Glassy dynamics in mono-, di- and tri-propylene glycol: from the α- to the fast β-relaxation

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Glassy dynamics in mono-, di- and tri-propylene glycol: From the α- to the fast β-relaxation

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1. Introduction

The dynamics of glassy matter reveals a variety of unusual and so far only poorly understood phenomena. A vast number of publications treat the structural α-relaxation, the 'excess wing' and the Johari–Goldstein (JG) relaxation [1]. While even the α-relaxation, known since the beginning of the research of glassy dynamics, still involves many open issues, the microscopic origin of the latter two features is unclarified at all. Sometimes excess wing and JG-relaxation peak are discussed on the same footing [2,3] but also this notion is not finally settled.

Another typical feature of glassy matter is the so-called fast β-relaxation, showing up in the GHz–THz range. This region attracted much interest during recent years as this fast dynamics may be the key for a better understanding of the glass transition and the glassy state of matter in general. By dielectric spectroscopy this high-frequency region is only accessible under great effort and likewise extremely challenging. For this reason, in the high-frequency regime only sparse data can be found in literature [4–10]. The fast β-relaxation is predicted by mode coupling theory (MCT) and ascribed to the 'rattling' motion of a particle in the transient cage formed by its neighboring molecules [11]. The fast β-process usually is assumed to show up as a shallow loss minimum in the GHz–THz region. There are also other approaches to explain the excess intensity observed in this high-frequency region. Maybe the most prominent one is the extended coupling model [12,13], which involves an explanation in terms of a nearly constant loss, a phenomenon which is known since long [14]. So far, two molecular and two ionic glass formers have been investigated by dielectric spectroscopy in a sufficiently broad and continuous frequency range to allow for a meaningful analysis of the fast β-relaxation [5,7–10]. In the present work, we considerably extend the database by providing results for another three glass formers.

Many groups studied the dynamics of propylene glycols, both the monomer as well as glycols exhibiting different grades of polymerization. Different techniques were used for characterization, among them neutron [15] and light scattering [16–21], optical Kerr-effect [22] as well as photothermal [23] and dielectric spectroscopy [3,24–31] including elaborated studies at elevated pressure [27–30]. Moreover, different models were applied to the experimental data, especially the coupling model [12,13,32], the minimal model [33,34], a non-monotonic relaxation kinetic model [35] and an approach considering the hybridization of a slower unresolved JG β- with a faster γ-relaxation [36]. However, in most dielectric experiments reported so far, the frequency range was restricted to frequencies below 10 MHz, except for [25] where spectra up to 1 GHz were reported.

In the present work, we show broadband spectra ranging from mHz up to the THz region for propylene glycol (PG) and its di- and trimer (DPG and TPG, respectively). The main purpose of the present work is to provide real broadband spectra, including the fast β-region for three more molecular glass formers, in addition to the two materials investigated so far (glycerol and propylene carbonate [5,9,10]). As the investigated glycols are closely related, one can expect information on the influence of the molecular size on glassy dynamics. In addition, a first phenomenological analysis of all the observed processes of glassy dynamics is presented and a preliminary evaluation of the fast β-relaxation in terms of the standard version of MCT is performed.

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2. Experimental procedures

A combination of different experimental techniques is necessary to record the real and imaginary part of the dielectric permittivity in the broad frequency range covered by the present work. For details the reader is referred to [5,37]. Propylene glycol was purchased from Fluka with a purity $\geqslant 99.5\%$, dipropylene glycol from Aldrich with a purity $>99\%$ and tripropylene glycol from Alfa Aesar with a purity $\geqslant 98.91\%$. All samples were used without further purification. Glass-transition temperatures were determined by differential scanning calorimetry (DSC) using a *Perkin–Elmer DSC Pyris 1* using cooling and heating rates of 10 K/min.

3. Results and discussion

In Fig. 1, the frequency-dependent dielectric loss $\varepsilon''(\nu)$ of (a) PG, (b) DPG and (c) TPG is shown for various temperatures. All spectra are dominated by the well-known α-relaxation peaks, whose strong temperature-dependent shift displays the tremendous slowing down of glassy dynamics towards the glass transition. In addition, at frequencies beyond the α-relaxation an excess wing is observed for PG whereas the dimer and trimer reveal well-pronounced secondary relaxation peaks [3,29,31]. With increasing temperature, the latter become successively submerged under the dominating α-peak leading to an excess wing at about 210 K. Finally, above about 240 K, α- and β-relaxations are merged. In the last two frequency decades, a shallow minimum shows up in all three materials. Its magnitude increases and it shifts to higher
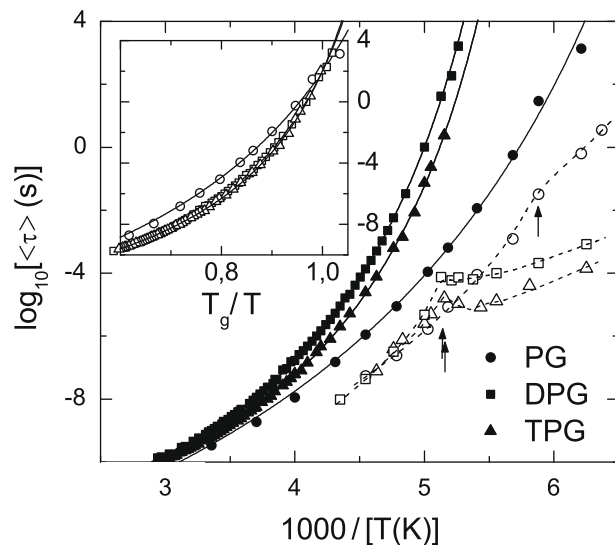

Fig. 2. Relaxation times determined from the fits shown in Fig. 1. The solid lines are fits of the α-relaxation times (closed symbols) with a VFT function. The open symbols correspond to the β-relaxation times. The dashed lines are guides to the eyes. The arrows indicate T_g. The inset shows an Angell-plot of the α-relaxation times.

frequencies with increasing temperature as found also in other glass formers [5,7–10].

3.1. The α-relaxation

To obtain information on the characteristic parameters of the α- and the β-relaxations, least-square fits of the experimental data were simultaneously performed for the imaginary *and* the real part (not shown) of the dielectric permittivity. For parameterization of the spectra at frequencies below the minimum region, a sum of the commonly employed Cole–Davidson (CD) [38] and Cole–Cole (CC) functions [39] was used. The former characterizes the α-relaxation and the latter the slow β-processes at temperatures below 220 K in PG and TPG and 230 K in DPG. It should be mentioned that the justification for using an additive superposition ansatz may be doubted and alternatives were promoted [40]. However, for a first parameterization such an ansatz works well and usually the convolution approach proposed, e.g., in [40] leads to very similar results (see, e.g., [6]). It should be noted that in the present work the use of the term 'β-relaxation' for the secondary relaxations in all three glycols has only practical reasons; in fact there is reason to believe that they are not due to the same microscopic mechanism (cf. Section 3.2). For PG, here we adopt the picture that the excess wing is due to a β-relaxation [2,3].

The fits yield the relaxation time τ_{CD} and the width parameter β_{CD} of the CD function, from which the average relaxation time $<\tau_\alpha> = \tau_{CD}\beta_{CD}$ can be calculated [5]. Its temperature dependence is shown in Fig. 2 (closed symbols). The phenomenological Vogel–Fulcher–Tammann function, $<\tau_\alpha> = \tau_0 \exp[DT_{VF}/(T-T_{VF})]$, well accounts for the observed non-Arrhenius behavior, typical for glassy dynamics. T_{VF} is the Vogel–Fulcher temperature, D the strength parameter [41] and τ_0 a prefactor. Resulting fit parameters are given in Table 1. They all agree reasonably well with the values reported in [24,25,31].

The fragility parameter m, defined as the slope at T_g in the Angell-plot [42,43] (inset of Fig. 2), gives evidence for intermediate fragility, increasing with increasing molecular size from $m = 48$ to 74. León et al. reported similar values ($m = 53$, 64 and 71 for PG, DPG and TPG, respectively) [31]. In [44] the fragility was related
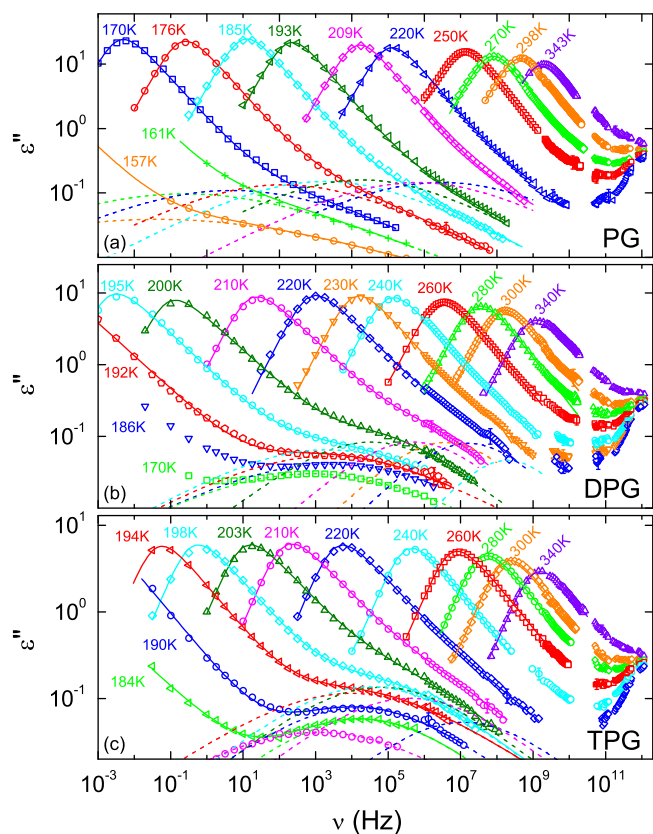

Fig. 1. Frequency-dependent dielectric loss of (a) PG, (b) DPG and (c) TPG for selected temperatures. The solid lines are fits with the sum of a CD and a CC function. (At $T \geqslant 220$ K for PG and TPG and $T \geqslant 230$ K for DPG the CC amplitude was set to zero.) The dashed lines show the CC parts of the fits. Data on PG are partly published in Ref. [3]. The errors are of similar magnitude as the symbol size, except where explicitly indicated.

Table 1

Relaxation parameters of the investigated glycols. In addition, the glass temperatures determined by DSC and from the condition $\tau_\alpha(T_g) = 100$ s are provided. For definition of parameters see text.

	PG	DPG	TPG
τ_0 (fs)	14	25	53
T_{VF} (K)	115	150	150
D	17	10	9
m	48	69	74
T_{CW} (K)	53	105	95
T_g (DSC)	170	196	194
T_g (diel.)	166	189	193

to the form of the potential energy landscape in configuration space. Within this framework, the observed increase in fragility of the glycols may be ascribed to a higher density of minima in the potential energy landscape for the more complex molecules. In [43], the fragility was shown to be related to intermolecular coupling and molecular complexity, which also may explain the observed fragility variation within the series PG, DPG and TPG.

Fig. 2 reveals that, contrary to naive expectations, the relaxation of the larger trimer molecules seems to be faster than for the dimer. As an explanation one may speculate that the relaxation of the trimer already indicates a transition to a polymer-like dynamics where usually segmental motions are the dominant relaxation processes. Also weakening of the intermolecular hydrogen bonding due to extended molecular size could provide an explanation. Interestingly, scaling the relaxation times in the Angell-plot (inset of Fig. 2) makes this puzzling behavior vanish.

The relaxation strength $\Delta\varepsilon_{CD}$ and width parameter β_{CD} of the α-relaxation are shown in Figs. 3(a) and (c), respectively. At high temperatures a saturation of β_{CD} below unity is observed for all three glycols. Such behavior was also found in other molecular glass formers [5,6]. $\Delta\varepsilon_{CD}$ increases with decreasing temperature and molecular size (Fig. 3(a)). Its temperature dependence can be best parameterized by a Curie–Weiss law, $\Delta\varepsilon \sim 1/(T-T_{CW})$ (lines). For DPG and TPG some deviations show up when approaching 200 K. This can be ascribed to the fact that the glass-transition temperatures for both compounds are in this region (cf. Table 1) and, thus, the sample falls out of thermodynamic equilibrium. A temperature dependence of the relaxation strength stronger than a simple Curie law ($\Delta\varepsilon \sim 1/T$) is often observed in glass-forming matter [9,45] and may point to increasingly cooperative relaxation for decreasing temperature.
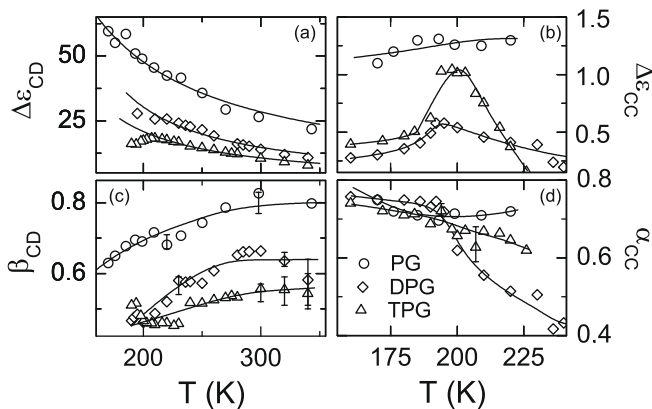
3.2. The secondary relaxation

The secondary relaxations of the glycols were extensively investigated in various earlier publications (e.g., [3,25,28]) and, thus, here we provide only a brief discussion. In [3], the secondary relaxation of PG was classified as a genuine JG relaxation [1], whereas the secondary peaks in DPG and TPG where identified as non-JG [25,28,30], according to criteria given in Ref. [46]. JG β-relaxations are assumed to be inherent to the glassy state and seem to show some properties that are common to all glass formers. In contrast, non-JG secondary relaxations are regarded as material specific, often ascribable to intramolecular modes, and of no deeper interest for the understanding of the glass transition. In the di- and trimer, high pressure experiments revealed an additional third relaxation process [25,28,30]. It is located in the frequency range between the α- and the β-process and was identified as the JG process [28,30]. Thus the β-processes observed in the present work seem to be of JG type for PG and of intramolecular type in DPG and TPG.

The β-relaxation times obtained from the fits in Fig. 1 are shown in Fig. 2 (open symbols). For many glass formers, in non-equilibrium at $T < T_g$ an Arrhenius behavior of the β-relaxation times is well established. However, for $T > T_g$, data analysis is hampered by strong merging of both processes and, thus, the behavior in this region is not finally clarified [47]. We show here the results of our additive fits, which we believe to at least provide a rough estimate of the behavior at $T > T_g$. Above T_g (indicated by the arrows), $\tau_\beta(T)$ shows a transition into a stronger temperature dependence than at $T < T_g$. Similar behavior was also found in various other glass formers [3,6,48]. It is interesting that the β-dynamics for all three glycols approaches each other with increasing temperature. At first glance, this seems to contradict a different nature of the secondary relaxations in PG and in DPG/TPG [25,28,30]. However, this finding seems to be just accidental as in [30] these relaxations were found to exhibit clearly distinct pressure dependences. In the sub-T_g region, a minimum in $\tau_\beta(T)$ is observed for DPG and TPG (Fig. 3). Grzybowska et al. [29,34] described this minimum in the framework of the minimal model [33] and a non-monotonic relaxation kinetic model [35]. In addition, in a recent work this phenomenon has been explained by the hybridization of the secondary relaxation with an unresolved JG β-relaxation at lower frequencies [36].

The width parameters α_{CC} and the relaxation strengths $\Delta\varepsilon_{CC}$ of the β-relaxations are provided in Figs. 3(b) and (d). In all three materials $\alpha_{CC}(T)$ tends to decrease, i.e., the β-peaks narrow with increasing temperature, a behavior typical for secondary relaxations (see, e.g., [6,49]). The di- and trimer both show a peak in $\Delta\varepsilon_{CC}(T)$, located close to T_g (Fig. 3(b)). An increase of $\Delta\varepsilon$ below T_g is predicted by the minimal model [33]. It is not clear if the decrease above T_g is an artifact of merging.

3.3. The fast β-relaxation

Glassy dynamics in the high-frequency region above GHz has found much interest during recent years. In the glycols (Fig. 4) and other glass formers [5,7,8,10], here a characteristic shallow loss minimum occurs. The interest in this region was mainly triggered by the MCT, whose main outcome is the prediction of non-trivial additional contributions to the susceptibility in the GHz–THz region. For a model-free check of such an excess intensity, the insets of Fig. 4 show curves which were calculated by the sum of a two power laws (dashed lines). The high-frequency power law was assumed to have an exponent of at least one as the increase towards the boson peak, which will appear beyond the investigated frequency region [5], should be steeper than linear [10,50]. Clearly, this ansatz is not sufficient for the description of the data and, thus, there is evidence for excess intensity in the minimum region.

Fig. 3. Relaxation strengths of the α- (a) and the β-processes (b) as obtained from the fits of the dielectric loss spectra shown in Fig. 1. Solid lines in (a) are fits using the Curie–Weiss law (c and d). Show the width parameters for α- (c) and β-relaxations (d). Lines in (b–d) are drawn to guide the eyes.
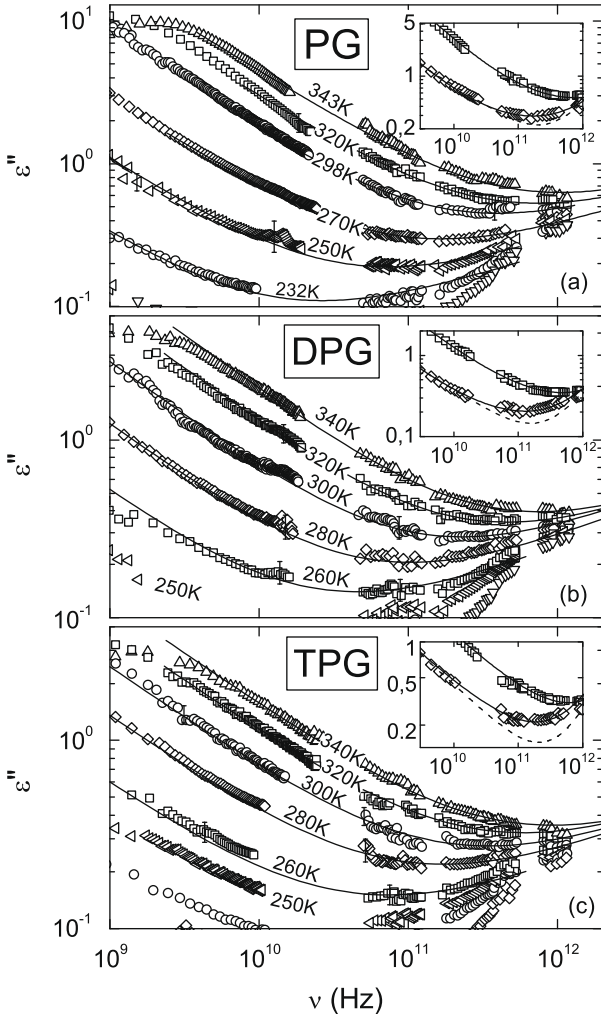
Fig. 4. $\varepsilon''(\nu)$ of PG (a), DPG (b) and TPG (c) at high frequencies for various temperatures. The lines are fits with MCT, Eqs. (2) and (3). The insets show spectra for two temperatures, fitted by Eq. (1) (solid lines). In addition, curves calculated by the sum of two power laws are shown (dashed lines, see text).

One possibility for the description of this excess contribution is provided by the assumption of a constant loss ε_c. Using an additive superposition of the above-mentioned two power laws with exponents $b < 1$ and $n \geqslant 1$, one arrives at

$$\varepsilon'' = c_b \nu^{-b} + \varepsilon_c + c_n \nu^n \tag{1}$$

As also found in other glass formers [7,8,51], for each glycol the minimum region of $\varepsilon''(\nu)$ can be reasonably well described by Eq. (1). This is shown for two temperatures by the fit curves included in the insets of Fig. 4 (solid lines). The presence of a constant loss in glassy matter has been considered since long (e.g., [14]) and quite recently found renewed interest due to its incorporation into the extended coupling model [12,13].

In the following, we will provide a preliminary analysis of our data in the framework of the idealized MCT; a treatment with the extended F12 model of MCT [52] will be published in a forthcoming paper. Within idealized MCT, for T above but near the critical temperature T_c, the minimum is approximated by the sum of the von Schweidler law, ν^{-b}, and the critical law, ν^a [11]:

$$\varepsilon'' = \frac{\varepsilon_{\min}}{a+b}\left[a\left(\frac{\nu}{\nu_{\min}}\right)^{-b} + b\left(\frac{\nu}{\nu_{\min}}\right)^{a}\right] \tag{2}$$

Here $\nu_{\min}$ and $\varepsilon_{\min}$ denote position and amplitude of the minimum. The temperature-independent exponents a and b are constrained by the system parameter

$$\lambda = \frac{\Gamma^2(1-a)}{\Gamma(1-2a)} = \frac{\Gamma^2(1+b)}{\Gamma(1+2b)} \tag{3}$$

where Γ denotes the Gamma function. The fits with Eq. (2) shown in Fig. 4 provide a good description of the experimental data over 2–3 frequency decades. The agreement is at least as good as in propylene carbonate or glycerol [5,10]. At low temperatures, especially in PG some deviations of data and fits occur in the region of the critical law, similar to the findings in glycerol [5]. In the latter compound they were ascribed to the additional increase towards the boson peak, not covered by MCT.

The critical temperature should manifest itself in the temperature dependence of the $\varepsilon''(\nu)$-minimum and the α-peak, according to the following three laws of MCT:
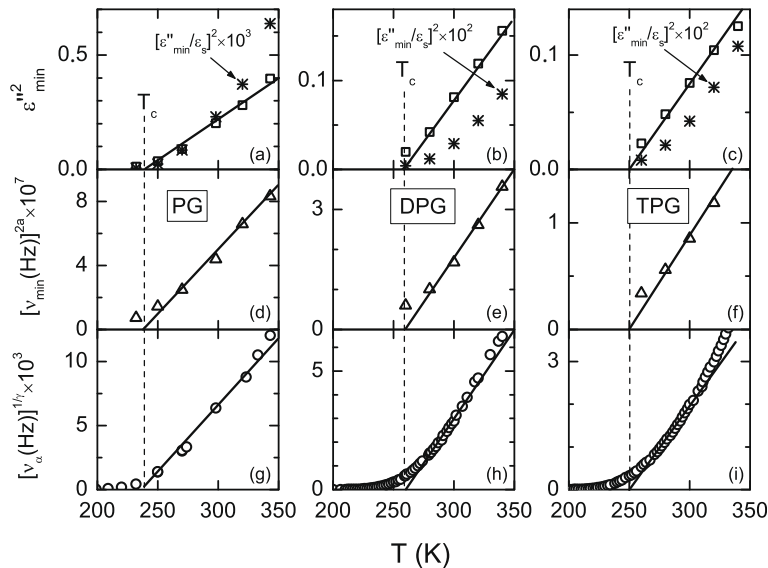

Fig. 5. Critical temperature dependence of the amplitude (a–c) and position (d–f) of the $\varepsilon''(\nu)$ minimum and of the α-relaxation rate (g–i). The lines correspond to the critical laws of MCT, Eqs. (4)–(6). In (a–c), additional results obtained from scaled loss data, as proposed in [53], are included (stars).

$$v_\alpha \propto (T - T_c)^\gamma \tag{4}$$

$$v_{\min} \propto (T - T_c)^{1/2a} \tag{5}$$

$$\varepsilon_{\min} \propto (T - T_c)^{1/2} \tag{6}$$

Here v_α is the relaxation rate $1/(2\pi\tau_\alpha)$. γ is defined as $\gamma = 1/(2a) + 1/(2b)$. $\varepsilon_{\min}$ and $v_{\min}$, taken from the fits with Eq. (2), as well as the α-relaxation rate v_α are shown in Fig. 5 in representations that should lead to straight lines extrapolating to T_c. For all three glycols the complete set of parameters can be consistently described with $T_c \approx 239$ K for PG, $T_c \approx 259$ K for DPG and $T_c \approx 250$ K for TPG (solid lines). When considering these results, one should be aware that the relations (4)–(6) should only hold above but near T_c and are expected to fail too far above T_c. Depending on the exact choice of the extrapolation curve, different critical temperatures can be obtained. For this reason the significance of the results of Fig. 5 should not be overemphasized, although similar evaluations are often used in literature. The parameters resulting from this preliminary analysis are summarized in Table 2. In literature, only for the monomer some inconsistent data were reported with critical temperatures of 198, 314 and 251 K [21,45]. The ratio T_c/T_g decreases from 1.41 to 1.28 with increasing molecular size. Hence it is slightly above 1.2, a value often mentioned in literature for this ratio. Finally it should be noted that in [53] it was proposed to scale the loss data with the static permittivity of the α-process before applying MCT. This leads to a change of the temperature dependence of $\varepsilon_{\min}$ (stars in Fig. 5(a)–(c)). Also these scaled data are consistent with the values of T_c given in Table. 2.

To compare the behavior of the fast β-relaxation of the different glycols, Fig. 6 shows the minimum frequency and the minimum amplitude, related to the α-relaxation strength, vs. the mean α-relaxation time $\langle\tau_\alpha\rangle$. This representation allows for a comparison of the minimum parameters at identical α-relaxation dynamics. The $v_{\min}$ values of all glycols approximately fall on a single line (Fig. 6(a)). However, the relative minimum amplitude and thus the strength of the fast β-relaxation increase with increasing molecule size. This may be ascribed to a better coupling of the dielectric loss to translational modes for the larger molecules. It is well known that the relative amplitude of the fast process is weaker in dielectric spectroscopy, compared to the susceptibilities determined by scattering methods, which more directly couple to density fluctuations [5,7,9,10]. For the spatially more extended DPG and TPG molecules, steric hindrance may rise and thus reorientational motions may be more strongly coupled to translational ones.

4. Summary and conclusion

In the present work, we have provided broadband dielectric spectra covering 15 decades in frequency for three glass-forming propylene glycols, thereby considerably increasing the number of glass formers for which data extending into the region of the fast β-relaxation are available. Besides the α-relaxation, these glycols show all the characteristic high-frequency processes of glassy dynamics that have attracted so much interest in recent years. Of special significance is the finding of clear evidence for a fast β-relaxation also in these materials. It exhibits characteristics, very similar to those in glycerol and propylene carbonate, the only molecular glass formers that so far have been investigated in a similarly broad and continuous frequency range. We have performed a thorough phenomenological characterization of the α- and the slow β-relaxations of these glass-forming glycols. In addition, it was demonstrated that the excess intensity in the region of the loss minimum can be described by a constant loss contribution. Finally, idealized MCT was shown to provide a reasonable description of the fast β-process. From this preliminary analysis, first estimates for the critical temperature and system parameters were obtained. One interesting aspect of this analysis is the increasing strength of the fast β-process with increasing molecular size, which may indicate an increasing coupling of reorientational and translational motion for the larger molecules. An analysis in the framework of the more sophisticated F12-model of MCT [52] will be provided in a forthcoming work.

Table 2
Parameters as derived from MCT analysis. For details see text.

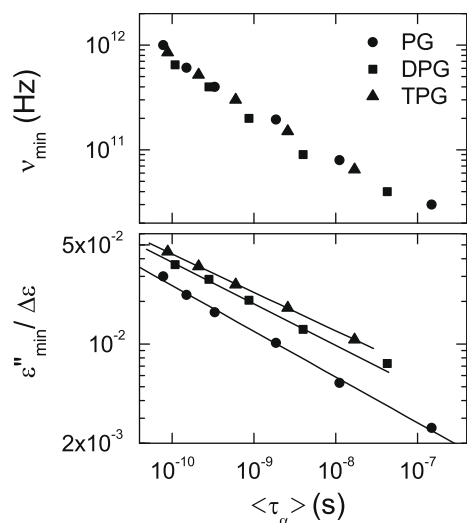|	PG	DPG	TPG
a	0.33	0.319	0.302
b	0.653	0.612	0.55
λ	0.692	0.717	0.7556
γ	2.28	2.38	2.566
T_c (K)	239	259	250
T_c/T_g	1.41	1.32	1.28

Fig. 6. Minimum position of PG, DPG and TPG (a) and minimum amplitude, relative to the α-relaxation strength, (b) vs. α-relaxation time. The lines in (b) are linear fits.

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