

**Brillouin light scattering from transverse and longitudinal acoustic waves in glycerol**

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Brillouin light scattering experiments from transverse acoustic (TA) and longitudinal acoustic (LA) waves in glycerol have been performed in the temperature range between 170 and 229 K, including both the supercooled and the glassy state of the system. The intensity of light scattered by TA modes is about four orders of magnitude lower than that scattered by LA modes, giving an exceptionally low ratio of the photoelastic constants  $P_{44}/P_{12} \approx 0.01$ . The analysis of the frequency positions of Brillouin doublets gives a linear temperature behavior of both transverse and longitudinal elastic moduli for temperatures higher than that of the glass transition  $T_g = 187$  K, a bend at  $T_g$  and a less pronounced variation below  $T_g$ . A linear relation is also found between shear and longitudinal moduli of glycerol with angular coefficient close to 3, suggesting a Cauchy-like relation to hold around  $T_g$ .

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**I. INTRODUCTION**

The adiabatic propagation of longitudinal acoustic (LA) waves in liquids is described, in the hydrodynamic regime, by two limiting elastic moduli, the relaxed elastic modulus  $M_0$  and the unrelaxed modulus  $M_\infty$ . The first gives the adiabatic sound velocity in the equilibrated sample at low frequency  $c_0 = (M_0/\rho)^{1/2}$ . The second gives the glass like velocity of sound in the high frequency limit when the period of the wave is smaller than the time required by the structure to equilibrate under an external perturbation (structural relaxation time  $\tau_\alpha$ ). Conversely, the propagation of transverse acoustic (TA) waves is described by a single limiting elastic constant  $G_\infty$  since transverse waves does not propagate in the low frequency limit, i.e.,  $G_0 = 0$ . When decreasing the temperature, the progressive cessation of viscous flow is driven by the slowdown of the structural relaxation time, which passes from picoseconds to hundreds of seconds when the glass transition is approached from above.<sup>1</sup> The glass transition temperature  $T_g$  itself can be defined as the temperature corresponding to  $\tau_\alpha = 100$  s. Transverse acoustic (TA) waves can eventually propagate in a liquid, provided that their frequency is comparable to, or higher than, the reciprocal of the structural relaxation time. The relaxation time—or the distribution of relaxation times—and the relaxed and unrelaxed moduli are the parameters which govern the dispersion (and absorption) of acoustic waves in a liquid, and their experimental determination has always received a great deal of attention (see, for instance, Ref. 2). This attention has been recently renewed by the experimental observation of the validity of a Cauchy-like relation at the transition from the liquid to the solid state of a curing epoxy resin<sup>3</sup> and of different glass forming liquids.<sup>4</sup>

Brillouin light scattering (BLS) offers substantial advantages compared with ultrasonic techniques for the study of acoustic waves in liquids, since the frequencies involved (1–10 GHz) are higher than the ultrasonic ones ( $\approx 1$ –10 MHz) and unrelaxed values of both  $M$  and  $G$  can be measured up to temperatures significantly higher than  $T_g$ .

The intensity of the light scattered from TA modes in BLS experiments is strong in liquids of highly anisotropic molecules, where the rotational dynamics provides an effective channel for depolarized scattering.<sup>5,6</sup> The scattered intensity is much lower in systems with low molecular anisotropy and where the contributions from different scattering mechanisms add out of phase. This is possibly the case of glycerol. Indeed, despite the wide interest for the viscoelastic characterization of this prototypical glass former, no evidence has been reported till now of light scattered from shear waves in glycerol. The lack of information on the temperature evolution of  $G_\infty$  has been covered so far by different extrapolation schemes applied to the low frequency data obtained by ultrasonic techniques.<sup>7,8</sup>

In this paper we report the results of a Brillouin light scattering investigation of transverse and longitudinal acoustic modes in supercooled and glassy glycerol. The temperature behavior of the unrelaxed shear and longitudinal modulus obtained by Brillouin spectra is discussed and the validity of a Cauchy-like relation between the moduli is tested. Moreover, the ratio of the intensity of Brillouin peaks relative to longitudinal and transverse modes is estimated, giving the ratio of the photoelastic constants  $P_{44}/P_{12}$ .

**II. EXPERIMENTAL**

Glycerol of 99.5% purity purchased from Aldrich Chemicals was employed without further purification. The sample was transferred directly into a cylindrical pyrex cell (inner diameter 1 mm) used for the measurements. The cell was sealed in dry nitrogen environment. The sample was placed into a copper holder which was used to regulate the temperature by means of Cryomech ST405 cryostat. A Coherent-Innova 300 model Ar<sup>+</sup> laser was operated with a typical power of 300 mW as a single mode 514.5 nm light source. The light scattered from the sample at an angle  $\theta = 90^\circ$  was analyzed by means of a Sandercock-type (3+3)-pass tandem Fabry-Perot interferometer characterized by a finesse of about 100 and a contrast ratio higher than  $10^9$ . Unpolarized

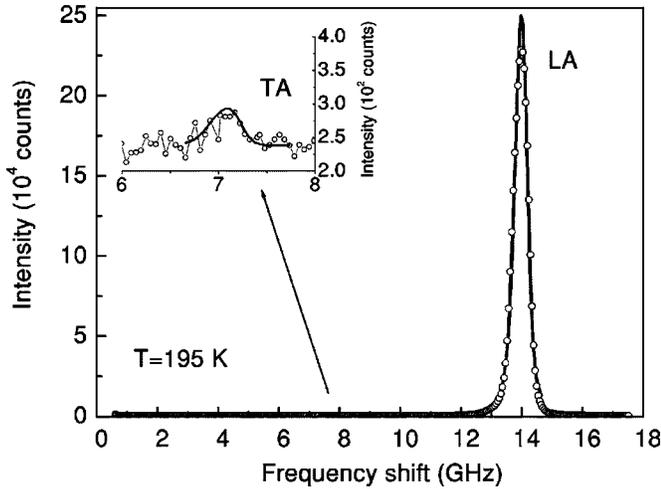


FIG. 1. Typical unpolarized Brillouin spectrum of glycerol collected in a  $90^\circ$  scattering geometry. The spectrum shows a peak due to the scattering from longitudinal acoustic (LA) modes at about 14 GHz. In the inset the small peak due to the scattering from transverse acoustic (TA) modes at about 7 GHz.

(VU) and depolarized (VH) spectra were collected above and below the glass transition temperature  $T_g = 187$  K. The peaks relative to transverse acoustic modes were detectable in the temperature region from 170 to 229 K. At higher temperatures, the transverse modes drown into the quasielastic peak of the longitudinal modes, whose intensity increases by increasing the temperature, while at lower temperatures the optical quality of the sample quickly degrades due to rising unrelaxed internal stresses.

### III. RESULTS AND DISCUSSION

The unpolarized spectrum in Fig. 1 shows a strong anelastic peak due to the scattering from LA modes at about 14 GHz and a second weaker peak at about 7 GHz due to the light scattered from TA modes. Only the peak at 7 GHz survives in depolarized spectra (Fig. 2) demonstrating the transverse nature of the mode. Since the depolarized condition improves the signal to noise ratio, this configuration is used to investigate the temperature evolution of the frequency of TA modes.

The characteristic frequency  $\omega_0$  and linewidth  $\Gamma$  of longitudinal and transverse acoustic waves are obtained by fitting the Brillouin peaks with the damped harmonic oscillator (DHO) model function

$$I(q, \omega) = \frac{I_0}{\pi} \frac{\Gamma \omega_0^2}{(\omega^2 - \omega_0^2)^2 + (\Gamma \omega)^2} \quad (1)$$

convoluted with the instrumental function. In this formula  $I_0$  is the area of the peak, which will be used to gain information on the photoelastic properties of the sample.

#### A. Elastic properties of glycerol

The unrelaxed longitudinal modulus is obtained from the frequency position of longitudinal modes  $\omega_0(\text{LA})$  through the relationship

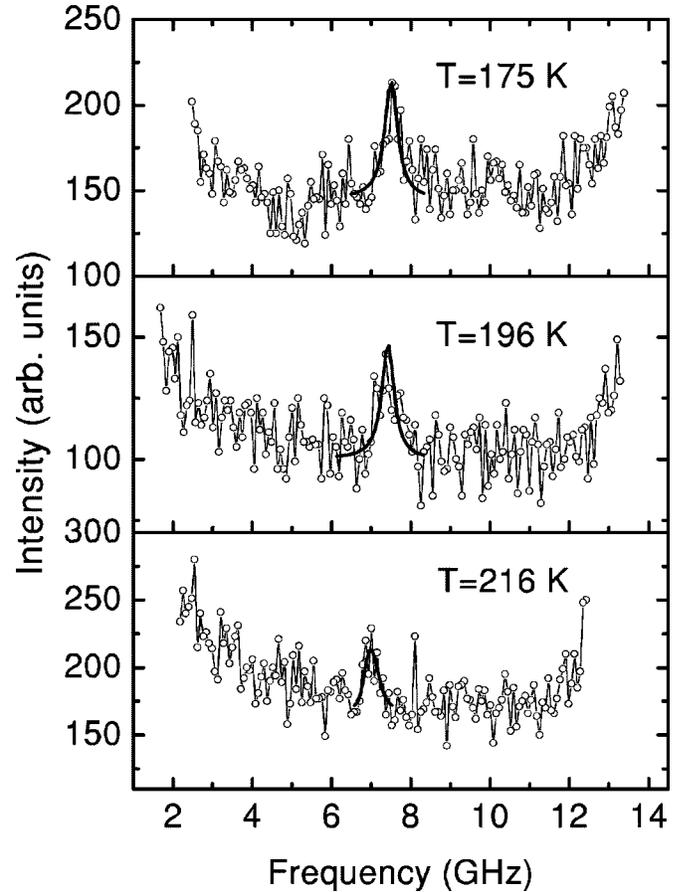


FIG. 2. Depolarized spectra of glycerol at three different temperatures. The typical accumulation time for each spectrum is about 12 h.

$$M_\infty = \frac{\omega_0^2(\text{LA})\rho}{q^2}. \quad (2)$$

The mass density  $\rho$  for the liquid phase ( $T > T_g$ ) is taken from Ref. 9,  $\rho(T) = 1.2723 - 6.55 \times 10^{-4}(T - 273.2)$ , with  $\rho$  in  $\text{g}/\text{cm}^3$  and  $T$  in K. For temperatures  $T < T_g$ , we use the thermal expansion coefficient of the glass at 180 K,  $\alpha_{\text{glass}} = 2.4 \times 10^{-4} \text{ K}^{-1}$  from Ref. 2 and the value  $\rho(T_g) = 1.3288 \text{ g}/\text{cm}^3$ .

The exchanged wave number  $q$  is related to the wave number  $K_i$  of the incident light through the relationship  $q = 2nK_i \sin \theta/2$ , where the refractive index  $n$  of the medium is estimated through the Clausius-Mossotti relationship using the known value  $n = 14754$  at 300 K and 514.5 nm.<sup>10</sup> The values of  $M_\infty$  obtained through this procedure are shown in Fig. 3 in comparison with those previously reported in a back-scattering investigation of glycerol.<sup>11</sup>

We notice that Eq. (2) rigorously holds in absence of relaxation processes. This is a reasonable approximation in the whole temperature range since we have shown in a previous investigation<sup>11</sup> that (i) the structural relaxation is the only process to considerably affect density fluctuations of glycerol in the GHz frequency region and (ii) the structural relaxation time is longer than  $1 \mu\text{s}$  in this  $T$  range, so that it has a negligible influence on the velocity of sound at high fre-

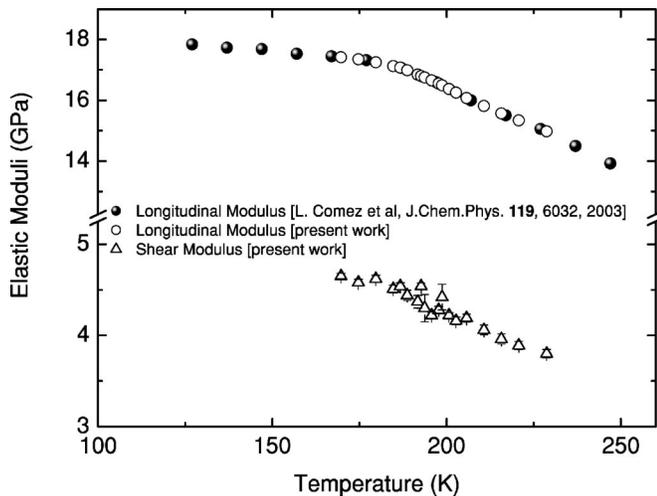


FIG. 3. Longitudinal and shear elastic moduli of glycerol from this work (open symbols) and from a previous investigation (full circles).

quency. To quantify this influence we can say that the value of the modulus at  $T=210$  K evaluated by means of Eq. (2) is  $(1.59 \pm 0.02) \times 10^{10}$  Pa, while the unrelaxed longitudinal modulus obtained through a full viscoelastic analysis of Brillouin spectra in Ref. 11 is  $(1.60 \pm 0.02) \times 10^{10}$  Pa.

The temperature evolution of the unrelaxed shear modulus  $G_{\infty}$  is obtained from  $\omega_0(TA)$  through the same procedure adopted for LA modes and the results are shown in Figs. 3 and 4 and reported in Table I. Figure 4 also shows a comparison between our data and literature data, which were obtained by an extrapolation of ultrasonic measurements (full squares, from Refs. 7 and 8). The gap of about 4% between the temperature behavior of  $G_{\infty}$  obtained from our measurements and that obtained from ultrasonic data might be ascribed both to different degrees of purity of the samples and to different data analysis procedures. In fact, the water content in our sample was less than 0.5%, while it was about 0.8% in the sample used in Ref. 6. Moreover, Brillouin spectra directly access the high frequency limit of the relaxing modulus, i.e.,  $G_{\infty}$ , in the whole temperature range, while in the ultrasonic investigation its value was calculated by an extrapolation of the relaxation curve at the single temperature  $T=233$  K, and assuming for  $G_{\infty}(T)$  the same temperature dependence as for the longitudinal modulus  $M_{\infty}(T)$ . Both the higher purity of the sample and the absence of extrapolations in our procedure plays in favor of the reliability of BLS results. Nonetheless, looking at Fig. 2 it is to be noticed that, apart from the small gap, the temperature behavior of our data is linear with a slope very close to that of the ultrasonic data. The linear fit of  $G_{\infty}(T)$  yields  $G_{\infty}(T) = (7.8 \pm 0.4) \times 10^9 - (1.77 \pm 0.20) \times 10^7 T$  where  $G_{\infty}$  is in Pa and  $T$  in K. This behavior is reported as a full line in Fig. 4. It has to be noticed that the linear dependence here obtained for  $G_{\infty}(T)$  is not in accordance with the linear behavior of the shear compliance ( $G_{\infty}^{-1}$ ) suggested in Ref. 12 and tested in different glass formers in a narrow temperature region above  $T_g$  (Ref. 13) including extrapolated data of glycerol.<sup>14</sup> The dashed line in Fig. 4 is obtained from that model, using the

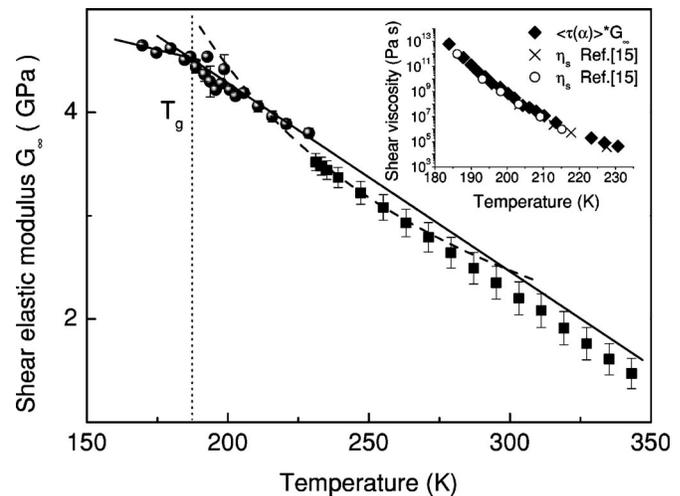


FIG. 4. Temperature dependence of the shear modulus of glycerol. Full circles are from this work, full squares are from Refs. 6 and 7. The solid line above  $T_g$  is the linear fit of the modulus  $G_{\infty}(T) = (7.8 \pm 0.4) \times 10^9 - (1.77 \pm 0.20) \times 10^7 T$ , where  $G_{\infty}$  is in Pa and  $T$  is in K. The dashed line is a fit of our data according to the empirical model of Refs. 11 and 12, where a linear dependence of  $G_{\infty}^{-1}$  is proposed. The solid line below  $T_g$  is a guide for the eye. Inset: Temperature dependence of static shear viscosity. Diamonds represent calculated shear viscosity using the relationship  $\eta_s = G_{\infty} * \langle \tau(\alpha) \rangle$ , where the values of  $G_{\infty}$  are from present work and  $\langle \tau(\alpha) \rangle$  data are from Ref. 16. Open circles and crosses represent measured shear viscosity from Ref. 15.

parameters for glycerol given in Ref. 13. It is apparent from the figure that this model fails in describing the temperature behavior of the unrelaxed shear modulus of glycerol obtained in our experiment.

The values of  $G_{\infty}$  here obtained can be used to estimate the shear viscosity  $\eta_s$  through the well-known Maxwell relationship  $\eta_s = G_{\infty} * \langle \tau(\alpha) \rangle_{TA}$ , to be compared with experimental values of static shear viscosity. The inset of Fig. 4 shows static shear viscosity data taken from Ref. 15 and  $\eta_s$  values obtained through the Maxwell equation, using  $\langle \tau(\alpha) \rangle_{DS}$  data measured by dielectric spectroscopy,<sup>16</sup> the only set of structural relaxation times available in the temperature region here investigated. The approximation  $\langle \tau(\alpha) \rangle_{TA} \approx \langle \tau(\alpha) \rangle_{DS}$  here adopted is established upon the merely phenomenological evidence that the time scale of different observables tends to collapse when approaching the glass transition.<sup>17</sup>

The good agreement among the different series of data is evident in the inset of Fig. 4. We notice that this procedure was successfully performed in a previous BLS investigation of  $ZnCl_2$ ,<sup>18</sup> a glass former which shares with glycerol an  $\alpha$ -relaxation-only scenario.<sup>11</sup> In that case Brillouin spectra were taken in the supercooled liquid, in a temperature region where both longitudinal and shear relaxation times could be estimated and a clear consistence between viscosity and relaxation data could be established.

Finally, we notice that appreciable deviations from experimental values of  $\eta_s$  are usually found in systems showing secondary relaxations or strong rototranslational coupling.<sup>19</sup> Even if translation-rotation coupling in glycerol has never

TABLE I. Temperature dependence of the shear modulus of glycerol.

$T(K)$	$G(\text{GPa})$
169.7	$4.65 \pm 0.03$
174.7	$4.58 \pm 0.04$
179.7	$4.62 \pm 0.04$
184.7	$4.51 \pm 0.04$
186.7	$4.54 \pm 0.04$
188.7	$4.44 \pm 0.06$
191.7	$4.37 \pm 0.07$
192.7	$4.54 \pm 0.03$
193.7	$4.30 \pm 0.15$
195.7	$4.22 \pm 0.04$
197.7	$4.28 \pm 0.04$
198.7	$4.42 \pm 0.14$
200.7	$4.22 \pm 0.03$
202.7	$4.16 \pm 0.04$
205.7	$4.19 \pm 0.04$
210.7	$4.06 \pm 0.05$
215.7	$3.96 \pm 0.05$
220.7	$3.89 \pm 0.05$
228.7	$3.80 \pm 0.05$

been investigated in depth up to now, the results of our analysis could be considered as an indication of weak coupling effects in the system.

**Cauchy-like relation.** If atoms interact through a central potential, the Cauchy identity holds which, in an isotropic system, reads  $M=3G$ .<sup>20</sup> Molecular liquids are frequently far from showing a central potential and Cauchy relation does not apply in its original version. Nevertheless, Zwanzig *et al.*<sup>21</sup> have calculated a generalized Cauchy relation  $M_\infty=3G_\infty+f(T,P)$  for isotropic liquids, which contains the additive term  $f(T,P)$  depending on both temperature and pressure. More recently Yamura *et al.*<sup>3</sup> and Krüger *et al.*<sup>4</sup> found a Cauchy-like relation to hold across the glass transition of different liquids, including curing epoxy systems

$$M_\infty = A + BG_\infty, \quad (3)$$

where  $A$  is a system dependent constant which, at ambient pressure, does not appreciably depend on temperature and  $B$  is found always close to 3. Remarkably, similar  $A$  and  $B$  parameters have been found in both freezing and polymerization experiments, showing a general behavior for the solid-like amorphous state.

In order to verify the validity of Cauchy relation in glycerol, the behavior of the unrelaxed elastic moduli  $M_\infty$  and  $G_\infty$ , is compared with Eq. (3). A linear law is fulfilled within experimental uncertainty, giving  $A=(4.61 \pm 0.85)$  GPa and  $B=(2.77 \pm 0.11)$  as best-fit parameters. Figure 5 shows experimental data together with the fit curve.

Krüger *et al.*<sup>4</sup> have also shown how the parameters  $A$  and  $B$  can be related to the acoustic mode Grüneisen parameters (MGP's)  $\gamma_i$  so that the Cauchy-like condition concerns also

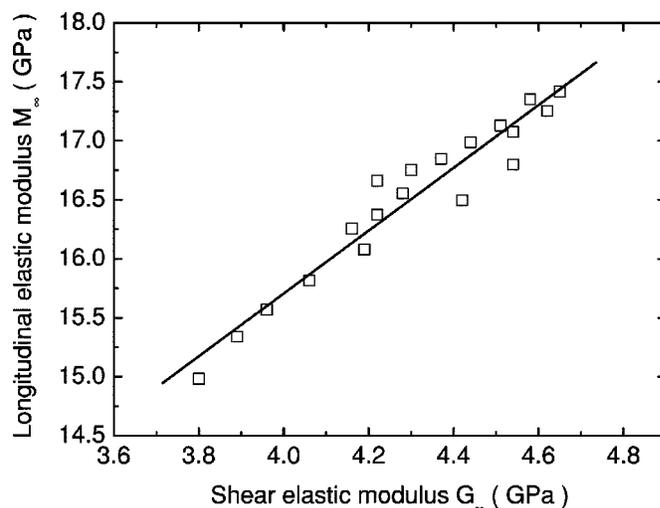


FIG. 5. Linear fit of the longitudinal elastic modulus vs. the transverse elastic modulus of glycerol in the whole investigated temperature range.

the nonlinear elastic properties of the isotropic state of matter. MGP's can be measured by means of Brillouin spectroscopy since they relate the temperature dependence of the characteristic frequency of the acoustic modes to the  $T$  dependence of density through the relationship

$$\gamma_i[v_i^q(T), \rho(T)] = -\frac{d \ln[v_i^q(T)]}{d \ln[\rho(T)]} = \frac{\rho(T_g)[dv_i^q(T)/dT]}{v_i^q(T)[d\rho(T)/dT]}, \quad (4)$$

where  $i=1,4$  is the polarization of the mode (longitudinal and transverse, respectively),  $q$  is the wave vector (it will be omitted in the following for simplicity),  $v_i^q(T)$  is the frequency of the acoustic mode,  $T$  is the temperature, and  $\rho(T)$  is the mass density of the sample.

Using the short notation  $k_{l,g}=2M_\infty(T_g)/(1+2\gamma_4^{l,g})$  and  $\Delta\gamma_{4-1}^{l,g}=\gamma_4^{l,g}-\gamma_1^{l,g}$  [each for liquid ( $l$ ) and glassy ( $g$ ) phases], Eq. (4) yields

$$M_\infty^{l,g} = k^{l,g} \Delta\gamma_{4-1}^{l,g} + \left( \frac{M_\infty(T_g)}{G_\infty(T_g)} - \frac{k^{l,g} \Delta\gamma_{4-1}^{l,g}}{G_\infty(T_g)} \right) G_\infty^{l,g}. \quad (5)$$

Equation (5) corresponds to Eq. (3), with  $A=k^{l,g}\Delta\gamma_{4-1}^{l,g}$  and  $B=[M_\infty(T_g)/G_\infty(T_g)-k^{l,g}\Delta\gamma_{4-1}^{l,g}/G_\infty(T_g)]$ . The simultaneous validity of the linear relationship, expressed in Eq. (5), for the liquid and for the glassy state has already been tested in different glass formers,<sup>4</sup> giving the same  $A$  and  $B$  parameters for temperatures above  $T_g$  as well as below  $T_g$ .

In order to extend this analysis to the case of glycerol, given the poor statistics of the data in the glassy state, our calculations are performed in the supercooled regime obtaining  $M_\infty(T_g)=(17.1 \pm 0.2)$  GPa,  $G_\infty(T_g)=(4.53 \pm 0.08)$  GPa,  $\gamma_1^l=3.08 \pm 0.03$ ,  $\gamma_4^l=4.12 \pm 0.25$ , and  $k^l=(3.70 \pm 0.21)$  GPa. Accordingly, the coefficients  $A$  and  $B$  of Eq. (5), are calculated:  $A=(3.83 \pm 0.24)$  GPa and  $B=(2.93 \pm 0.17)$ . Notice that the values of both  $A$  and  $B$  are in satisfactory agreement with those obtained through Eq. (3) and that the value of  $B$  close to 3 agrees with previous evaluations on organic and inorganic glass formers.<sup>4</sup>

TABLE II. Absolute value of the ratio of the photoelastic constants of glycerol as obtained from Brillouin spectra using Eq. (6) and from the DID formula of Eq. (7).

$T(K)$	$ P_{44}/P_{12} $	$P_{44}/P_{12}$ (DID)
188.7	$0.011 \pm 0.002$	$0.203 \pm 0.002$
190.7	$0.011 \pm 0.001$	$0.203 \pm 0.002$
192.7	$0.010 \pm 0.001$	$0.202 \pm 0.002$
194.7	$0.007 \pm 0.002$	$0.202 \pm 0.002$

### B. Photoelastic properties of glycerol

The propagation of elastic waves in a continuous medium produces fluctuations in the strain tensor which induces fluctuations in the dielectric constant responsible for Brillouin scattering processes. The dielectric constant can be expressed as linear combination of strain tensor elements,<sup>22</sup> the coefficients of the combination being the photoelastic (or elasto-optic or Pockels) constants  $P_{ij}$ . In isotropic systems there are only two independent photoelastic constants and the ratio of these constants can be related to the intensity  $I_0$  and the frequency  $\omega_0$  of the peaks in our unpolarized (VU) experiments through the relation

$$\left(\frac{P_{44}}{P_{12}}\right)^2 = \frac{I_0(TA) \omega_0^2(TA)}{I_0(LA) \omega_0^2(LA)}. \quad (6)$$

The values obtained for the ratio  $|P_{44}/P_{12}|$  at four different temperatures close to  $T_g$  are reported in Table II. The very low intensity of the light scattered by TA modes is reflected in the low value of  $|P_{44}/P_{12}|=0.01$ , almost temperature independent.

A microscopic pure dipole-induced-dipole (DID) model was recently proposed for evaluating photoelastic constants in disordered isotropic solids composed of units carrying spherical polarizability.<sup>23</sup> That model predicts that the photoelastic constants are only determined by the dielectric constant  $\epsilon$  of the system

$$\left[\frac{P_{44}}{P_{12}}\right]_{\text{DID}} = \frac{0.2(\epsilon - 1)}{1 + 0.2(\epsilon - 1)}. \quad (7)$$

A comparison with photo-elastic constants measured in a number of silicate glasses having a refractive index ranging between 1.5 and 2 was also reported.<sup>23</sup> A systematic difference of  $0.40 \pm 0.06$  was found between theoretical and experimental values of the photoelastic ratio attributed to possible “local” contributions, such as bond polarizability effects.

Our evaluations of Eq. (7) for glycerol are reported in Table II together the measured values of  $|P_{44}/P_{12}|$ . A large deviation of about 0.2 is found between the prediction of the DID model and the measured values of the photoelastic ratio. This suggests that also in case of glycerol the spherical polarizability approximation is not sufficient and the presence of local effects can be hypothesized. To this respect the absence in glycerol of the strong bonds typical of  $\text{SiO}_2$  molecules could explain the better agreement of Eq. (7) with the values of  $P_{44}/P_{12}$  measured in glycerol with respect to those measured in silicate glasses.

### IV. CONCLUSIONS

The task of revealing light scattered from transverse acoustic modes of glycerol is particularly challenging and, up to now, inaccessible to experimentalists due to the very low Brillouin cross section of these modes. Our measurements succeeded in achieving this task thanks to the appropriate temperature range around  $T_g$  chosen to minimize the damping and to get a good optical quality of the sample, and thanks to the use of a high contrast Sandercock-type Fabry-Perot interferometer.

The microscopic origin of the very low cross section of TA modes in glycerol, which is responsible for the low ratio  $P_{44}/P_{12} \approx 0.01$ , is still unclear. This ratio is even lower than that which can be expected by a pure dipole-induced-dipole mechanism of scattering<sup>23</sup> and deserves further theoretical investigation, possibly invoking bond polarizability effects.

From the analysis of the frequency position of Brillouin peaks a linear behavior of the unrelaxed shear modulus was recognized above  $T_g$ , rather unexpected when compared with the linear dependence of the unrelaxed compliance previously documented in a number of glass forming systems.<sup>12</sup> Moreover, a linear relation was found between shear and longitudinal moduli of glycerol with an angular coefficient close to 3. This evidence, corroborated by a detailed analysis of the acoustic mode Grüneisen parameters, plays in favor of the generalized Cauchy-like relation for glycerol around  $T_g$ . An even more general validity of this relation could be inferred from the results of a systematic investigation of silicate glasses,<sup>23</sup> where a ratio ( $M/G$ ) close to 3 was found. On the other hand, it should be noticed that the values of  $M$  and  $G$  of  $\text{ZnCl}_2$  reported in Ref. 18, seems to be not consistent with the Cauchy-like relationship. Thus, it results that the degree of universality of the Cauchy-like relation is not well understood up to now and constitutes a challenge for further experimental investigations.

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