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RESEARCH ARTICLE

Influence of temperature on quasi-elastic scattering in GeO₂ glass

S. Caponi^{a,b,*}, A. Fontana^{b,c}, F. Rossi^b, and M. Zanatta^b

^a*IOM-CNR, c/o Università di Perugia, I-06123, Perugia, Italy;*

^b*Dipartimento di Fisica, Università di Trento, I-38050 Povo, Trento, Italy;*

^c*IPCF-CNR, UOS Roma, c/o Università di Roma "La Sapienza", I-00185 Roma, Italy;*

(October 6, 2010)

We report Raman scattering spectra for the network glass former vitreous germania (v-GeO₂) measured in the 10 K–1200 K temperature range. The results show an interesting temperature dependence of the quasi-elastic scattering. Comparing this study with acoustical attenuation measured by ultrasonic absorption and Brillouin light scattering, we found that the mechanism giving rise to quasi-elastic scattering in this system seems to be the same as the one causing the acoustic attenuation.

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*Corresponding author. E-mail: caponi@science.unitn.it

data in the framework of Jäckle model [22]. Our results suggest, indeed, that the acoustic attenuation involves the same physical mechanisms responsible for QES as discussed in this paper.

2. Experimental and Results

Our sample was prepared from GeO₂ powder (Aldrich, 99.99%) melted in platinum crucibles for about 1h at 1600°C. The homogeneous and bubble free melt was subsequently quenched in air. The absence of crystallization was checked before the experiment by X-ray diffraction measurements. The samples were then polished in order to obtain good optical surfaces.

Raman scattering experiments, in a 90° scattering geometry, were done using a standard experimental setup Jobin-Yvon U1000 in a wide frequency range, from -300 to 3000 cm⁻¹, in order to correctly subtract the underlying weak background of luminescence [25]. The light polarizations were perpendicular, VV and orthogonal, HV, to the scattering plane.

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In order to compare spectra taken at different temperatures, they are normalized at the high frequency peaks, as shown in fig. 1c). The spectra show the BP, peaked at ~ 50 cm⁻¹, and at lower frequency the QES. The lower panel, figure 1b) shows that, increasing temperature up to room temperature, there is no intensity variation of the BP. On the contrary, a strong increase of the scattering is observed at the

lower frequency. Above room temperature the scenario is different, fig. 1a). Here, increasing temperature, the QES present an increasing less marked than before, while the BP shifts to lower frequency increasing its intensity. The boson peak temperature behaviour is not the object of the present work and we do not discuss it further, this study can be found in ref. [26].

We can model the Raman intensity in HV polarization as the sum of QES and BP contributes [17]:

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where:

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where $\tau(V)$ is the relaxation time for potential barriers of height V , and $P(V)$ is the distribution of barrier heights. Different choices of the distribution of barriers have been proposed in literature [22–24, 27]. Concerning the model dependence of the analysis, it should be noticed that the frequency dependence it is found to be almost unaffected by the choice of the distribution function as already reported by Gilroy and Phillips [23] in the case of silica, while slight differences have been evidenced in the temperature evolution. We choose a Gaussian distribution, $P(V)$, of barriers V having a variance σ_V . This distribution has been already used in the case of vitreous germania to optimize the acoustic attenuation data in a wide frequency and temperature range [24, 29].

The results are shown in fig.3. Here we report the acoustic attenuation measured in the ultrasonic region (20 MHz) [30], the one measured by Brillouin light scattering (25 GHz) [28] and the rescaled QES data at 180 GHz. The continuous curves are computed with parameters: $\sigma_V = 1750\text{ K}$, $\tau_0 = 1.47 \cdot 10^{-14}\text{ s}$ and $A = 0.18$. The parameter are the same reported in ref. [29] where the analysis of the acoustic

attenuation has been analyzed over more than three decades in frequency.

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To report the QES intensity in fig 3, we normalized the data on the extrapolation at 180 GHz. The good agreement between the QES and the acoustic attenuation extrapolated between 10 K e 800 K at the same frequency indicates the common origin of the two phenomena. At temperatures higher than 800 K the parallel ends and the QES increases while the extrapolation of acoustical attenuation at higher temperatures decreases. This difference could be due to the occurrence at these temperatures (higher than T_g) of the structural modifications from the glassy state to the liquid one, which contributes to QES with another relaxational process.

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