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Effects of pressure on the boson peak of tellurite $(TeO_2)_{1-x}(ZnO)_x$ glasses: Evidence of an elastic glass-to-glass transition

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The Raman spectra of three $(TeO_2)_{1-x}(ZnO)_x$ (x=0.1, 0.2, and 0.4) glasses have been measured under pressure up to 13 GPa. We report an unusual pressure dependence of the Boson peak frequency which is quite different from that observed in relevant experiments for other network glasses or predicted by theory. The unexpected observed effect indicates that tellurite glasses respond like elastic, rather than plastic, media to both compression and decompression treatments. We suggest that, with respect to plastic versus elastic deformation mode, the behavior of glasses under pressure *is not universal*. Further, we conclude that a medium range order elastic glass-to-glass transition from a rigid to a more flexible network is induced by pressure for two of the glasses (x=0.2, 0.4). This transition is reversible upon decompression, merely exhibiting a small hysteresis.

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I. INTRODUCTION

The effects of high pressure on a variety of network glass systems have drawn considerable experimental[1–14] and theoretical[15–19] attention in recent years in an effort for a better understanding of their structure and dynamics. As a consequence of their topological disorder and large free volume, these glasses sustain significant densification at high pressure which can become permanent for several cases above certain threshold. Besides, pressure-induced structural changes are often observed in network glasses involving modifications of atomic coordination[2–13,15–19] or/and transformations of the medium range order.[1,7–10,12,13,16,17] In this respect, most of these glasses are known to exhibit polyamorphism by undergoing a series of glass-to-glass transitions under pressure.

It is generally accepted that the low-frequency vibrational component observed in inelastic light or neutron-scattering spectra of glasses, the so-called Boson peak (BP), is a characteristic feature of the disordered state and represents an excess vibrational density of states above the Debye level of the crystalline counterpart. In spite of numerous experimental and theoretical studies over the past decades, the scattering mechanism responsible for the BP is still a matter of debate and controversy.[20–25] However, it is broadly accepted that the BP is due to vibrations of the disordered medium beyond the atomic distance scale and, consequently, related to the medium-range order (MRO). Further, in all current theories[20–25] for the BP, additional non-Debye modes are considered to interact (hybridize) with Debye acoustic phonons. To throw some light in these issues, the effects of pressure on the BP and the vibrational density of states have been reported in experimental[14,26–31] and theoretical[32–35] studies of network[14,26–29,33–35] and polymer[30–32] glasses. Because of their large free volume, macroscopic quantities and properties of glasses, such as volume and density, are affected more strongly by pressure compared to the crystalline counterparts. Thus, most glasses studied so far respond to pressure largely like plastic media often resulting in a permanent densification. All relevant studies[14,26–35] agree in that the frequency (intensity) of the BP increases (decreases) with increasing pressure. In most experimental studies, though, the range of pressure exerted is limited to a few gigapascal.

In an analytical approach based on the soft-potential model (SPM) (Refs. 20 and 21) which takes into account both the influence of internal random forces acting on quasilocal vibrations and the pressure-induced (external) contribution to the total force, Gurevich *et al.*[35] have deduced the following expression for the frequency ω_b of the BP with pressure:

$$\omega_b(P) = \omega_b(0)\left(1 + \frac{P}{P_0}\right)^{1/3}, \quad (1)$$

where $\omega_b(0)$ is the BP frequency at ambient pressure and P_0 a parameter proportional to the ambient-pressure bulk modulus K_0 of the glass. For low pressures ($P \ll P_0$), Eq. (1) yields a linear variation in ω_b with pressure, while for $P \gg P_0$, a $P^{1/3}$ dependence is obtained. The SPM prediction for the frequency shift [Eq. (1)] is in good overall agreement with *in situ* BP data under pressure for the network glasses g-SiO_2 (Ref. 26), g-As_2S_3 (Ref. 27), and g-GeS_2 (Ref. 28), provided that $P_0(K)$ is (almost) pressure independent. However, significant discrepancies between theory and experiment have been noticed by Hong *et al.*[31] for the BP data of polymer glasses because of the strong pressure dependence of the bulk modulus. The above three network glasses have either tetrahedral (g-SiO_2, g-GeS_2) or trigonal (g-As_2S_3) local coordination and are highly compressible, hence displaying large BP shifts[26–28] with pressure. All experimental studies so far of the pressure dependence of the BP refer to selected glasses, rather than systematic studies of several members of a multicomponent glassy system with varying composition and essential properties like elastic moduli and glass transition temperatures.

Here, we report an unexpected pressure dependence of the BP in $(TeO_2)_{1-x}(ZnO)_x$ (x=0.1, 0.2, and 0.4) glasses deduced from *in situ* Raman measurements at pressures up to 13 GPa. In particular, the BP frequency ω_b displays an overall trend with pressure which is entirely different from that predicted[35] by the soft-potential model or observed[26–28] in other network glasses. This trend implies that the response of the various glassy systems to pressure cannot be considered as universal.

II. STRUCTURE AND PROPERTIES OF $(\mathrm{TeO_2})_{1-x}(\mathrm{ZnO})_x$ GLASSES

$(\mathrm{TeO_2})_{1-x}(\mathrm{ZnO})_x$ glasses have a three-dimensional structure with nontetrahedral, but tightly bound, local coordination. The stiffness at ambient conditions is relatively large,[36] i.e., the bulk modulus K varies between 35 and 50 GPa, depending on composition. These values of bulk modulus are much larger than those of chalcogenide glasses g-$\mathrm{As_2S_3}$ (Ref. 37) and g-$\mathrm{GeS_2}$ (Ref. 38) ($K \approx 13$ GPa and 12 GPa, respectively), and comparable to that of g-$\mathrm{SiO_2}$ (Refs. 39 and 40) ($K \approx 37$ GPa).

Pure $\mathrm{TeO_2}$ appears in several crystalline phases[41] and, under certain conditions, also forms a rather unstable glassy phase.[41] When combined with a metal-oxide modifier such as ZnO, stable $(\mathrm{TeO_2})_{1-x}(\mathrm{ZnO})_x$ glasses are obtained over the composition range: $0.05 \leq x \leq 0.45$ (Refs. 36 and 42–44).

The structure of pure and mixed tellurite glasses has been studied by diffraction[42] and spectroscopic[43,44] techniques showing that, like in the crystalline phases[41] of $\mathrm{TeO_2}$, the basic structural unit of their network is the $\mathrm{TeO_4}$ bipyramid. The introduction of the ZnO modifier results in a gradual deformation of $\mathrm{TeO_4}$ bipyramids leading to $\mathrm{TeO_{3+1}}$ polyhedra (with one Te—O bond being elongated and another shortened relatively to the rest) and trigonal $\mathrm{TeO_3}$ pyramids (having one nonbridged Te═O bond).[42,44] A neutron-diffraction study[42] suggests that ZnO is incorporated in the network in the form of $\mathrm{ZnO_6}$ octahedra linked with all other units through corner-sharing oxygen atoms. However, spectroscopic evidence[44,45] above the respective glass transition temperatures T_g has shown that mixed tellurite glasses crystallize to the same structure, that is to the *paratellurite* α-$\mathrm{TeO_2}$ structure,[41] irrespective of the type[44] or concentration[44,45] of the metal modifier. In fact, this evidence[44,45] raises a question whether the ZnO modifier participates in the $\mathrm{TeO_2}$ glassy network as has been suggested in Ref. 42.

In a network glass, the glass transition temperature T_g can be considered[8] as a measure of connectivity, that is, the higher the T_g the higher the percolation threshold for a breakup of network linking. We have determined the T_g values of the three glasses used in this study by differential scanning calorimetry:[45] $T_g = 658 \pm 4$, 644 ± 4, and 620 ± 4 K for the $x=0.1$, 0.2, and 0.4 compositions, respectively. Hence, the connectivity of the glasses should decrease with increasing x. This trend is plausible because the population of $\mathrm{TeO_{3+1}}$ and $\mathrm{TeO_3}$ units increases with x.

III. EXPERIMENTAL DETAILS AND DATA ANALYSIS

Small chips (typical dimension of ~ 50 μm) of melt quenched[46] samples of tellurite glasses were loaded into a diamond anvil cell (DAC) along with a tiny sphere of ruby for measuring the pressure through the shift of its R-line luminescence bands.[47] A mixture of methanol-ethanol-water in the ratio 16:3:1 was used as pressure-transmitting medium. Raman spectra were excited by the 476.5 nm line of an $\mathrm{Ar^+}$ laser at a power of ~ 10 mW measured just outside the pressure cell. Backscattered light was collected, analyzed, and detected by a Jobin-Yvon T 64000 triple spectrometer, operated in subtractive mode, combined with a liquid-nitrogen-cooled charge-coupled device. The spectral resolution of the system was about 3 $\mathrm{cm^{-1}}$ in all experiments. For each sample composition, Raman spectra were recorded in two runs, each of them including both compression and decompression.

The fitting procedures applied for the determination of Raman band frequencies, and particularly those used for the frequency of the BP, have been given in previous relevant articles.[45,48] As in Ref. 45, we have carried out tests to estimate the depth of overlapping between the high-frequency tail of the quasielastic scattering and the low-energy side of the BP profile, and assess whether the former scattering component affects the determination of the BP frequency. Details about the outcome of these tests for the BP spectra of this work are presented below in Sec. IV.

IV. RESULTS AND DISCUSSION

Representative Raman spectra of the $x=0.4$ glass at various pressures are shown in Fig. 1 for both (a) increasing and (b) decreasing pressure. The inset of Fig. 1(a) shows the well-resolved profile of the BP recorded at ambient conditions outside the high-pressure cell. As in previous studies,[14,26–28,30,31] the BP hardens (blueshifts) with pressure and this is accompanied by a suppression of its intensity. The shoulder on the high-energy tail of the BP (at ~ 130 $\mathrm{cm^{-1}}$) can be attributed to residuals of low-frequency optic modes of $\mathrm{TeO_2}$ crystal; the Raman spectrum of the latter[41] displays strong phonon bands in this region. The peak position of the midfrequency band is sensitive to ZnO content (it shifts to lower frequencies with increasing x) ranging from 446 ($x=0.1$) through 430 ($x=0.2$) to 414 $\mathrm{cm^{-1}}$ ($x=0.4$). This band, also observed in the Raman spectrum of pure $\mathrm{TeO_2}$ glass[41] at 450 $\mathrm{cm^{-1}}$, has been assigned[43,44] to bending vibrations in Te—O—Te bridges linking $\mathrm{TeO_4}$, $\mathrm{TeO_{3+1}}$, and $\mathrm{TeO_3}$ units. The position of the shoulder at 670 $\mathrm{cm^{-1}}$ (strongly present in pure $\mathrm{TeO_2}$ glass[41]) is composition independent and has been assigned[43,44] to stretching vibrations in $\mathrm{TeO_4}$ units. Finally, the intensity and position of the band at 750 $\mathrm{cm^{-1}}$ (absent in pure $\mathrm{TeO_2}$ glass[41]) depend on composition and the band is attributed[44] to stretching vibrations in $\mathrm{TeO_{3+1}}$ and $\mathrm{TeO_3}$ units.

Plots of the BP frequency ω_b against pressure for the three glasses are shown in Fig. 2. To make sure that the estimated ω_b values at ambient pressure, outside and inside the DAC, are not influenced by any contribution of the quasielastic scattering (QES) which may overlap with the low-energy side of the BP, we have fitted the former to a power-law function, as in recent works of tellurium-zinc-oxide glasses under variable temperature[45] and QES from polymer glasses at high pressure.[49] Then, the fitted QES has been extrapolated for $\omega > 15$ $\mathrm{cm^{-1}}$ and subtracted from the overall recorded spectrum. We have found that the contribution of

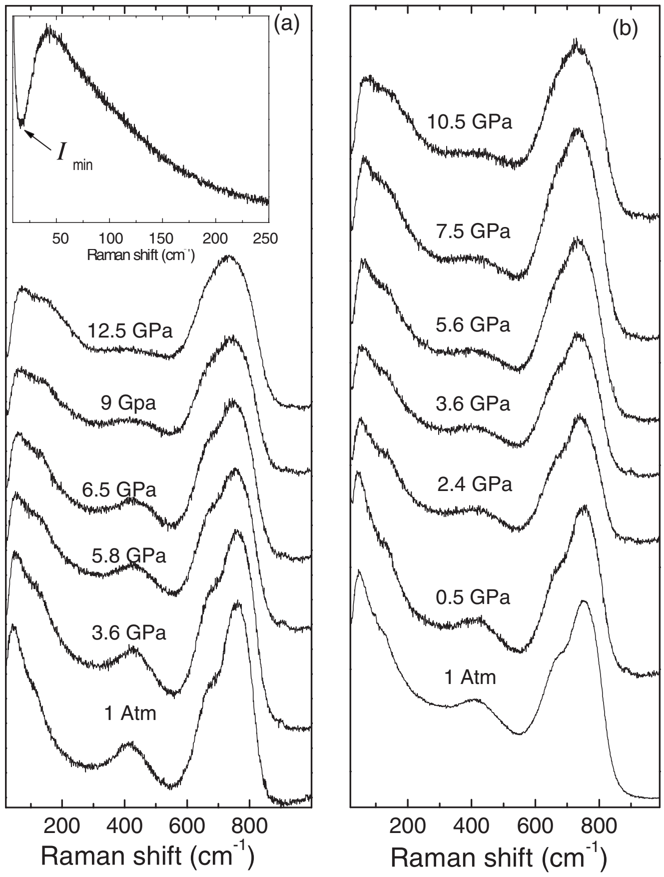

FIG. 1. Raman spectra of the $(TeO_2)_{0.6}(ZnO)_{0.4}$ glass at various pressures for the (a) compression and (b) decompression runs. The inset in (a) shows a detailed spectrum of the Boson peak at ambient conditions outside the high-pressure diamond anvil cell.

QES at the peak of the BP is negligible and the two ω_b values obtained with the sample outside and inside the DAC differ marginally, that is, within the error bars. This is not surprising as in strong glasses, like the tellurite glasses of this work, the vibrational component is dominant[50] over the relaxational (QES) one in the spectral region above the low-frequency intensity minimum I_{min} located below the BP frequency [inset of Fig. 1(a)]. With pressure application, the influence of the QES vanishes altogether since the BP shifts upward in frequency.

Throughout the full pressure range, the experimental points in the frequency ω_b versus pressure plots for the $x=0.1$ glass can be well fitted by a linear relationship [Fig. 2(a)]. A similar linear dependence on pressure of the midfrequency band (at 446 cm^{-1}) of this glass is observed in the corresponding plot of ω_{mfb} versus pressure and shown in Fig. 3. With the reminder that the composition of this glass is close to that of the crystalline compound, this result implies that at both medium and short-range order levels, the glass responds to pressure like an elastic medium for either compression or decompression, showing negligible hysteresis.

In the plots of the $x=0.2$ [Fig. 2(b)] and $x=0.4$ [Fig. 2(c)] glasses, an overall increase in the slope $\partial\omega_b/\partial P$ is clearly observed at $P_c \approx 7.5$ GPa and 6.0 GPa, respectively. The slope increase can be traced even without the two straight lines which have been produced, to assist the eyes, by fitting linearly the two sets of data points below and above P_c. Such a "concave" pressure dependence of the BP is unprecedented in all other known cases of network[26–28] or polymer[31] glasses which display a continuous decrease in $\partial\omega_b/\partial P$ slope with pressure, that is, a "convex" dependence. The increase in slope at P_c is most likely related with changes in elastic moduli and an elastic transition from rigid to a relatively flexible (compressible) network. Such changes in elastic moduli (and anomalies of shear sound velocity) were observed recently in g-$GeSe_2$ at high pressure by Antao *et al.*[10] who attributed them to a transition to a more flexible network involving also an increase in local coordination.

The change in slope and the proposed rigid-to-flexible elastic transition at P_c for the two glasses [Figs. 2(b) and 2(c)] suggest a drastic modification of their MRO. Such an MRO modification is most likely related with a decrease in network connectivity brought about by a reduction in cross-linking between units, and particularly TeO_{3+1} and TeO_3 polyhedra which tend to break from the backbone network of TeO_4 bypiramids at high pressures. This conclusion is compatible with the absence of such a structural change in the low ZnO concentration ($x=0.1$) glass in which the population of these units may not be sufficient[42] to cause significant decrease in connectivity. The occurrence of an MRO transition is further supported by the observation of a clear short-range order (SRO) change (discussed below) in the $x=0.4$ glass at about the same $P_c \approx 6.0$ GPa.

To find out whether there is any time dependence effect after pressure application, we have recorded, at several fixed pressure values (spanning through the entire pressure range of measurements), successive spectra of the $x=0.4$ glass at various time intervals, i.e., shortly after increasing the pressure, and at intervals of 12 h up to a total time lag of 48 h. We have found that the frequency of all Raman bands is practically unchanged as time elapses for both compression and decompression. Data points for the BP corresponding to different time lags are given in the ω_b-P plot of the $x=0.4$ glass [Fig. 2(c)]; in the compression run, a small consistent upward shift of ω_b with time is attributed to an increase in pressure in the DAC by up to 10% after 48 h. In fact, the time invariance of ω_b confirms the network elasticity of the glass. A slight hysteresis of ω_b displayed around P_c by this glass appears also to be time independent [Fig. 2(c)]. However, unlike the change in slope at P_c observed in the compression run, recovery of the glass to its original MRO structure appears to be continuous and almost linear [empty symbols of Fig. 2(c)]. A similar hysteresis is also displayed at P_c by the $x=0.2$ glass [Fig. 2(b)].

The midfrequency band of the $x=0.2$ glass hardens with pressure showing an overall decrease in slope $\partial\omega_{mfb}/\partial P$ (Fig. 3). In contrast, in the $x=0.4$ glass, after an initial moderate hardening up to ~6.0 GPa, this band softens and decreases in intensity with pressure (Fig. 1). This implies a change in the SRO with an increase in local coordination being the most likely scenario. This conclusion is supported by the apparent relative increase in intensity with pressure of the band at 670 cm^{-1} (fourfold coordinated TeO_4 bipyramids) to that at 750 cm^{-1} (lower coordinated TeO_{3+1} and TeO_3 units) (Fig. 1), indicating a population increase in the former units at the expense of the latter.

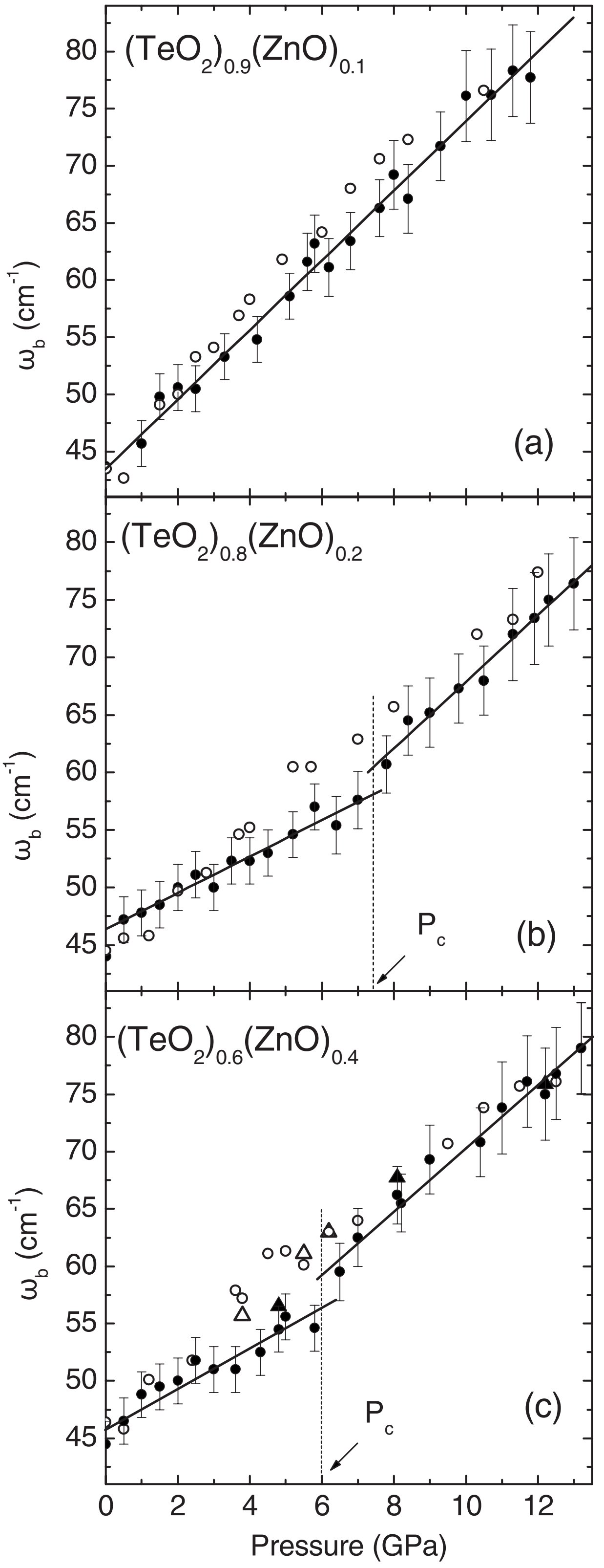

FIG. 2. Plots of Boson peak frequency ω_b against pressure for the three $(TeO_2)_{1-x}(ZnO)_x$ glasses: (a) $x=0.1$; (b) $x=0.2$; and (c) $x=0.4$. Full and open symbols denote the compression and decompression runs, respectively; circles: shortly after pressure application; triangles: after 48 h of pressure application. The linear fittings correspond to data points denoted by full circles (compression, shortly after pressure application).

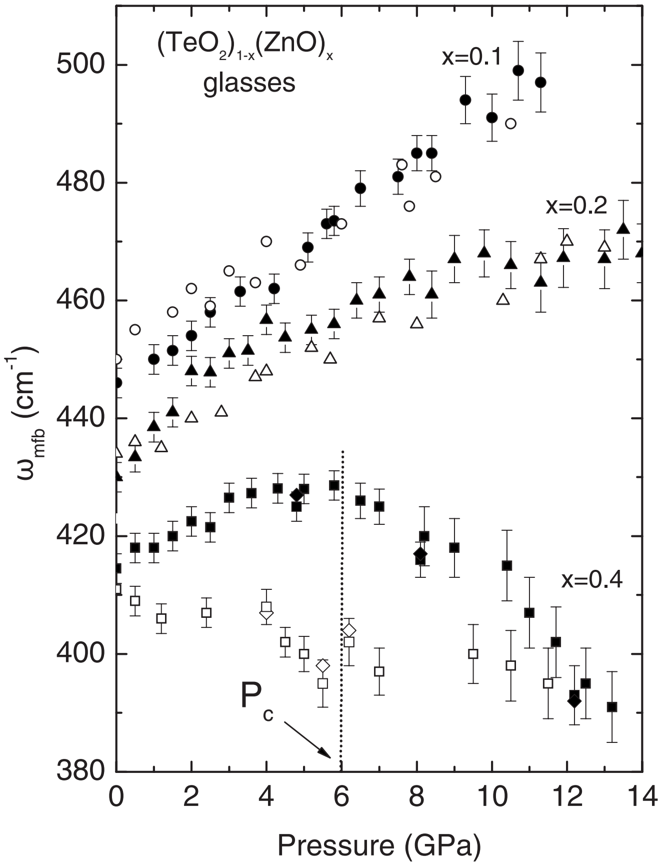

FIG. 3. Plots of frequency ω_{mfb} of the midfrequency Raman band in the 410–450 cm^{-1} region against pressure for the three $(TeO_2)_{1-x}(ZnO)_x$ glasses. Full and open symbols denote the compression and decompression runs, respectively; circles: $x=0.1$; triangles: $x=0.2$; squares: $x=0.4$, soon after pressure application; and diamonds: $x=0.4$, after 48 h of pressure application.

From neutron-diffraction data[42] of $(TeO_2)_{1-x}(ZnO)_x$ glasses at ambient pressure, the mean coordination number of Te for the $x=0.2$ glass was estimated to be 3.35 compared to 4 for crystalline TeO_2. Thus, in the $x=0.4$ glass this coordination number should be lower than 3.35 since the population of TeO_{3+1} and TeO_3 units is the highest for this composition. Therefore, an increase in Te coordination and local structural changes, under pressure, are more likely to occur in the latter glass than for any other composition of the system. The path of decompression for the SRO is almost reversible in the cases of the $x=0.1$ and 0.2 glasses (Fig. 3). In contrast, although the SRO of the $x=0.4$ glass appears to be fully restored after pressure release, the recovery route is different showing significant hysteresis (Fig. 3). However, like the MRO, the SRO of this glass appears also to be time independent after changing the pressure (Fig. 3).

Using the bulk modulus values $K_0 \approx 35$ and 42 GPa which are known[36] for the $x=0.1$ and 0.2 glasses and considering the $\partial\omega_b/\partial P$ slopes (linear fits) below P_c (Fig. 2), we have estimated the Grüneisen parameters for the BP: $\gamma_T=2.4\pm0.2$ ($x=0.1$) and 1.6 ± 0.2 ($x=0.2$). There are no K_0 data available for the $x=0.4$ glass, but an approximate value can be obtained by linearly extrapolating the data[36] in the composition range up to $x=0.2$ and by doing so, we obtain $K_0 \sim 50$ GPa and $\gamma_T = 1.7 \pm 0.3$ for this glass. These γ_T values are lower than those of g-SiO_2 ($\sim$7.0),[26] g-As_2S_3 ($\sim$4.0),[27] and g-GeS_2 ($\sim$2.6).[28] This indicates that tellurite glasses are not as compressible as the above glasses at low pressures but, it appears, they become more compressible for $P > P_c$ (larger $\partial\omega_b/\partial P$ slopes above P_c, Fig. 2). However, the nearly linear dependence of ω_b with pressure indicates that tellurite glasses remain elastic also above P_c.

As mentioned in Sec. II, a comparison of the glass transition temperatures T_g of the present glasses indicates that the connectivity decreases with increasing x. This trend provides a plausible explanation for the scaling with pressure of structural rearrangements observed in these glasses.

In a Raman study of chalcogenide Ge_xSe_{1-x} glasses under pressure, Wang *et al.*[8] reported that the effect of pressure on the SRO bonding is not immediate in most glasses, but it begins after pressure exceeds a threshold which depends on composition. We do not observe any pressure threshold in our results for either the BP (Fig. 2) or the midfrequency band (Fig. 3). This implies an immediate response of both MRO and SRO bonding and an elastic behavior of the glasses, similar to that of crystalline compounds.

Considering the broader category of disordered solids, an increase in local coordination number (change in SRO) can be viewed as a low- to high-density amorphous-to-amorphous transition (polyamorphism). Such a transition with abrupt or gradual increase in the average coordination number under pressure has been experimentally recognized in a variety of network disordered solids such as elemental amorphous solids,[51–53] amorphous ice[54] and oxide,[2–7] and chalcogenide[8–13] glasses. It is pointed out that the above-mentioned amorphous solids have networks which are built up by tetrahedral and also pyramidal (trigonal) units. On the other hand, a modification of the MRO has been also observed under pressure for a variety of network glasses such as SiO_2 (Refs. 1 and 55), GeO_2 (Ref. 7), $GeSe_2$ (Refs. 9 and 10), and GeS_2 (Refs. 12 and 13). Furthermore, this MRO change has been found to occur at a pressure close to the onset of the increase in coordination number.[5,7,10,13] The MRO modification can also be inferred from the anomalous pressure dependence of the elastic moduli observed for SiO_2 (Refs. 39 and 40) and $GeSe_2$ (Ref. 10) glasses. The effect is connected with the network rigidity and it occurs at pressures below the appearance of the SRO modification. As a result, the possibility of two different, overlapping or not, densification mechanisms[5] in glasses (i.e., changes in both MRO and SRO) must be further examined. In this respect, the present work can help toward a better understanding of the nature of the different densification mechanisms, since the network connectivity and coordination number variations in the glasses studied here result in different critical pressure regimes for each process. Raman spectroscopy, and in particular the systematic recording of the Boson peak if observable, could be useful for the study of the medium- and short-range orders in other categories of network glasses under pressure.

V. CONCLUSIONS

We have observed a unusual pressure dependence of the Boson peak in three $(TeO_2)_{1-x}(ZnO)_x$ glasses which is different from that seen in other network[26–28] and polymer[31] glasses, or predicted by the soft-potential model.[35] This indicates that the behavior of the Boson peak under pressure is *not universal* among the various glassy systems. In general, the response of $(TeO_2)_{1-x}(ZnO)_x$ glasses to high pressure is reminiscent more of an elastic (crystalline) medium than a typical plastic disordered solid. Furthermore, the pressure effects are imminent and independent of time after pressure application. An increase in the $\partial\omega_b/\partial P$ slope, in the frequency versus pressure plots of the Boson peak, observed in two glasses at high pressure implies an elastic transition from a rigid to a more flexible structure and this is accompanied, in one glass, by a local structure modification. All pressure-induced effects are reversible upon decompression, confirming the network elasticity of the glasses. The scaling of the pressure effects on the three glasses is compatible with their glass transition temperature T_g and the inferred network connectivity variation as a function of composition. The observed unexpected pressure dependence of the Boson peak frequency and the network elasticity displayed by tellurite glasses may prompt relevant experimental studies in other glassy systems and provide useful input for a theoretical description of the origin of the Boson peak in glasses.

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