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RESEARCH ARTICLE

Influence of temperature on quasi-elastic scattering in GeO_2 glass

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(*October 6, 2010*)

We report Raman scattering spectra for the network glass former vitreous germania (v-GeO_2) 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.

Keywords: glasses, vitreous germania, quasi elastic scattering, Raman light scattering

1. Introduction

During the past 10 years, many works have been devoted to study the structural and dynamical properties of complex materials [1–5]. One of the main problems is to understand the modification of the vibrational dynamics due to topological disorder of the system [6–11]. The fast dynamics in disordered systems includes two main contributions: the first is the low-frequency collective vibrations, the so-called boson peak (BP), and the second the relaxation dynamics that appears in light and neutron scattering spectra as quasielastic intensity (QES). Both these contributions are observed in spectra of all glass-forming systems as well as porous systems, polymers, and biological systems [13–21]. Hence, they are common features of dynamics in complex systems. Long times ago [22], the QES was assigned to the light coupling with relaxations of atoms or group of atoms between almost equivalent equilibrium positions described by a double well potential. This coupling can be direct or indirect; recently Sokolov et al.[19] have reported convincing results on v-SiO_2 and polystyrene in favor of the indirect mechanism. Substantially, according to this hypothesis, the contribution to the light scattering comes from the mechanical coupling of defects to the sound waves. Different studies were preformed to relate the QES temperature and frequency behaviors with the damping of acoustic modes [16–19, 23]. In this paper, we report a detailed investigation of the low frequency Raman spectrum (< 200 cm^{-1}) of vitreous and melted germania in a very large temperature range, from 10 K up to 1000 K, extending the temperature range investigated till now [21]. We compare our data with acoustic attenuation

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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 GeO_2 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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Figure 1. pannel a) and b) Raman spectra at chosen temperatures as shown in the label. c) high frequency part of spectra at some significant temperatures.

Figure 1 shows the depolarized reduced intensity measured on v-GeO_2, defined as:

$$I_R^{Red}(\omega) = \frac{I^{Exp}(\omega)}{[n(\omega,T)+1]\omega}; \quad (1)$$

where $I^{exp}(\omega)$ is the experimental datum and $n(\omega,T)$ is the Bose-Einstein population factor.

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]:

$$I_R^{Red} = I_{QES}(\omega) + I_{BP}(\omega); \tag{2}$$

where:

$$I_{BP}(\omega) = C^v(\omega)g(\omega)\frac{[n(\omega,T)+1]}{\omega}, \tag{3}$$

and

$$I_{QES}(\omega) = C^{rel}(\omega)L(\omega,T), \tag{4}$$

are the QES and BP contribution to the spectrum. Here v indicates the vibrational part, and rel the relaxation part; $C(\omega)$ is the coupling light-vibration function and $g(\omega)$ is the vibrational density of states. The QES contribution is described by the function $L(\omega,T)$.

In the present investigation of the low frequency Raman scattering, we are able to measure the spectrum down to frequency shifts of 4 cm^{-1}. The increase below ~ 4 cm^{-1} is caused by an elastic scattering due to a finite resolution of the spectrometer (~ 2 cm^{-1}). From the Raman spectra we have extracted the scattered intensity as function of temperature at 6 cm^{-1} integrating the signal in a frequency window of 1 cm^{-1}. The temperature dependence $I_{QES}(6\text{cm}^{-1})$ is shown in fig. 2 as dots: we can note a rapid increase up to $T = 500$ K, then a plateau which extend up to 800 K and, finally, a strong increase at higher temperatures. It's worth to notice that, at the chosen frequency, the evolution of the QES is not influenced by the shift and the intensity variation of the BP. This is evidenced in figure 2 where is also reported the evolution of the QES obtained from the squeezed spectra (triangles). In fact, dividing the x axis of the spectra by the frequency position of the Boson Peak maximum and scaling the Raman intensity imposing as well the conservation of the total number of vibrational states, a BP master curve has been obtained in a wide class of materials [13–15, 17]. This procedure, called *squeezing*, is well described in the case of GeO_2 in ref. [26], where the squeezed spectra are presented at chosen temperatures from the liquid to the glassy phase. In that representation, the evolution of the QES is completely disentangled by the eventual intensity modification due to the BP variations. As reported in fig. 2, the comparison of the QE signals obtained in different ways presents slightly differences in the high temperature range, but the overview is not modified. In this work we try to understand the microscopic origin of QES analyzing its temperature evolution.

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As already suggested in literature [22] the QES can be due to the scattering from structural defects or damped sound waves which are characteristic of glassy state. In fact the thermally activated relaxation process (TARP) can couple to light

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Figure 2. Intensity of Quasi Elastic Scattering measured from the Raman spectra (dots) at 6 cm^{-1} as function of temperature; and intensity of Quasi Elastic Scattering at some chosen temperatures obtained from the squeezed spectra [26] in order to single out eventual contribution of the Boson Peak intensity variation (triangles).

directly, as was originally proposed by Jäckle [22] or indirectly, by coupling to phonons [19]. In order to compare our data with those obtained by the acoustic attenuation, we consider the internal friction, Q^{-1}, which is related to the QES scattering by $Q^{-1} \propto L(\omega)\omega$ [17]. In Eq. 4, the QES contribution, described by the function $L(\omega, T)$, can be formulated as [22]

$$L(\omega, T) \propto \int dV P(V) \frac{\tau(V)}{1 + \omega^2 \tau^2(V)} \tag{5}$$

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: $\sigma_V = 1750$ K, $\tau_0 = 1.47 \cdot 10^{-14}$ s and $A = 0.18$. The parameter are the same reported in ref. [29] where the analysis of the acoustic

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Figure 3. experimental data for Q^{-1} measured at 20 MHz (circles) [30], 25 GHz (stars). Lines are the evaluation of the TARP model at the same frequencies and the extrapolation at 6 cm^{-1}. Dots are the rescaled I_{QES}.

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.

4. Conclusions

In conclusion, the similar spectral shape and temperature dependence of Raman data, in comparison with the temperature dependence of the acoustical attenuation, support the idea that QES and sound attenuation are due to the same mechanism. Following the interpretation of ultrasonic absorption this mechanism is described by a hopping model for structural defects with different atomic configurations of the structure.

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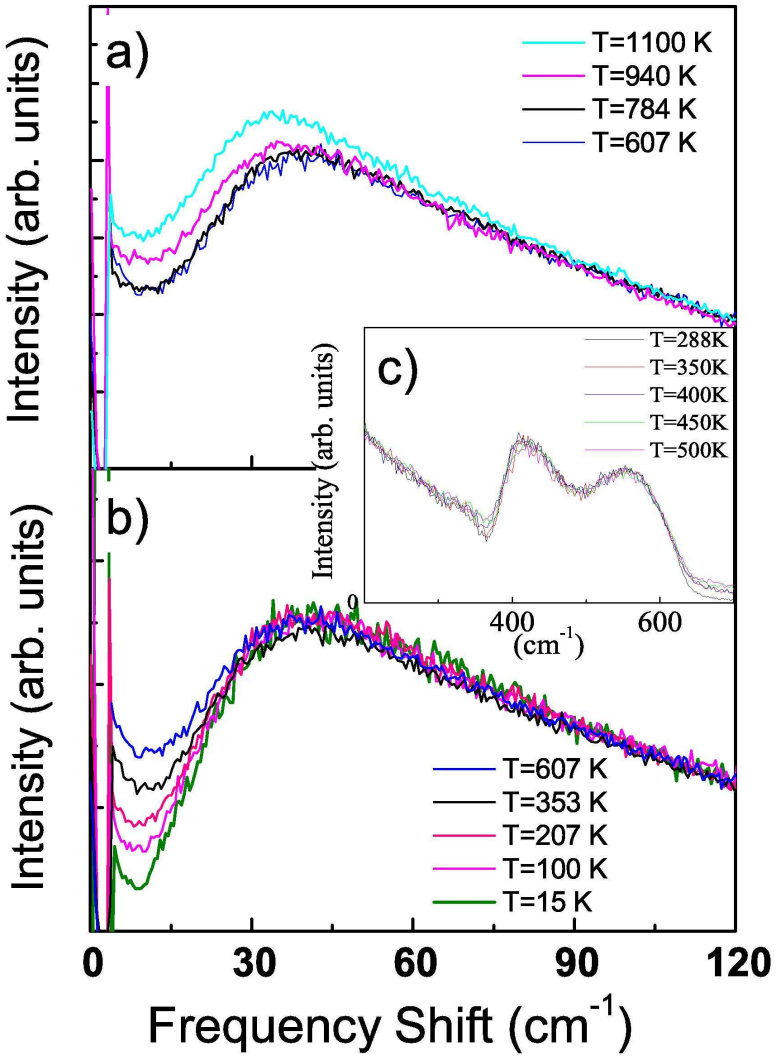

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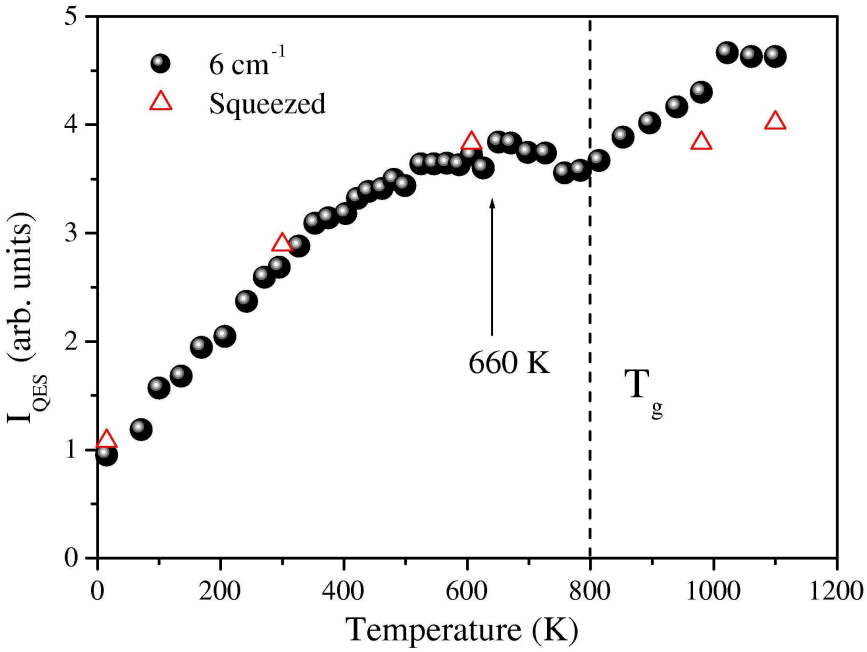

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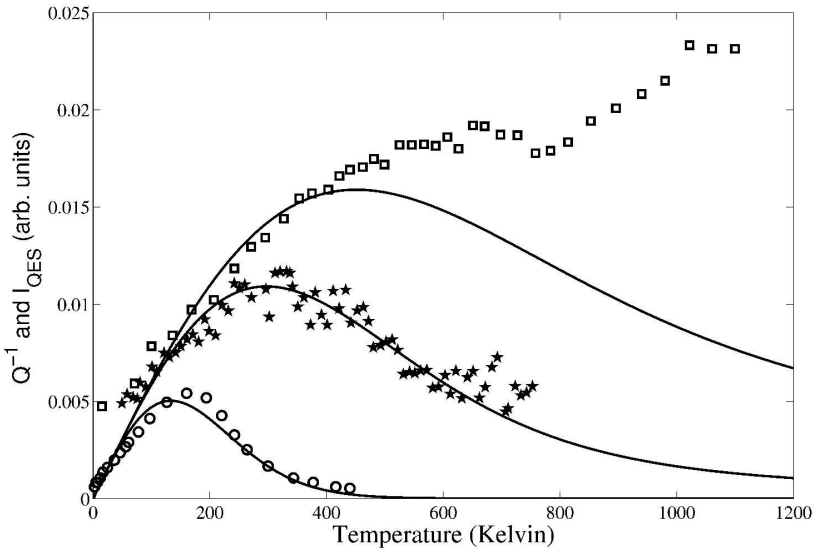

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RESEARCH ARTICLE

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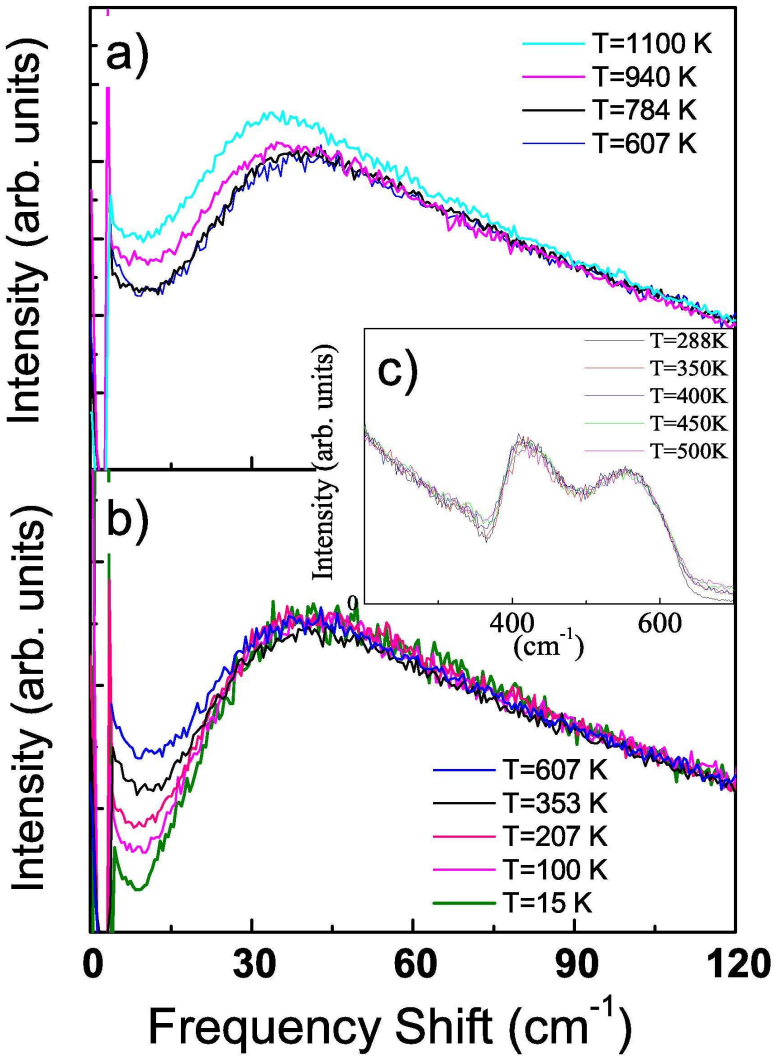

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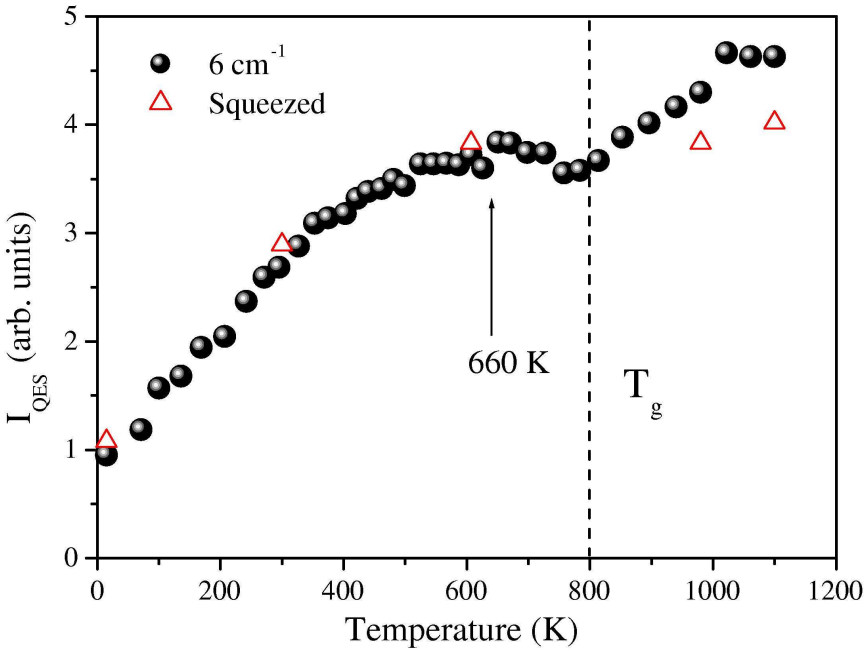

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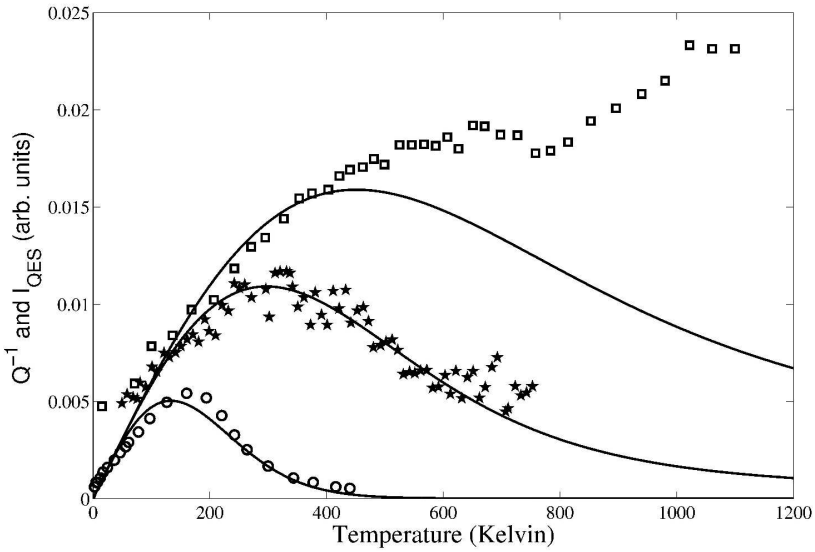

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