boson peak) is temperature independent in the low-temperature regime 50 to 150 K. This is again an indication that in this low-temperature regime the boson peak is likely to be of harmonic origin. The temperature dependence at higher T (or smaller frequency) due to the anharmonic interaction may come about – like in crystalline solids [74, 85, 123] – in two different ways: (i) indirectly via a change of the elastic constants, (viz. the sound velocities) with T (quasi-harmonic effect), or (ii) via a direct change of the excitation spectrum.

Among the many subsequently reported neutronscattering results on the vibrational spectrum of glasses [13, 33–35, 86, 148, 196, 205, 214, 217] many investigations focussed on the extraction of the Raman-coupling function $C(\omega)$ by comparing the neutron spectra with the Raman spectra [2, 27, 56–58, 82, 89, 99, 151, 176, 181– 186, 216]. All of these showed that $C(\omega) \propto \omega$ in the BP frequency regime is linearly proportional to the frequency ω . This was used subsequently to study the DOS by Raman spectroscopy [38, 48, 54, 83, 84, 92, 127, 138, 149, 166].

In 2008 Schmid and Schirmacher [161] published a theory of Raman scattering, which makes it possible to describe Raman spectra and other spectra containing information on the DOS in a unified way. This was used subsequently, in combination with heterogeneous-elasticity theory [152, 155–157] to describe and reconcile Raman, neutron-scattering and specific-heat data in a unified way [28, 134, 167, 194].



FIG. 11. Comparison of the density of states (a), (d) and the reduced density of states (b),(e) obtained with the nuclear inelastic scattering technique, and of the specific heat (c),(f) for ambient silica glass and α -quartz (a)–(c), and for densified silica glass and α -cristobalite (d)–(f) (from Chumakov *et al.* [45]).

C. Nuclear inelastic scattering

A powerful method for measuring the vibrational DOS is nuclear inelastic scattering (NIA) [168, 179]. It had already been pointed out as early as 1960 by Singwi and Sjölander [175] that the resonance fluorescence spectrum of Mössbauer impurities essentially gives the same information as the incoherent neutron scattering law. But in conventional Mössbauer experiments or by applying conventional X-Ray sources the frequency range was not extended enough to observe phonon spectra. This situation changed with the advent of high-brilliance synchrotrotron sources, and it was shown [168, 179] that the NIA method can successfully be used to extract phonon spectra of solids. A severe drawback of this method is that there are only a few Mössbauer isotopes available with resonance energies accessible by synchrotron radiation, namely ⁵⁷Fe ($E_0 = 14.4 \text{ keV}$), ¹¹⁹Sn ($E_0 = 23.9 \text{ keV}$), ¹⁵¹Eu ($E_0 = 21.6 \text{ keV}$). Chumakov *et al.* [42] imbedded ferrocene molecules with 57 Fe as central atom into several glassy hosts in order to successfully extract the vibrational DOS of the hosts. The authors made sure that the ferrocene molecules were tightly bound into the hosts, so that the spectra are not due to an extra motion of the impurity, but represent the host spectra. In Fig. 10 the results for the vibrational DOS of four glass-forming materials are shown [42]. The authors found that all spectra (and further spectra from the literature) exhibited an exponential decrease of the DOS beyond the boson peak. It has been shown recently [193], by applying instanton techniques [39], that this exponential decrease is a disorder-induced phenomenon like the band tails observed in the electron spectra of disordered materials [46].

Later the NIA method was used to compare the spectra of glassy silicates with the DOS, obtained from the phonon dispersions of the corresponding crystals [44, 45, 218]. Following the conclusions of the model calculations of Taraskin et al. [190] the authors substantiated the point of view that the boson peak is a broadened version of the lowest (transverse-acoustic) van-Hove singularity of the corresponding crystal. They explained the fact that in crystals in the low-frequency regime of ~ 1 THz no such singulary occurs by pointing out that glasses usually have a lower density than the corresponding crystals. In the case of SiO_2 they compared the DOS of the ambient SiO₂ glass with that of crystalline α quartz and that of densified SiO_2 glas with that of cristobalite, which has a higher density. In both cases the glassy boson peak is observed near the corresponding van-Hove singularity. The boson peaks in the specific heat of these materials corroborate these findings (see Fig. 11). The authors state the opinion that quite generally in all glassy materials the boson peak would be due to the leveling-off of the transverse acoustic sound dispersion near the quasi-Brillouin zone (quasi-van-Hove singularity), i.e. the BP would in general *not* be caused by the structural disorder. We will comment on this in the discussion section.

3. Inelastic X-Ray scattering

A breakthrough in the vibrational spectroscopy of glasses was the development of inelastic-X-ray spectrometers at synchrotron sources with extremely narrow resolution [169, 170]. Due to the very high brilliance of the synchrotron radiation it was possible to devise monochromators and analyzers (using very-high-order reflections) with resolution of ~ 1 meV, comparable to that of thermal neutron scattering. In contrast to the latter for the X-rays there is no upper kinematic limit in energy. For the neutron investigations it was not possible to investigate acoustical waves above the ~ 1 Thz or 4 meV regime. Similar to neutrons, the X-rays are sensitive to the *longitudinal* degrees of freedom.

Instead of averaging over the momenta, as done in the incoherent approximation, the observation of the \mathbf{k} dependence of the spectra can provide useful information on the nature of the vibrational wavefunctions, which underly the observations. The spectra are given by the *coherent* scattering law

$$S_{\rm coh}(k,\omega) \propto |n(\omega) + 1| \chi''(k,\omega) \tag{9}$$



FIG. 12. Momentum (Q) dependence of the DHO resonance frequency parameter $\Omega(Q)$ and $\Gamma(Q)$, Eqs. (9) and (10), for glassy glycerol, from Sette *et al.* [169].

where $\chi(k,\omega) = \chi'(k,\omega) + i\chi''(k,\omega)$ is the complex dynamical susceptibility, which is usally parametrized as a damped harmonic oscillator (DHO) with resonance frequency $\Omega(k)$ and sound attenuation (damping) coefficient $\Gamma(k)$

$$\chi(k,z) = \frac{k^2}{-\omega^2 + \Omega(k)^2 - i\omega\Gamma(k)}$$
(10)

Inelastic scattering experiments featuring a resonance $\Omega(k)$ are usually referred to as Brillouing spectra and $\Gamma(k)$ is then the Brillouin line width (full width at half maximum, FWHM for $\Gamma \ll \Omega$).

Indeed the first inelastic X-ray scattering (IXS) experiments on glassy glycerol [115, 169], LiCl:6H₂O [115] and SiO₂ [26] revealed a pronounced propagating longitudinal sound excitation, i.e. $\Omega(k) \propto k$ which extends into the $k \sim 5 \text{ nm}^{-1}$ range, which corresponds to a wavelength of ~ 1 nm. Interestingly, in all cases the propagating wave-like excitations extended beyond the boson-peak energy $E_{BP} = \hbar \omega_{BP}$, as indicated in the top panel of Fig. 12 for the case of glycerol [169]. Similar features were found in glassy SiO₂ [26]. Some authors were astonished that above the boson peak and the Ioffe-Regel frequency propagating modes would exist [67, 195], but later experiments confirmed the presence of these modes, which proved to be of longitudinal character.

In subsequent times the resolution of the X-ray beamlines was increased and further details about the vibrational excitations of glasses were revealed. Here, we focus on the example of glassy SiO_2 . In the top panel



FIG. 13. Upper panel: Dispersions of maxima of the dynamical structure factors obtained by inelastic X-ray and neutron scattering experiments ([23] and citations quoted therein). Lower panel, top: Frequency-dependent sound velocity $v_L(\nu) = \Omega_L(q)/q$ vs. $\nu = \Omega(q)/2\pi$, middle: Brillouin line width (sound attenuation) $\Gamma(\nu)$, bottom: reduced DOS $g(\nu)/\nu^2$ obtained by neutron scattering, using the incoherent approximation (from Baldi *et al.* [22]).

of Fig. 13 the dispersion $\Omega(q)$ is shown together with the maxima of inelastic neutron scattering (INS) data obtained by the same authors. The latter show the boson peak near $\hbar\omega = 6$ meV as a wavenumber independent feature. Two years later Baldi et al. [21, 22] found a crossover in the sound attenuation (Brillouin line width) from a Ravleigh-like behaviour $\Gamma(\omega) \propto \omega^4$ to ω^2 . as predicted by heterogeneous-elasticity theory (HET) [113, 152, 155-157] near the boson-peak frequency 1.5 THZ = 6 meV. This crossover is accompanied by a kink in the frequency-dependent longitudinal sound velocity $v(\omega)$, where $v(\omega) = \Omega(q)/q|_{\omega=\Omega(q)}$, predicted as well by HET theory. Similar findings were obtained as well for glassy glycerol [124] and sorbitol [150]. In the latter materials, as remarked by Baldi et al. [21] the connection between $\Gamma(\omega)$ and $v(\omega)$, as given by HET theory is sufficient to explain the boson peak in the INS data, whereas



FIG. 14. Top: Non-affine displacements in a computer SiO₂ glass subject to a global shear; bottom: reduced DOS $g(\nu)/\nu^2$ scaled with the transverse sound velocity and the correlation length ξ for the SiO₂ glass and a Lennard-Jones-glass [103] (from Léonforte *et al.* [104]).

in glassy SiO_2 it is not. The missing boson-peak intensity could then be additionally due to optic-like or van-Hove-singularity like modes [19, 20, 24, 41, 44, 45].

IV. SIMULATIONS

Since the first molecular-dynamics (MD) simulation of a Lennard-Jones glass⁶ of Rahman *et al.* [145], in which the DOS was calculated and was shown to agree to the Fourier transform of the velocity autocorrelation function [81, 157], a very large number of MD simulations has been published, and, again, we can only mention a few, which – to our opinion – contributed most to the understanding the low-frequency anomalies of glasses.

In the 1970-80 years only limited computer power and storage capacity was available, so the simulated systems could have only particle numbers of N = 500 to 2000. In simulations of such small systems Laird and Schober

 $^{^6}$ A Lennard-Jones glass is a liquid, in which particles interact via a Lennard-Jones potential $\phi(r) \propto Ar^{-12} - Br^{-6}$, quenched to a very low temperature.

[98, 165] found low-frequency modes, which, by analyzing the participation ratio appeared to be localized.

The participation ratio p is defined by [50]

$$p = \left[N\sum_{i} (\mathbf{e}_{i} \cdot \mathbf{e}_{i})^{2}\right]^{-1}$$
(11)

of the eigenvectors \mathbf{e}_i of the dynamical matrix (1) normalized as $\sum_{i} (\mathbf{e}_i \cdot \mathbf{e}_i) = 1$. *p* takes values of the order unity for extended states and small values of order 1/Nfor localized ones. In macroscopic large disordered materials, as stated above, the low-frequency vibrational excitations are acoustical waves, because the material on a large scale is homogeneous and isotropic. In the systems of only ~ 1000 particles, the low-frequency acoustic waves do not exist, because their wavelengths would be much larger than the sample size. Therefore Laird and Schober [98] argued that the localized excitations, which they observed at small frequencies, would – in a larger system – hybridize with the acoustical waves (like heavy-mass impurities in crystals [60]), and called these excitations "quasi-localized". Quite recently a revival of the investigation of such small systems took place in the literature [12, 18, 105, 106, 122, 135, 137, 201]. It was found that the DOS of the quasi-localized modes of small systems follows a scaling $q(\omega) \propto \omega^{\alpha}$, where in many investigated systems $\alpha \approx 4$ [105]. In other systems the observed exponent α ranged between 2 and 4, dependent on the preparation protocol [12, 135, 137].

In the 2000er years the advancement of machine power and storage, in particular the advent of parallel computing [139] made it possible to treat systems with 10^5 to 10^7 particles, enabling to disentangle the boson peak from the finite-size resonances of the acoustic waves. One of the first of such studies treated a Lennard-Jones system [103] and glassy SiO₂ [104]. It was shown, confirming other similar findings [102, 204] that the overwhelming part of the vibrational displacements in a glass do not follow an external strain in an affine way. Instead, these displacements display non-affine patterns, as shown in Fig. 14. Léonforte et al. [104] found that the correlation length ξ of these patterns is as large as ~ 10 interatomic spacings both in the Lennard-Jones and SiO₂ glass. In a representation against $\omega \xi / v_T$ – in accord with the empirical law of Elliott [63] and Duval *et al.* [55] – the observed boson peaks co-incide, as displayed in Fig. 14. Further studies of model glasses focussed on evaluating the local spatial fluctuations of elastic constants. As shown in the two seminal papers of Lutsko [109, 110] local elastic constants - including the non-affine contributions - can be evaluated by a coarse-graining procedure. Such a procedure was applied in a number of papers [53, 113, 120, 121]. These studies confirmed the presence of spatial fluctuations of elastic constants in glasses, which is the basic assumption of heterogeneous-elasticity theory, HET [152, 156, 157]. In fact, using the example of a simulated soft-sphere glass, i.e. a quenched system of $N = 10^7$ particles with interparticle potential varying as $\phi(r) \propto r^{-12}$ Marruzzo *et al.* [113] showed



FIG. 15. Comparison of the results of a soft-sphere glass simulation (symbols) for three different temperatures $T = 5 \cdot 10^{-5}$, $T = 5 \cdot 10^{-4}$, $T = 5 \cdot 10^{-3}$ (in Lennard-Jones units), with the prediction of heterogeneous-elasticity theory (HET) [152, 155] (blue lines). Top panel: reduced DOS $g(\omega)/\omega^2$. Bottom panels: Real and imaginary part of the complex shear modulus, obtained from both, the complex transverse and longitudinal sound velocities, see text (from Marruzzo *et al.* [113]).

that the observed boson peak is due to the spatially fluctuating elastic constants and can be perfectly described by HET using exactly the observed statistics. The data were evaluated for the DOS $g(\omega)$ from the velocity autocorrelation function and the longitudinal (L)and transverse (T) current correlation functions [81, 157] $C_{L,T}(k,\omega) = (\omega/k^2) \text{Im} \{\chi_{L,T}(k,\omega)\}$ with the longitudinal and transverse dynamic susceptibility given by the DHO formula (10). The quantities $\Omega_{L,T}(k)$ and $\Gamma_{L,T}$ were converted to complex frequency-dependent elastic constants $G(\omega)$ (shear modulus) and $M(\omega) = K + \frac{4}{3}G(\omega)$ (longitudinal modulus) as

$$G(\omega) = \frac{\rho_m}{k^2} [\Omega_T(k)^2 - i\omega\Gamma_T(k)]_{\omega=\Omega_L(k)}$$

$$M(\omega) = \frac{\rho_m}{k^2} [\Omega_L(k)^2 - i\omega\Gamma_L(k)]_{\omega=\Omega_L(k)}$$

$$= K + \frac{4}{3}G(\omega). \qquad (12)$$

Here ρ_m is the mass density. The transverse and longitudinal data sets could be reconciled by taking the bulk moduls K frequency-independent and real, as demonstrated in the bottom panel of Fig. 15, i.e. it was found that $\Gamma_L(\omega) = \frac{4}{3}\Gamma_T(\omega)$ and, indeed, $M'(\omega) = K + \frac{4}{3}G'(\omega)$. Furthermore, the prediction of heterogeneous-elasticity theory, HET [113, 152, 155] (blue lines) lie on top of the simulated frequency dependent spectral quantities. This holds for three very different temperatures. It is remarkable that the spatial distribution of the fluctuating local shear moduli, extracted from the simulation, agrees with the distribution used in the theory. The temperature dependence at very low frequencies could be traced to anharmonic effects [114]. The downward kink of the real parts of the elastic coefficients at the BP frequency, which correspond to the downward kink of the k dependent sound velocity [21, 124], which is predicted by HET, arises as a consequence of the Kramers-Kronig relation between the real and imaginary parts of the complex moduli. Similar features had also been observed in an earlier simulation of a Lennard-Jones glass [125].

V. DISCUSSION

Let us try to summarize, what has been found in the last half century about the origin of the boson-peak related vibrational anomalies of glasses. Beside that – on the theoretical side – the subject is highly controver-

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sal, we find that – on the experimental (and computerexperimental) side – the evidence is that the boson peak is not universal, i.e. in different materials the spectral features leading to an enhanced reduced DOS in the THz regime are caused by different physical mechanisms. For the prototypical glass SiO_2 there is clear evidence for the presence of local oscillators in the THz regime, which are most probably librations⁷ of SiO_2 tetrahedra [33], giving rise to optical modes and van-Hove peaks in the corresponding crystal [45]. On the other hand, in non-network glasses like metallic glasses or the computer models, it has become clear that the structural disorder is responsible for the boson peak. In such materials the BP marks the transition from sound-like vibrational excitations to random-matrix like excitations. It has been demonstrated by investigating the vibrational spectrum of a macroscopic model glass that both features, the disorderinduced boson peak and a smeared van-Hove singularity coexist at different frequencies [202]. In fact, as remarked by Baldi et al. [21] both mechanisms might be present in glassy SiO_2 . For a simulated metallic glass Brink *et al.* [30] recently demonstrated that the BP clearly comes from the disorder and not from a smeared van-Hove singularity. Particularly difficult in this respect is the interpretation of specific heat data. As stated above, a BP in the specific heat is observed not only in glasses but in in an extended number of (complex) crystalline materials [96]. In the latter, they are most probably due to low-frequency "rattlers", i.e. loosely bound heavy-mass molecules or residues

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