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## A link between quasielastic scattering and sound attenuation in silver phosphate glasses

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### Abstract

New inelastic Raman scattering experiments on the superionic glasses  $(\text{AgI})_x(\text{AgPO}_3)_{(1-x)}$  were performed from room temperature up to the melting temperature. The results are compared with the acoustic attenuation measured in the frequency range between 8 and 9 GHz by Brillouin light scattering. This comparison shows that the quasi-elastic scattering intensity can be associated with the same mechanisms that cause the sound attenuation, thereby demonstrating the same physical origin of these phenomena in good agreement with suggested models.

*Keywords:* glasses, Raman spectroscopy, phosphates

### 1. Introduction

The subject of glassy materials has been of considerable interest for many years because disordered systems show universal physical properties that are strikingly different from those of their corresponding crystalline solids [1]. These peculiar characteristics can be found both in the low-temperature thermal conductivity and in the low-frequency vibrational density of states, which have been widely investigated by means of inelastic scattering experiments and specific heat measurements respectively [1]. At frequencies below some meV ( $\sim 100\text{cm}^{-1}$ ), inelastic neutron and Raman scattering measurements show the existence of modes in excess of the predictions of Debye theory. The intensity of the maximum of the bump in the  $g(\omega)/\omega^2$  versus  $\omega$  plot follows the Bose-Einstein law and is called the boson peak (BP) [2-9].

Moreover, at frequency below  $\sim 10\text{cm}^{-1}$ , the scattering data show an extra scattering close to the elastic line, which is temperature dependent and does not follow the temperature dependence expected for harmonic vibrations. This phenomenon is the so-called quasi-elastic scattering (QES), which can be considered as a broadening of the Rayleigh line [10-13]. The QES can be explained as arising from relaxation process. In glasses the relaxation processes can be mainly divided in two classes: the  $\beta$ -processes due to relaxations at temperatures usually far for the glass

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2 transition  $T_g$ , and the  $\alpha$ -processes which are the structural relaxations connected to the slow  
3 dynamics of the systems near  $T_g$ .

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5 Jackle [14-16] has found an explicit relation between the QES scattering intensity and the sound  
6 attenuation predicting that the QES intensity vs temperature should have the same behaviour of the  
7 acoustic attenuation. Recently Caponi et al. [17] have reported convincing results on  $v$ -SiO<sub>2</sub>  
8 demonstrating that the QES intensity and the  $\beta$ -relaxation are strictly connected.  
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11 In order to check the validity of the Jackle model for different types of relaxations we studied the  
12 QES behaviour as a function of temperature on two samples of superionic glasses  
13  $(\text{AgI})_x(\text{AgPO}_3)_{(1-x)}$  at different concentrations  $x$ . These glasses are a good candidate for our aim,  
14 not only because they are easy to prepare and have an easily accessible  $T_g$  ( $\sim 400$  K), but more  
15 interestingly they display an additional phenomenon due to the superionic diffusion (in the  
16 following labelled with the symbol  $\sigma$ ), well outside the range where  $\beta$  and  $\alpha$  relaxations take  
17 place. This superionic phenomenon is well known and it has been observed in low-frequency  
18 mechanical [18], ultrasonic [19], and high-frequency (GHz by Brillouin) studies [20]. Hence if the  
19 QES intensity shows a similar behaviour to the damping, this would be a further significant proof  
20 that they have the same physical origin.  
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## 32 33 2. Experimental details

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35  $(\text{AgI})_x(\text{AgPO}_3)_{(1-x)}$ , with molar fraction  $x = 0.4$  and  $x = 0.5$ , were obtained by a standard melt-  
36 quenching method. The original reagent-grade powders AgI (99% Aldrich), Ag<sub>2</sub>O (99% Aldrich)  
37 and H<sub>3</sub>PO<sub>4</sub> (crystals 98% Aldrich), accurately weighted and mixed, were melted in alumina  
38 crucibles for about 1 h at 400 °C and for 30 minutes at 500 °C. Subsequently, they were poured in  
39 stainless-steel moulds at room temperature. The obtained samples have different colours as a  
40 function of their  $x$ -composition, varying from white to yellow.  
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46 Raman scattering experiments were done using a double-monochromator Jobin Yvon U1000 in a  
47 standard experimental set-up. Measurements were performed in a wide frequency range (from 3 to  
48 3000 cm<sup>-1</sup>) in order to identify the shape of an underlying weak background of luminescence [21].  
49 At very low frequency ( $< 10$  cm<sup>-1</sup>) the double monochromator was set in order to achieve a 0.5  
50 cm<sup>-1</sup> resolution. The Raman spectra were taken in polarized (VV) geometry and collected on both  
51 samples as a function of temperature.  
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## 58 59 3. Result and discussion

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Figure 1 and 2 display the low-frequency Raman data at some significant temperatures for the two samples. The spectra are multiplied by such a coefficient that the high-frequency peak centred at  $1100\text{ cm}^{-1}$  are normalized to each other, as shown in the inset of figure 1.

### Figure 1 and 2 around here

In the inset of figure 2 the low-temperature spectrum at  $T = 15\text{ K}$  is reported only in order to evidence the position and the spectral shape of the BP in the absence of the QES contribution. The temperature behaviour of the BP is outside the scope of the present work. As can be seen from the figures, the QES contribution firstly increases as a function of  $T$  (open symbol) and then it decreases at high temperatures (full symbol).

In order to compare the QES intensity with the acoustic attenuation, the Raman intensity has been integrated in a spectral window from  $5$  to  $13\text{ cm}^{-1}$  for all the measured temperatures. The temperature dependence of this integrated QES intensity is shown in the upper panels of figures 3 and 4 for the two samples: we note an increase up to  $500\text{ K}$  then a decrease at higher temperatures; for the  $x = 0.5$  sample we observe two very large bumps centred at  $\sim 370$  and  $\sim 480\text{ K}$ , then a slow decrease at high temperatures, while for the  $x = 0.4$  sample we have a large peak centred at  $\sim 550\text{ K}$  and a decrease at high temperatures.

### Figure 3 and 4 around here

In the same figures (low panels) the temperature behaviour of the acoustic attenuation in the same samples is shown. These data are taken from the literature [20,22]. The Brillouin data for the  $x = 0.4$  sample disclose two distinct absorption peaks separated by  $\sim 50\text{ K}$ . The peak at the lowest temperature is attributed to damping of phonons by superionic diffusion ( $\sigma$ ) of  $\text{Ag}^+$  ions, while the second peak arises from structural relaxation ( $\alpha$ ). Comparing the positions between these two peaks for both samples it can be seen that their distance decreases with increasing AgI concentration as already reported in the literature [22]

As is evident from the experimental results reported in figures 3 and 4, there is a good qualitative agreement between the temperature behaviour of the QES intensity and the acoustic attenuation. It is worth noting that the precision in the evaluation of the QES intensity is not sufficiently high to separate the two peaks revealed by Brillouin scattering, as shown in figure 4 for the  $x = 0.4$  sample. Nevertheless the large bump present in the QES temperature behaviour in the  $x = 0.4$  sample is in the same range as the two peaks observed in the acoustic attenuation. This similarity is clearer in the  $x = 0.5$  sample where the separation of the two peaks measured by the acoustic attenuation is larger, and the QES data show the presence of two distinct features.

## 4. Conclusion

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The observed behaviour of the QES suggests it has the some microscopic origin as that which causes the damping of Brillouin lines. This similarity has been found also in v-SiO<sub>2</sub> regarding the  $\beta$ -relaxation [17]. Here we found that the QES scattering also seems to follows the damping behaviour due to  $\alpha$ -relaxation and also to Ag<sup>+</sup> ions diffusion mechanism.

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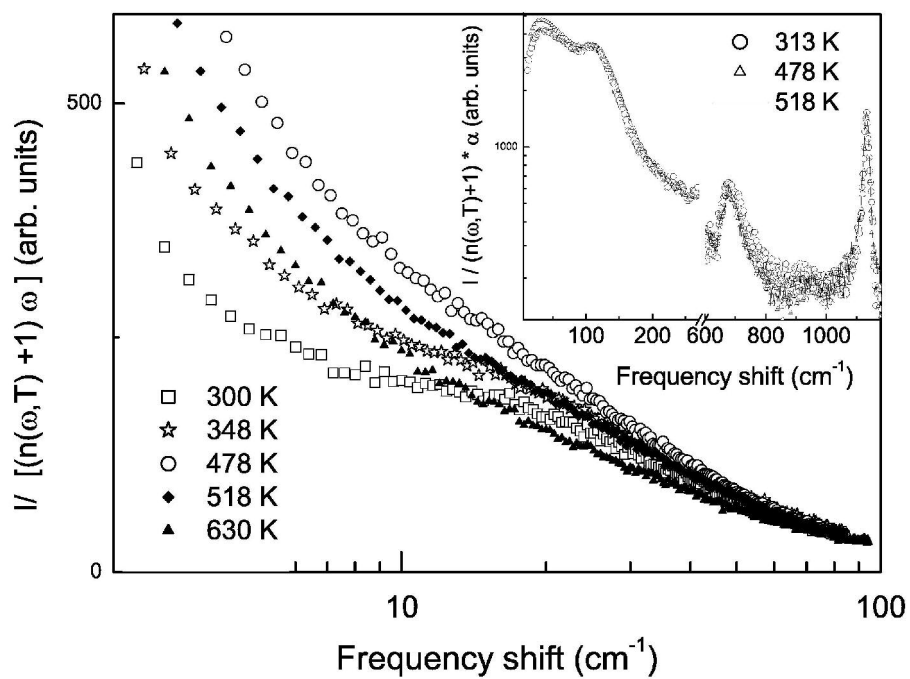
#### 28 FIGURE CAPTIONS

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32 Figure 1 Reduced Raman spectra of the  $x = 0.5$  sample taken in VV polarization at different  
33 temperatures as shown in the figure legend. In the inset, as a normalization example, we report the  
34 Raman spectra at  $T = 313$  K, 478 K and 518 K divided by the Bose-Einstein population factor and  
35 multiplied by an appropriate coefficient.  
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41 Figure 2. Reduced Raman spectra of the  $x = 0.4$  sample taken in VV polarization at different  
42 temperatures as shown in the figure legend. In the inset the reduced Raman spectrum taken at  $T =$   
43 15 K is reported.  
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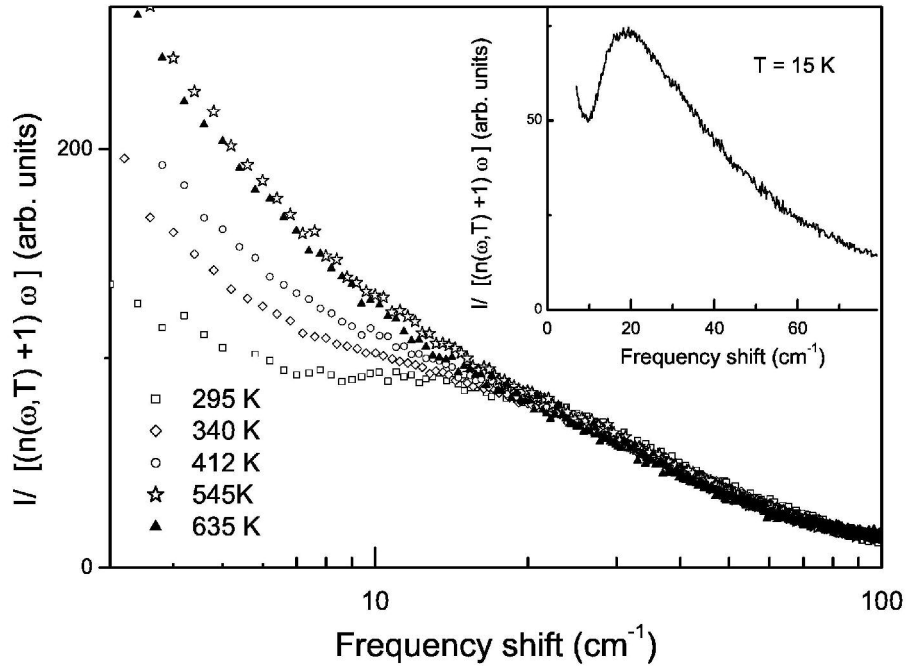
47  
48 Figure 3. Comparison between the QES data (upper panel), obtained as described in the text, and  
49 the acoustic attenuation (lower panel) [22], for  $(\text{AgI})_{0.5}(\text{AgPO}_3)_{0.5}$  in the temperature range  
50 between 300 K and 700 K.  
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55 Figure 4. Comparison between the QES data (upper panel), obtained as described in the text, and  
56 the acoustic attenuation (lower panel) [20], for  $(\text{AgI})_{0.4}(\text{AgPO}_3)_{0.4}$  in the temperature range  
57 between 300 K and 700 K.  
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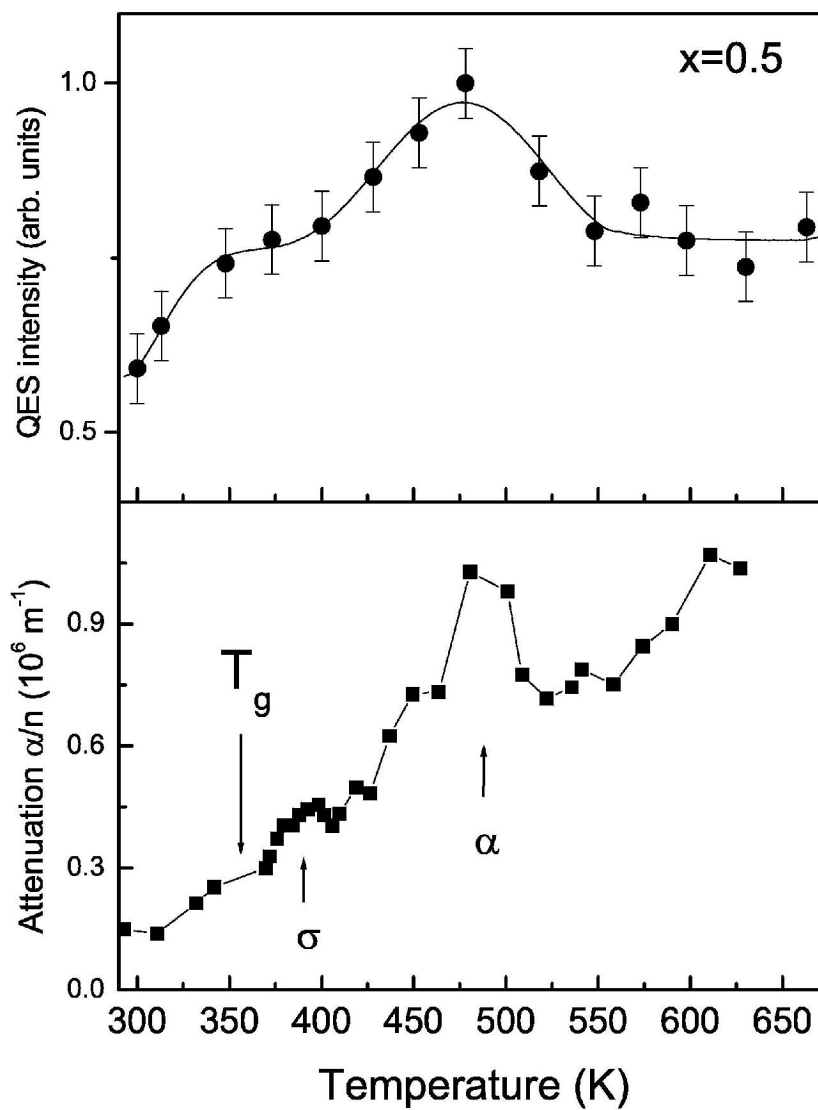
**Figure 1**  
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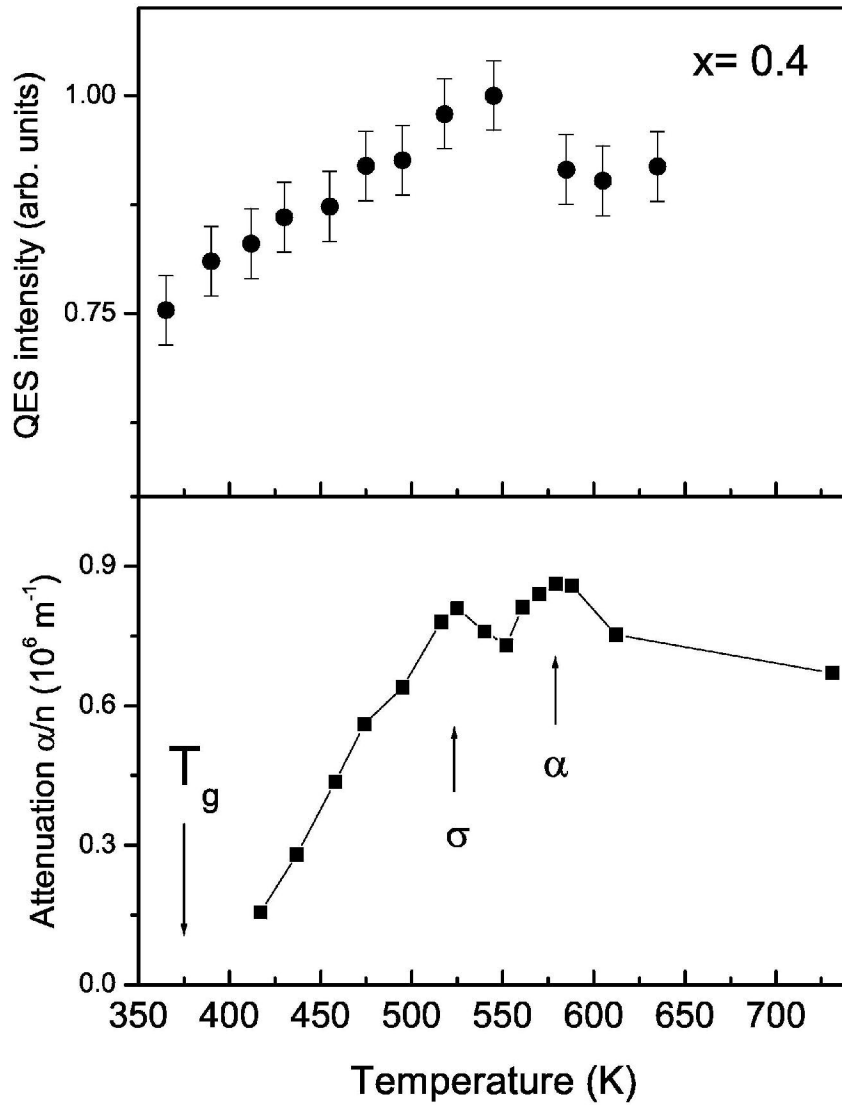


**Figure 2**  
109x83mm (600 x 600 DPI)

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**Figure 3**  
79x103mm (600 x 600 DPI)



**Figure 4**  
80x103mm (600 x 600 DPI)