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**Temperature evolution of the boson peak and Debye scaling
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RESEARCH ARTICLE

Temperature evolution of the boson peak and Debye scaling in
vitreous B_2O_3 M. Zanatta^{a*}, C. Armellini^b, A. Fontana^{a,c}, and F. Rossi^a^a*Dipartimento di Fisica, Università di Trento, I-38050 Povo, Trento, Italy;* ^b*CNR-IFN, Istituto di Fotonica e Nanotecnologie, CSMFO Group, 38050 Povo, Trento, Italy;*^c*CNR-IPCF, UOS Roma, c/o Università di Roma "La Sapienza", I-00185 Roma, Italy.*

(December 10, 2010)

We present a Raman scattering study of the B_2O_3 as a function of temperature. Measurements have been done from liquid helium up to the supercooled liquid state. We found that the BP shift could be completely ascribed to the temperature elastic medium transformation and we do not need the introduction of other mechanisms. Furthermore we show also in this system, that the boson peak spectral shape is invariant.

Keywords: glasses, boson peak, elastic medium transformation, Raman light scattering

1. Introduction

A common feature of glasses and supercooled liquids is the presence in the vibrational density of states $g(\omega)$, of an excess of modes over the Debye level. In Raman and neutron inelastic scattering spectra it appears as a broad maximum in the so called reduced density of states $g(\omega)/\omega^2$, and it is known as boson peak (BP), [1–7]. Many different approaches have been proposed to explain the origin and the nature of these extra modes, but this topics are still objects of debate, [8–18].

In recent years many experimental efforts have been devoted to understand the evolution of the BP as a function of thermodynamical parameters like temperature [23, 24] and pressure [25]. Moreover, the BP has been also studied as a function of density [26–28] and time during a chemical vitrification process [29]. A general result is that, when the density is increased, no matter how, the BP position ω_{BP} shifts towards higher frequencies and it decreases its intensity. Moreover, plotting spectra corresponding to different densities as a function of ω/ω_{BP} , they collapse on a master curve [23, 27]. This shows that the BP spectral shape is invariant: any intensity variation is only apparent and it is due to the BP shift. The explanation of the origin of this shift is now the open problem. Assuming an acoustic nature for the BP mode [8, 9, 14], the shift can be explained by considering the elastic medium evolution i.e. the variation of the Debye frequency ω_{DB} [23–27, 29]. This approach has been applied to many different systems with contrasting results. As a matter of fact it provides a good scaling only in some cases [24, 26, 29] so we

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1 can conclude that elastic medium transformation does not always cover the whole
2 story.

3 In this paper we present a Raman scattering study of the boson peak temper-
4 ature evolution in vitreous boron oxide $v\text{-B}_2\text{O}_3$, from the liquid helium up to the
5 supercooled region. Boron oxide is the object of many detailed studies (e.g. refer-
6 ences [19–22]), in what follows we point out our attention on two specific points:
7 the existence of the already mentioned master curve and the comparison between
8 BP shift and elastic medium modification.
9

10 2. Experimental details

11 Vitreous B_2O_3 is a strong glass with $m \simeq 32$ and a rather low glass transition
12 temperature, $T_g \simeq 576$ K [30].

13 The sample was prepared starting from Aldrich boron oxide powder, purity
14 greater than 99.995%. The powder was dehydrated at 100°C and then melted
15 at 1100°C in an alumina crucible. The homogeneous and bubble free melt was sub-
16 sequently quenched in air. The obtained samples were polished in order to obtain
17 good optical surface.

18 Raman scattering experiments were performed using a Jobin-Yvon U1000 double
19 monochromator in 90° geometry. As a light source it was used the 514.5 nm line of
20 an argon ion laser. The spectra were acquired in a wide range of frequencies, from
21 -300 cm^{-1} to 1300 cm^{-1} with different resolutions. In the low frequencies region the
22 resolution is about 2 cm^{-1} . The measurements were performed in both VV and HV
23 polarization geometries, where V and H indicate the polarizations perpendicular
24 and parallel to the scattering plane.
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33 3. Results and discussion

34 The first order Raman scattering intensity for a Stokes process can be written as
35 [31]:
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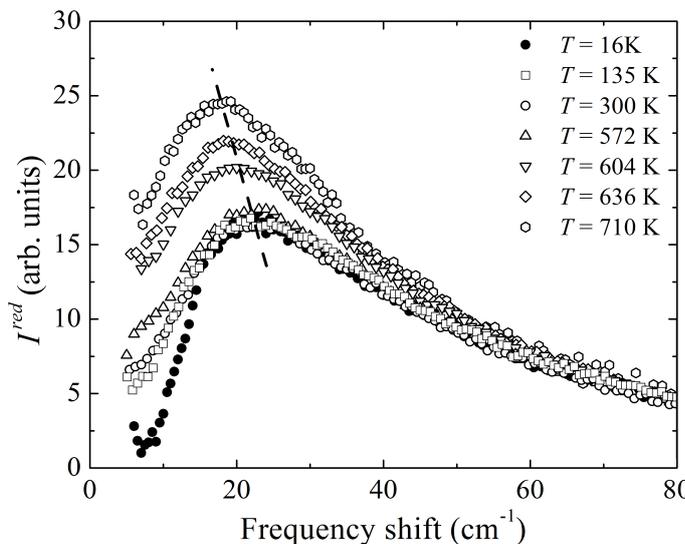
$$37 I_R(\omega) = C(\omega)g(\omega)\frac{[n(\omega, T) + 1]}{\omega}; \quad (1)$$

38 where $n(\omega, T)$ is the Bose-Einstein population factor and $C(\omega)$ is the light-
39 vibrations coupling function. Dividing the Raman intensity for $[n(\omega, T) + 1]$ and ω
40 we obtain the so called reduced Raman intensity $I^{red}(\omega)$, which is proportional to
41 the reduced density of vibrational states $g(\omega)/\omega^2$, namely
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$$48 I^{red}(\omega) = C(\omega)\frac{g(\omega)}{\omega^2}; \quad (2)$$

49 Reduced Raman depolarized (HV) spectra are reported in figure 1 for selected
50 temperatures. Spectra at different temperatures are normalized to the same total
51 area in order to be compared. High temperature spectra are characterized by the
52 rise of a quasielastic scattering signal (QES) which slightly modifies also the BP
53 parameters. The QES contribution has then been subtracted using the procedure
54 described in ref. [23]. It can be seen that, when the temperature is increased, hence
55 the density decreases, the BP peak shifts towards lower frequencies and increases
56 its intensity. In the temperature range investigated, the position shift is $\sim 20\%$
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1 while the intensity shift is $\sim 50\%$. To determine the peak frequency ω_{BP} we fit
 2 each spectrum using a log-normal function.
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 25 Figure 1. Reduced depolarized Raman intensity for the $v\text{-B}_2\text{O}_3$, symbols are as in the caption; the
 26 dashed line is a guide for the eyes and it represents the temperature evolution of the BP.

27
 28
 29 We start our analysis of present data by considering the BP spectral shape by
 30 using a rescaled frequency $\nu = \omega/\omega_s$ and performing the variable transformation
 31 $g(\nu)d\nu = g(\omega)d\omega$. Assuming that, in the BP frequency range $C(\omega) \sim \omega$, [32, 33],
 32 the squeezed intensity can be written as:
 33

$$I_s(\nu) = I^{red}(\omega) \times \omega_s^2. \quad (3)$$

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 38 The so squeezed spectra are reported in figure 2; within the experimental uncer-
 39 tainty they collapse on a single master curve without any adjustable parameter.
 40 The frequency ω_s is defined as the frequency which allows this scaling for each
 41 spectrum. In this case $\omega_s = \omega_{BP}$, as it is clearly visible in the inset of figure 2. The
 42 small differences, smaller than the uncertainty, could be ascribed to the difficulty
 43 in obtaining Raman spectra in absolute intensities.
 44

45 The existence of this master curve shows that the BP evolution is ruled by its
 46 shift. To understand the origin of this feature, we compare ω_{BP} to the elastic
 47 properties of the systems, which are described through the Debye frequency ω_{DB} .
 48 The Debye frequency has been computed using sound velocities from ref. [34],
 49 measured by Brillouin light scattering, and density from [35]. The T evolution of
 50 the sound velocities is reported in the inset of figure 3 where a transverse sound
 51 wave is still visible also in the supercooled and liquid region, up to ~ 1300 K.
 52 This allows the use of the Debye frequency formula also above the glass transition
 53 temperature. In the explored T range the density decreases of about 18%. The
 54 comparison between ω_{BP} and ω_{DB} is reported in figure 3: it can be seen that
 55 they exhibit the same T behavior. The boson peak position scales with the Debye
 56 frequency ω_{BP} , i.e. the transformation visible in the reduced density of states
 57 $g(\omega)/\omega^2$ can be ascribed to the elastic medium modifications.
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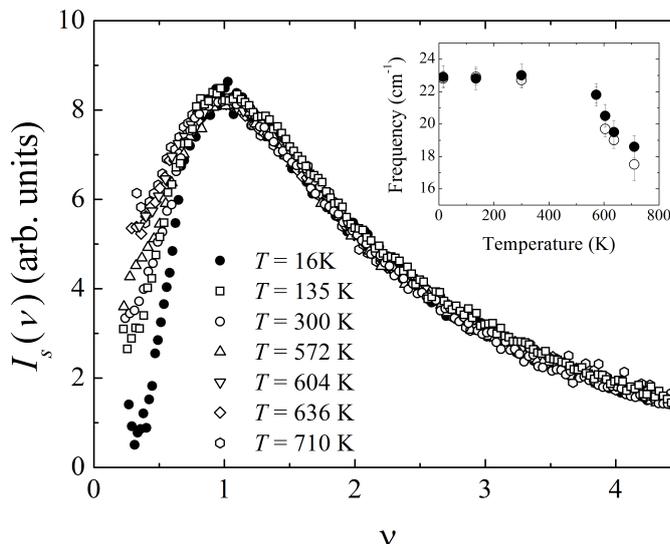


Figure 2. Master curve obtained as described in the text, symbols are as in the caption; in the inset is reported a comparison between ω_{BP} and ω_s , respectively indicated with dots and open circles.

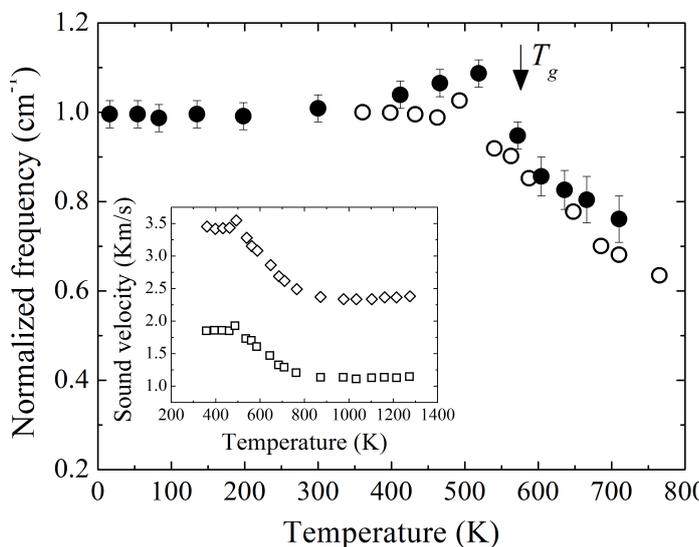


Figure 3. Comparison between ω_{BP} (dots) and ω_{DB} (open circles); longitudinal (open diamonds) and transverse (open squares) sound velocity are reported in the inset [34].

4. Conclusions

We investigated the temperature evolution of the boson peak in vitreous B_2O_3 . We find a master curve for the BP, showing the invariance of the shape. Moreover we find also that the BP shift in this glass could be accounted using the elastic medium transformation approach. Similar studies performed in other strong systems, like vitreous silica [23, 28] and vitreous germania [27], pointed out the absence of such a scaling property, thus invoking the existence of further mechanisms that rule the BP shift. This completely different result can add a piece to understand the nature of these processes.

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