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Some remarks on the low-energy excitations in glasses: Interpretation of Boson peak data

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Abstract

The frequency-reduced representation $g(\omega)/\omega^2$, of the vibrational density of states $g(\omega)$, known as the *Boson peak* is frequently used to extract information on structural and dynamical properties of glasses and supercooled liquids. The $g(\omega)/\omega^2$ representation is preferred over the true excess of the vibrational modes – defined as the difference between the total $g(\omega)$ and the Debye contribution – for practical reasons. Analysis of a large body of the available experimental data from inelastic neutron and Raman scattering reveals that reduction procedures distort to a great extent the otherwise symmetric excess density of states. The frequency and the intensity of the Boson peak are very sensitive to minor modifications of the distribution of the excess. The quantitative use of the $g(\omega)/\omega^2$ function leads to some peculiar results that contradict our general perception about structure and dynamics of glass-forming materials. We stress that the physics of the low-energy excitations are hidden in the excess density of states, which should attract more attention both from the experimental as well as the theoretical point of view.

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Our general perception when considering vibrational properties of a material in its non-crystalline (amorphous or glassy) state at low energies is that these properties differ dramatically from the corresponding ones of the same material in its crystalline state. The differences obviously emerge from the lack of long-range periodicity and are epitomized in the universal presence of excess low-energy excitations in the amorphous state, which are situated in the interval $0.5 - 2$ THz ($1 \text{ THz} \hat{=} 33.3 \text{ cm}^{-1} \hat{=} 4.13 \text{ meV}$) [1]. The term low-energy excitations encompasses a number of properties such as “anomalies” in the specific heat and the thermal conductivity [2] as well as the presence of non-Debye scattering mechanisms in inelastic neutron and Raman scattering. The present paper will focus on the latter, i.e. on the non-Debye scattering mechanism in low energy inelastic scattering spectra, which is widely known as the Boson peak.

Let us first compare the vibrational density of states (VDoS) $g(\omega)$ between a glass and its corresponding crystal. See for example Fig. 2 in [3] and Fig. 2 in [4] for two typical glass-formers, OTP and toluene, respectively. The comparison reveals worth-noticing differences only in the high-energy modes while below ~ 1 THz, where the Boson peak dominates, there is no “visible” difference. In particular, the genuine excess of the modes, which is provided by the difference between the VDoS of the glass and the crystal $g(\omega) - g^{cr}(\omega)$ (or in some approximation by the difference between the VDoS of the glass and the Debye expectation for the glass, $g(\omega) - g^D(\omega)$), is negligible around the Boson peak energy. Naturally, a question arises here: “are these differences between crystalline and non-crystalline phases at low energies really profound, as usually considered, or is this perception a result of the way we analyze the data?” To answer this we have to trace back to the definition of the Boson peak, i.e. the peak in the *reduced* VDoS, $g(\omega)/g^D(\omega)$. Since $g^D(\omega)$ is not usually known with high accuracy $g(\omega)/\omega^2$ is used as an alternative because $g^D(\omega) \propto \omega^2$. It should be emphasized here that although vibrational differences at low energies are usually discussed between glassy and crystalline phases, $g^D(\omega)$ refers to the Debye VDoS of the glass and not to that of the crystal.

The appearance of the Boson peak is therefore simply the result of the reduction of the VDoS. Apart from some important shortcomings that usage of the *frequency-reduced* VDoS (fr-VDoS) entails, as

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2 discussed below, one can also notice some strange effects brought about by such a reduction procedure.
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4 (The terms Boson peak and fr-VDoS will be used interchangeably here with the same meaning). In
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6 particular, Boson peaks in purely crystalline materials have been discovered and discussed; some
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8 examples include shape memory alloys [5], zeolites [6], as well as several other materials, especially
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10 crystalline polymers. Obviously, this adds some confusion to the interpretation of the Boson peak and its
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12 relation to glass or supercooled liquid properties. The fact that peaks in the $g(\omega)/\omega^2$ exist also for
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14 crystalline solids calls for an examination of how the ω^2 term influences the spectral features of the real
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16 excess.
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20 An extended discussion on this issue has already been presented elsewhere [7]. A detailed
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22 comparison between the true *excess* VDoS (e-VDoS) defined as $g(\omega) - g^D(\omega)$ and the fr-VDoS,
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24 $g(\omega)/\omega^2$ revealed the following facts: (i) e-VDoS is a symmetric (bell-shaped) distribution which
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26 transforms to an asymmetric peak upon divided by the ω^2 term. The observed “universal” asymmetry of
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28 the Boson peak is therefore an artifact and hides no special physical significance. (ii) In cases where it was
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30 possible – because of the availability of INS data and Debye frequencies – the temperature dependence of
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32 the e-VDoS was calculated [7]. In all cases, the temperature rise causes a shift of the e-VDoS to lower
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34 energies as well as a decrease of its intensity. The most interesting behavior is that appreciable changes
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36 occur also at $T < T_g$. (iii) The position of the maximum of the Boson peak Ω^{BP} is not directly related to
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38 the maximum of the distribution of the e-VDoS; the former is by a factor of 2 to 3 smaller than the latter.
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40 Further, Ω^{BP} is sensitive to the type of the tails (convex- or concave-like) of the distribution of the excess
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42 modes. It would be therefore risky to use the Boson peak frequency either in estimations of correlation
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44 lengths characterizing the intermediate range structural order of glasses or in correlations with other glass
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46 parameters [8]. This is particularly true since *the Boson peak is a not a real vibrational peak; instead it is*
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48 *an artifact of the product of the Bose factor and the phonon propagator term, which modulates the VDoS.*
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54 Observation (ii) mentioned above calls for a re-examination of a correlation between the fragility m
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56 of a liquid (defined as the slope of $\log(\text{viscosity})$ against T_g/T plot extrapolated at the glass transition
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2 temperature T_g) and the strength of the vibrational Boson peak defined as $g(\omega)/g^D(\omega)$ [9]. Indeed,
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4 figure 1 shows that – with exception of SiO_2 which has a particularly high ratio of excess modes – all
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6 other glasses, for which we could find the relevant data, exhibit an almost constant $g(\omega)/g^D(\omega)$ ratio
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8 with a value at about 2.5 ± 1 . In all cases data refer to the glassy phase. For SiO_2 , glycerol and PB data
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10 are shown for more than one temperature. Interestingly, for SiO_2 we notice a considerable reduction of the
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12 ratio $g(\omega)/g^D(\omega)$ in the case where the excess modes were measured at a temperature much higher than
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14 ambient temperature but still below T_g . It should be emphasized that data shown in figure 1 represent the
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16 excess of the low energy modes as measured by INS. We prefer to avoid plotting together values of the
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18 Boson peak strength given by the heat capacity bump $C_p(T)/C_p^D(T)$ because this could be misleading.
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20 Actually, it has also been demonstrated that there is no correlation between the heat capacity ratio and m
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22 [10]. The scattering of the data on the Boson peak strength in figure 1, owing to the fact that we included
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24 data at various temperatures, provides a flexibility of choosing several linear or non-linear relations
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26 between $g(\omega)/g^D(\omega)$ and m as shown by some arbitrarily placed straight lines.
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33 Paying the appropriate attention to the e-VDoS one can also reexamine several conclusions that
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35 have been reached in the past concerning the dependence of the Boson peak intensity on external stimuli
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37 (temperature, quenching rate, annealing condition, thermal history, pressure, irradiation, glass
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39 composition, etc.) [7]. This dependence has been frequently used to extract information of the glass
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41 structure at an intermediate range order which Boson peak is considered to represent. The key point here
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43 is that the asymmetry and more importantly the intensity (peak area) of the Boson peak is much more
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45 sensitive to a slight shift of the e-VDoS distribution rather than on intensity changes of the latter in the
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47 presence of an external stimulus. Therefore, intensity changes of the Boson peak cannot be used safely to
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49 conclude about corresponding changes of the true excess density of states.
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53 The experimental observations (i) and (ii) mentioned above are to a good extent predicted by recent
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55 theoretical approaches that use model systems to calculate the e-VDoS. Indeed, using the Euclidian
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57 random matrices theory (ERMT) Parisi and coworkers [11] studied the inherent structures (local minima
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59 of the potential energy) where the Boson peak was connected with a phase transition from a minima-
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2 dominated phase (with phonons) at low energy to a saddle-point-dominated phase (without phonons).
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4 According to the analytical result of the ERMT, the spectrum of the excess modes is of a semicircular
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6 shape whose edge energy depends linearly on temperature. Figure 2(a) demonstrates the support of the
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8 experimental data (for glycerol [12], polybutadiene [13] and SiO₂ [14]) to this prediction. As regards the
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10 intensity or the *height* of the Boson peak ERMT predicts $g(\Omega^{BP})/(\Omega^{BP})^2 \propto (T_c - T)^{-1/2}$ where T_c is the
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12 critical point at which the aforementioned phase transition occurs. While for most glasses the height of the
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14 Boson peak increases when heating the glass the opposite occurs for SiO₂ [12]. Therefore, predictions
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16 about the temperature dependence of the Boson peak height, i.e. $g(\Omega^{BP})/(\Omega^{BP})^2$, are not quite useful,
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18 especially in view of the fact that e-VDoS shifts to lower energies (as temperature increases); hence the
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20 increase of the $g(\Omega^{BP})/(\Omega^{BP})^2$ ratio may just originate from the e-VDoS shift and may imply no real
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22 change of the density of the excess modes. Predictions about the temperature dependence of e-VDoS are
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24 desired in view of the experimental data shown in figure 2(b).
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30 Apart from temperature, pressure plays also an important role in our efforts to decipher the
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32 microscopic origin of the Boson peak although the number of such studies has been limited due to
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34 inherent experimental difficulties. It is interesting to note that in all glasses studied experimentally up to
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36 now (see for example [15] and references therein) the general conclusion is that the application of pressure
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38 induces a shift of Ω^{BP} to higher energies and at the same time a considerable suppression of its intensity.
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40 Figure 3 shows a representative example for As₂S₃ glass. Theoretical predictions [16] on the pressure
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42 dependence of Ω^{BP} agree with experimental findings. Considering the pressure effects on the Boson peak
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44 in the spirit of the e-VDoS it was revealed [15] that one should not overlook the pressure dependence of
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46 $g^D(\omega)$ as well as the suppression of the Raman coupling coefficient. Taking this into account it has been
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48 found that experimental data concerning e-VDoS show an increase of the excess modes with pressure
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50 application because $g^D(\omega)$ was found to decrease faster than the total VDoS.
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55 We proceed now to the discussion of the implications of some experimental “peculiarities” arising
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57 from the use of the fr-VDoS.
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(A) *Temperature dependence of the Boson peak strength.* For all glasses studied so far, excluding SiO₂, the ratio $g(\Omega^{BP})/(\Omega^{BP})^2$ is strongly increasing with increasing temperature. Obviously, the rapid increase of the Debye contribution and the shift of the excess modes to lower energies are the dominant factors for this effect. In a naïve interpretation, the enhancement of $g(\Omega^{BP})/(\Omega^{BP})^2$ is accounted for by considering an increase of the excess modes excitations. On the contrary, analyzing data for several glasses it turns out that e-VDoS exhibits a clear decrease with increasing temperature [7]. In the case of SiO₂ interpreting the low energy data in terms of the fr-VDoS one is confronted with an unexpectedly *decreasing* Boson peak intensity with temperature rise [14, 17]. In this case, the anomalous behavior of the Debye contribution of silica with increasing temperature seems to be the decisive factor. The area of the e-VDoS of silica decreases with increasing temperature and at the same time exhibits a shift to lower energies, as shown in figure 2.

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(B) *Contrasting behavior in the temperature-dependence of the Boson peak frequency and sound velocity.* Perhaps, the most commonly accepted idea in Boson peak studies is that Ω^{BP} and the longitudinal/transversal sound velocities $v_{l,t}$ can be used to estimate a structural correlation length, i.e. $R_c \propto v_{l,t} / \Omega^{BP}$. As discussed above, and in detail in Ref. [7], Ω^{BP} is the energy of the maximum of a peak that does not represent the VDoS. Indeed, fr-VDoS represents some kind of histogram of the amplitude of the excess vibrational modes and the area under this peak does not account for the density of the excess modes. Experiments show that the maximum of the true e-VDoS is located at much higher energy. Therefore, the use of Ω^{BP} in estimations of structural correlation length becomes questionable. Support to the above-mentioned facts is also provided by the following example. In fact, for all glasses studied so far the temperature dependences of Ω^{BP} and $v_{l,t}$ exhibit slopes of the same sign (positive for silica [11]; negative for all other glasses). A peculiar situation, however, occurs for a modified silica glass where Ω^{BP} and $v_{l,t}$ exhibit different signs in their slopes with temperature; the former is negative [18] (decrease of Ω^{BP} against T) while the latter is positive [19] (increase of both $v_{l,t}$ against T). As a result, R_c values estimated for K₂Si₄O₉ show an unexpectedly dramatic increase with increasing temperature,

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2 even for $T > T_m$. Interpreted in the sense that R_c stands for a structural correlation length the above
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4 finding imply an increasing length scale of the solid-like correlations.
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7 (C) *Non-coincidence of the polarized and depolarized Ω^{BP} energies.* Studies of the polarized (VV)
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9 and depolarized (VH) low frequency Raman spectra of some oxide glasses [20] have revealed that the
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11 energies of the Boson peak in these two polarization geometries are different. Actually, this non-
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13 coincidence effect occurs between the combination $VV - (4/3) * VH$, known as the isotropic part and the
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15 VH spectrum or anisotropic part. The difference $\Omega_{ANISO}^{BP} - \Omega_{ISO}^{BP}$ can be as large as $10-15 \text{ cm}^{-1}$. To account
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17 for this effect, the authors [20] invoked arguments from the theory of molecular liquids where changes of
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19 the intermolecular potentials under pressure are used to describe non-coincidence effects for
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21 intramolecular vibrational modes. The same non-coincidence effects were also observed for the Boson
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23 peak of a silicate glass where its temperature dependence was also recorded [18]. We believe that the non-
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25 coincidence effect can be rationalized without considering that the freezing of the liquid structure
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27 preserves this effect to the glass [20]. Actually for the silicate glass [18] the non-coincidence effect
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29 diminishes in the melt. In a simpler and more reasonable explanation we have to recall that in Raman
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31 scattering the e-VDoS is modulated by the coupling coefficient $C(\omega)$, which is the only parameter that
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33 carries polarization information. Obviously, even minor differences between the polarized and depolarized
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35 components of $C(\omega)$ will modulate e-VDoS accordingly and the division by the ω^2 term will reveal this
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37 change as a different energy of the Boson peak in the two polarization geometries.
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43 (D) *Resolvability of the Boson peak in the normal liquid state at $T > T_m$.* The fact that for several
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45 glass-forming materials the Boson peak is well resolved, i.e. the minimum at its low energy side is
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47 preserved, even when the supercooled liquid is driven to the equilibrium liquid state [11] is a fundamental
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49 issue whose clarification could unravel some mysteries of the Boson peak. It seems that the Boson peak is
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51 not a genuine feature of glassy and supercooled (viscous) phase since it can also exist in the normal liquid
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53 state, that is, above the melting point of the corresponding crystal. This observation casts serious doubts
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55 on the consideration of a vividly increasing quasi-elastic line with temperature rise and on the medium-
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57 range structural order origin of the Boson peak. SiO_2 , ZnCl_2 , ZnBr_2 and their mixtures, $\text{K}_2\text{Si}_4\text{O}_9$ and B_2O_3
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(see Ref. 14 for a detailed discussion) are representative examples of materials for which the Boson peak is well resolved in the melt. In several other cases – glycerol, $\text{Na}_{0.5}\text{Li}_{0.5}\text{PO}_3$, As_2O_3 , Sb_2O_3 , etc. – the Boson peak is not well resolved but there is clear hint that it still survives in the melt.

Bearing in mind that those glasses for which the Boson peak survives in the melt are network-forming and considering the predictions of the ERMT [11] the resolvability of the Boson peak in the liquid state can be reasonably rationalized. In the spirit of ERMT the preservation of the Boson peak might be related to the details of the potential energy landscape. Indeed, the shift of the spectrum of the excess modes to negative frequencies could be due to structure softening taking place above some critical temperature T_c . The relevant changes of the topology of the potential energy landscape involve a merging of the inherent structures (local minima) with saddle points. At the phonon→saddle transition [11] – occurring when crossing T_c (moving upwards) – the e-VDoS crosses the zero energy point $\omega = 0$. As a result, the finite (non-zero) density of states at $\omega = 0$ exhibits a remarkable increase in the fr-VDoS, i.e. $g(\omega)/\omega^2$ representation, which blurs the sharpness of the peak initially seen at $T < T_c$. Given the above context, the Boson peak, i.e. the peak in $g(\omega)/\omega^2$, will continue to show-up as a sharp or well-resolved peak as long as the lowest energy eigenvalue of the e-VDoS has not yet moved to the negative part of the spectrum. Most of the liquids mentioned above are over- or optimally-constrained structures and hence can withstand temperature changes more effectively than floppy structures. Finally, the preservation of the Boson peak in the molten state might at first seem unexpected owing to our general perception that is related to the medium range structural order or to some correlated motion of several tens of atoms. This idea attracted much attention in early studies of low energy excitations in glasses and still continues to be considered as a possible origin of the Boson peak. On the other hand, bearing in mind that the Boson peak is a manifestation of the excess modes in relation to the Debye level it would be not surprising that it can also exist in the liquid state.

Finally, we would like to briefly comment on the existence of “Boson peaks” in materials that possess topological order. Here we can discriminate between two cases: (a) materials called orientational glasses (or orientationally disordered crystals) where molecules have fixed but different orientations, such

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2 as ethanol [21]; (b) materials that accommodate guest molecules such as ice clathrates [22], mixed
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4 fullerene crystals [23], and other inclusion compounds [24]. In both cases, excess (over the Debye level)
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6 modes exist at low energies. Obviously, any contribution in that spectral region of the VDoS will
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8 transform to a “Boson peak” when divided by the ω^2 term. In orientationally disordered crystals the
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10 microscopic origin of the “Boson peak” has been frequently assigned to librational motions of the
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12 molecules. In order to classify such peaks as Boson peaks we have to recall the definition of the Boson
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14 peak, namely the fact that its temperature dependence should scale with the Bose factor. For the case of
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16 guest molecules in crystals [22, 24] the deviations from the Debye expectation – frequently shown as a
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18 peak in C_p/T^3 function – have been successfully accounted for by considering modes (rattling of guest
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20 molecules) as Einstein-type local oscillators. In this second case it is more obvious that these excess
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22 modes have no connection with the Boson peak observed in structural glasses. Therefore, the use of the
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24 peak in the C_p/T^3 curve may cause considerable misunderstanding in view of the fact that in many cases
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26 [25] the “Boson peak” for a material in its crystalline state is found to be stronger than the Boson peak of
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28 the corresponding glass.

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34 The Boson peak continues to be a thorny issue in the physics of glasses. Its dependence on external
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36 stimuli is frequently used to unravel details about structural and dynamical features of the glass. In this
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38 paper, we attempted a short survey on some problems associated with the use of the Boson peak to extract
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40 quantitative information or to propose “universal” correlations. A critical comparison of the true excess
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42 with the frequency-reduced density of vibrational states [7] has led to the conclusion that the physics of
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44 low energy excitation in non-crystalline phases is hidden in the excess VDoS. It would be useful if future
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46 experimental and theoretical work would focus more on these issues so as to enlighten the microscopic
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48 origin of the modes responsible for the Boson peak. The examples given above, concerning the existence
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50 of “Boson peaks” in crystalline phases, reveal that correlations between the spectral features of the Boson
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52 peak and dynamical properties of the glassy or the supercooled liquid phases should be considered with
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54 great care.
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Figure Captions

Fig. 1: Plot of the Boson peak strength derived from INS against the quantity m . Solid triangles correspond to data used for the correlation of Ref. [9], semi-filled circles correspond to new data analyzed in the present work. Temperature legends besides symbols denote the temperature at which the Boson peak strength was estimated for the corresponding glass. Data for GeO_2 are from E. Fabiani *et al.* arXiv:cond-matter/0502207.

Fig. 2: (a) Temperature dependence of the eigenvalue of the maximum of the e-VDoS for glycerol and polybutadiene and silica (inset). The lines represent linear fits according to the ERMT. (b) Temperature dependence of the area of the e-VDoS for the same glasses.

Fig 3: Low-frequency Raman spectra of As_2S_3 as a function of pressure, from Ref. [15]. The arrow denotes qualitatively the energy of the Boson peak.

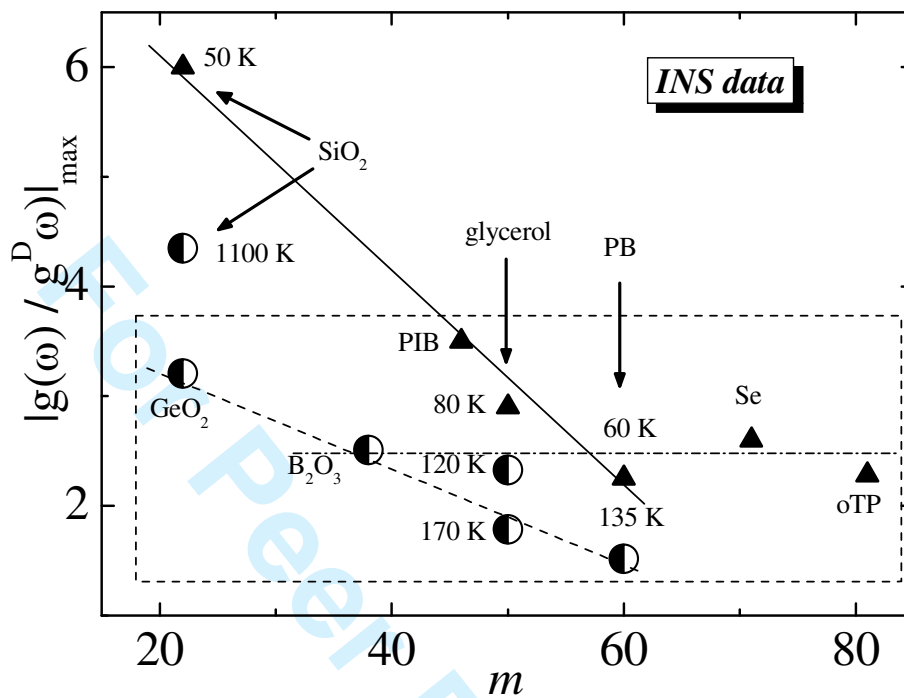


Figure 1

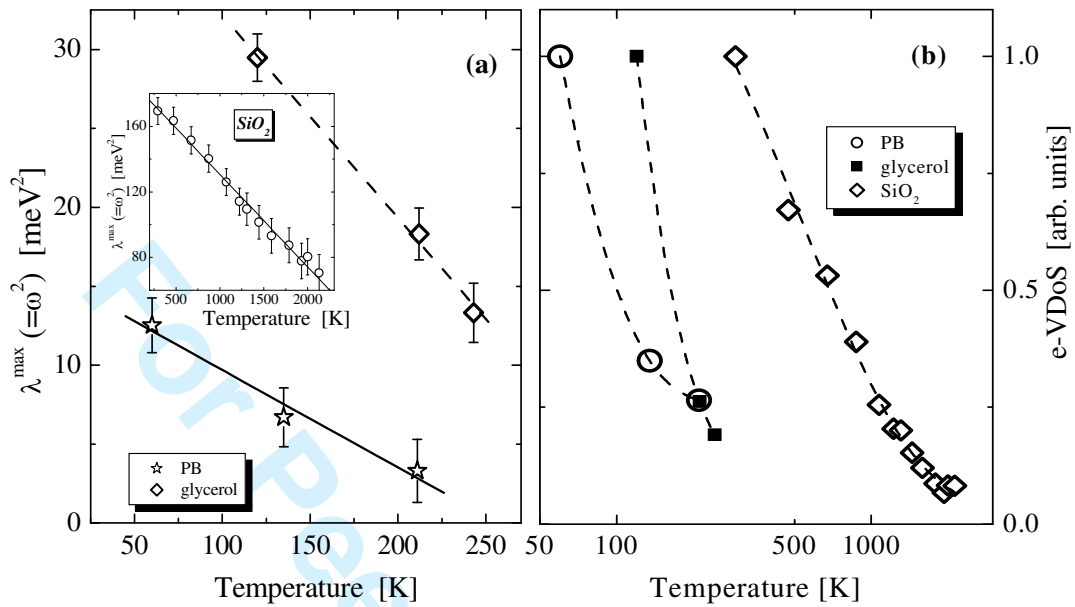


Figure 2

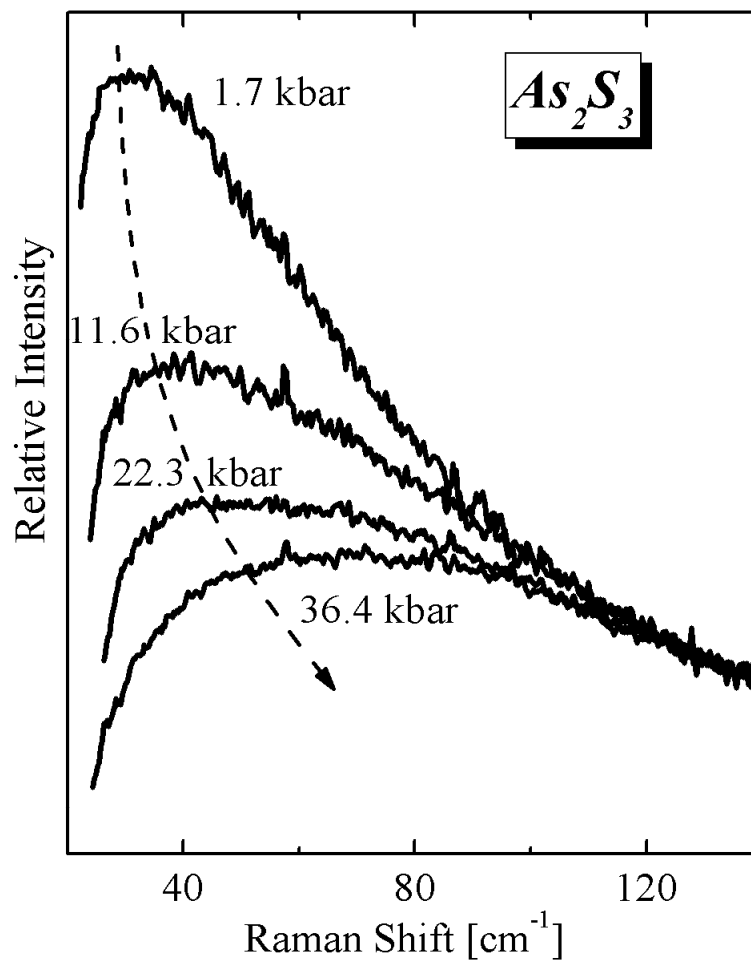


Figure 3