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## Fast and slow relaxation processes in glasses

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**Abstract.** We present dielectric relaxation (DS) and light scattering (LS) data of several glass formers. Relaxational features are compiled which are not yet properly taken into account by current models. (i) We distinguish two types of glass formers. Type A systems do not show a slow  $\beta$ -process whereas type B systems do. A full line-shape analysis of  $\varepsilon(\omega)$  is presented ( $10^{-2}$  Hz  $< \nu < 10^9$  Hz). In type A systems the evolution of the high-frequency wing of the  $\alpha$ -process is the most prominent spectral change while cooling and leads to an essentially constant loss at  $T < T_g$ . The analysis of  $\varepsilon(\omega)$  of type B systems is carried out within the Williams–Watts approach and we focus on the temperature dependence of the  $\beta$ -relaxation strength. (ii) Concerning fast relaxations below  $T_g$  as revealed by LS ( $10^9$  Hz  $< \nu < 10^{13}$  Hz) we identify relaxation with a low-frequency power-law behaviour. No indication of a crossover to a white noise spectrum as previously reported and discussed within MCT is found. Analysing this relaxation we recourse to the model of thermally activated transitions in asymmetric double well potentials. We show that the model works well in some cases and the distribution of barrier heights may be extracted, but in other systems pronounced deviations occur.

### 1. Introduction

Supercooling a liquid leads to a pronounced increase of viscosity, and the slow dynamics in such glass formers is well studied by numerous methods. Concerning the evolution of the main relaxation ( $\alpha$ -process), its susceptibility is broadened as compared to a Debye process, and an interpolation by, e.g., a Cole–Davidson function allows one to reproduce its main spectral feature. However, there exist deviations at high frequencies, which have been known for a long time [1], but only recently was this high-frequency wing of the  $\alpha$ -process studied systematically [2–4]. Its nature is discussed controversially. For example, it is not clear whether it is a part of the  $\alpha$ -process [2] or whether it constitutes a separate relaxation process [5]. The high-frequency wing fills the gap between the slow dynamics ( $\alpha$ -process peak) and the fast dynamics ( $\nu > 1$  GHz). With the success of mode coupling theory (MCT) [6], attention has been drawn to details of the fast dynamics. Neutron (NS) [7] and light scattering (LS) experiments [8] demonstrated that in addition to spectral contributions from the microscopic dynamics a non-white noise relaxation spectrum is observed in the GHz regime. The interplay of fast and slow dynamics leads to a susceptibility minimum which can be described by MCT well above the glass transition temperature  $T_g$ .

Here, the question may arise: how does the fast dynamics evolve below  $T_g$ ? Coming from low temperatures, say  $T > 10$  K, the dynamics in glasses are usually described by assuming thermally activated transitions in asymmetric double well potentials (ADWPs) [9], e.g., within

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the Gilroy and Phillips approach (GP) [10]. At even lower temperatures ( $T \ll 10$  K) tunnelling within the ADWPs is believed to be responsible for the low-temperature anomalies of glasses. Thus, we may ask whether there exists a crossover from the fast dynamics observed in the liquid ( $T > T_g$ ) and the fast processes found deep in the glass ( $T \ll T_g$ ). We will focus on this point by presenting results from LS experiments in the frequency and temperature range  $3 \text{ GHz} < \nu < 5 \text{ THz}$  and  $6 \text{ K} < T < 300 \text{ K}$ , respectively.

In addition to these relaxational features many glass formers show a secondary relaxation process ( $\beta$ -process) which is faster than the  $\alpha$ -process but still significantly slower than the fast relaxation described within MCT or the GP model [11]. This  $\beta$ -process persists also below  $T_g$ . At  $T \gg T_g$  the  $\alpha$ - and  $\beta$ -process merge and only a single slow relaxation process survives. Recently, new arguments have been compiled that the  $\beta$ -process is of intermolecular origin, thus confirming the picture presented already by Johari [12–15]. However, besides the question of its origin, several features of the  $\beta$ -process are not well understood. In particular, the relaxation strength varies significantly among the different glass formers and in some cases even no  $\beta$ -process peak is observed at all. Here, we want to promote the idea that indeed two kinds of glass formers exist. One class exhibits a  $\beta$ -process (type B glass former), and the other class does not show discernible  $\beta$ -relaxation (type A glass former) [5, 15].

Thus, three kinds of relaxation process have to be distinguished in simple organic glasses: (i) fast dynamics, (ii)  $\beta$ -process and (iii) some kind of background noise being reminiscent of the high-frequency wing of the  $\alpha$ -process. In this contribution slow dynamics ( $\alpha$ - and  $\beta$ -process) will be analysed by applying dielectric spectroscopy (DS) and fast dynamics by LS.

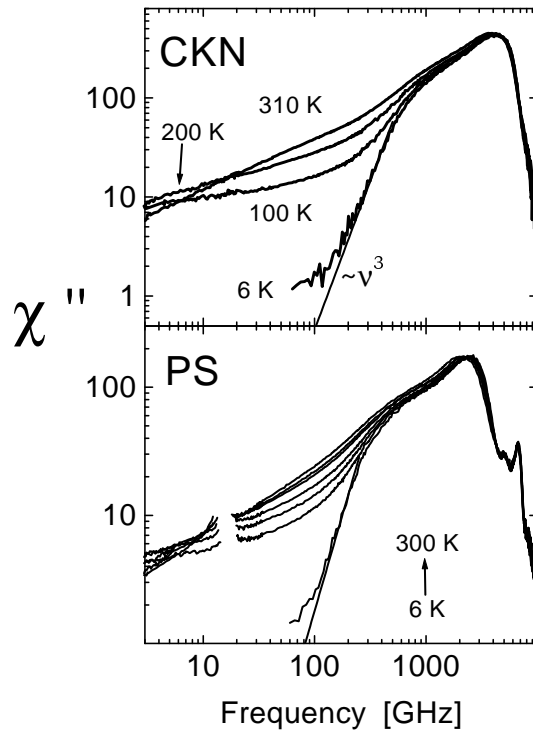
## 2. Results

### 2.1. Fast dynamics studied by light scattering

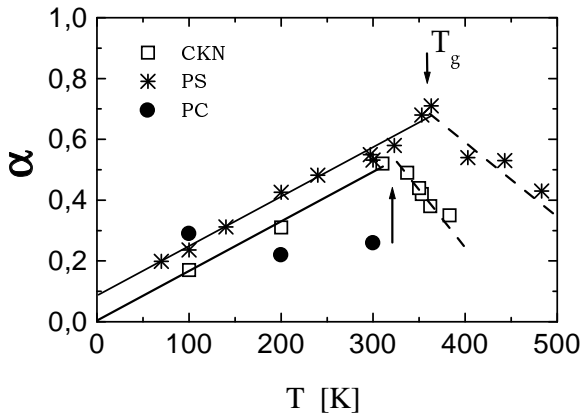
The glass formers calcium potassium nitrate (CKN), polystyrene (PS) and polycarbonate (PC) were measured by tandem Fabry-Pérot interferometry and the data combined with previously reported Raman scattering data [16, 17]. The scattering intensity  $I(\nu)$  was converted to the susceptibility  $\chi''(\nu) = I(\nu)/[n(\nu) + 1]$ , where  $n(\nu)$  is the Bose factor. As an example, the susceptibility spectra of CKN and PS are shown in figure 1. At the lowest temperature (6 K) the spectra are dominated by the boson peak and by the microscopic dynamics spectrum. At higher temperatures a significant quasi-elastic contribution is observed with a low-frequency wing exhibiting a power-law behaviour. The corresponding exponent  $\alpha$  is larger for higher temperature. Explicitly, the temperature dependence of  $\alpha$  is shown in figure 2. In the case of CKN  $\alpha$  increases proportionally with  $T$ , whereas for PS a linear increase with a small deviations from proportionality is found. This temperature dependence extends up to  $T_g$ . Above  $T_g$  an opposite behaviour shows up when recently published data are included [18, 19]: here  $\alpha$  decreases with temperature. Thus, the evolution of  $\chi''(\nu)$  changes by surpassing  $T_g$ , and the models applied above  $T_g$ , e.g., MCT, cannot be expected to work below.

We note that we do not find indications of the so-called knee, i.e. a crossover from power-law behaviour with  $\alpha < 1$  to a white noise spectrum ( $\alpha = 1$ ) at low frequencies as previously reported for CKN [8] and expected from idealized MCT [6]. This experimental difference is due to the fact that we applied additional interference filters in order to suppress the intensity from higher transmission orders. Similar results are reported by Gapinski *et al* [20].

In order to describe the fast dynamics reflected in our LS spectra we recourse to the model of Gilroy and Phillips (GP) [10]. They explained the relaxation in glasses at say  $T > 10$  K, by assuming thermally activated transitions in ADWPs. The model was successfully applied to



**Figure 1.** Light-scattering susceptibility spectra of CKN and polystyrene (6, 70, 100, 140, 200, 240, 300 K).



**Figure 2.** Temperature dependence of the exponent  $\alpha$  characterizing the low-frequency behaviour of the LS susceptibility; data at  $T > 310$  K cf [18] and [19].

interpret acoustic attenuation data [9]. Taking a flat distribution of the asymmetry parameter  $\Delta$ , GP arrived at a simple expression for  $\chi''(\nu)$

$$\chi''(\nu) \propto \int_0^\infty \frac{2\pi\nu\tau}{1 + (2\pi\nu\tau)^2} g(V) dV. \quad (1)$$

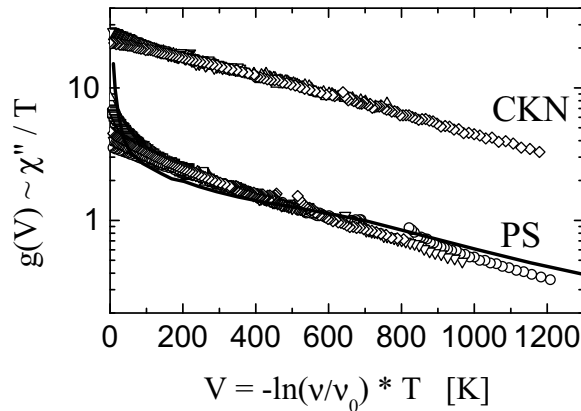
Here  $g(V)$  is the distribution of barrier heights, and the correlation time  $\tau$  at a given site is

connected to  $V$  by an Arrhenius law, explicitly  $\ln(\tau/\tau_0) = V/RT$ . In this approximation the asymmetry parameter  $\Delta$  does not enter in equation (1) any longer. Applying this approach to our data, of course, at high frequencies a crossover to the vibrational contribution is expected, and equation (1) does not hold any longer. The most simple approach to separate relaxational and vibrational contributions is given by subtracting the vibrational contribution as obtained from the respective low-temperature spectrum, where essentially no relaxational contribution is expected. The results obtained do not strongly depend on the actual separation procedure chosen [21]. This holds in particular because, within our approach, we are interested in the high-energy tail of  $g(V)$ . The latter dominates  $\chi''(\nu)$  at low frequencies and there  $\chi''(\nu)$  is not influenced by vibrational contributions.

Restricting ourselves to activated dynamics within in the ADWPs we can go a step further and assume a broad distribution  $g(V)$ . In this case the Debye term in equation (1) can be approximated by a delta function. Then  $\chi''(\nu)$  directly reflects the distribution of correlation times  $G(\ln \tau)$  caused by the distribution  $g(V)$ , and the data  $\chi''(\nu)$  can be scaled by temperature in order to provide a master curve which directly yields  $g(V)$ . Thus, we write [21, 22]

$$g(V) \propto \chi''(\nu)/RT \quad \text{with } V = -RT \ln(2\pi\nu\tau_0). \quad (2)$$

The only parameter needed is the attempt frequency  $\nu_0 = 1/(2\pi\tau_0)$  which we estimate from the peak frequency observed in the spectra after having subtracted the vibrational contribution. We find  $\nu_0 = 150$  GHz for CKN and 180 GHz for PS.

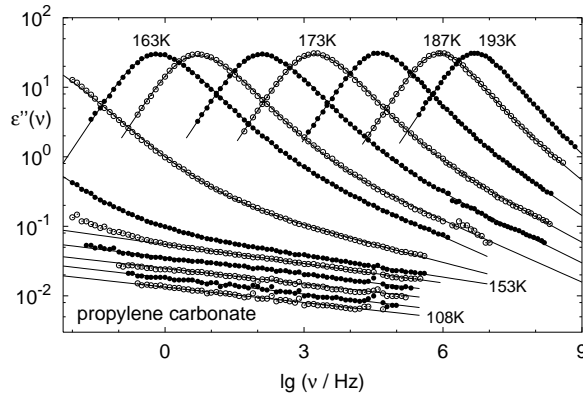


**Figure 3.** Rescaled relaxational spectra providing a master curve which directly reflects the distribution of barriers  $g(V)$ .

The rescaled data are shown in figure 3. Indeed, the data for CKN and PS reveal a master curve. The distribution  $g(V)$  is exponential in good approximation. Within our approximations no cut-off energy can be identified at low energies  $V$ . The typical barrier energies are of the order of some 100 K, i.e. in the range of  $T_g$  [10]. We note that already GP assumed an exponential distribution  $g(V) = V_0^{-1} \exp(-V/V_0)$ , which is very close to the one observed here. By applying equation (1) an exponential  $g(V)$  leads to a power-law distribution  $G(\ln \tau)$  and finally to power-law susceptibility at low frequencies. Then the exponent is given by  $\alpha = RT/V_0$  as observed in figure 2. Therefore we conclude that the GP model well describes the fast relaxation in CKN and PS. However, the data for PC cannot be explained in this frame. As displayed in figure 2, the exponent does not show any temperature dependence [12].

## 2.2. Slow dynamics studied by dielectric spectroscopy

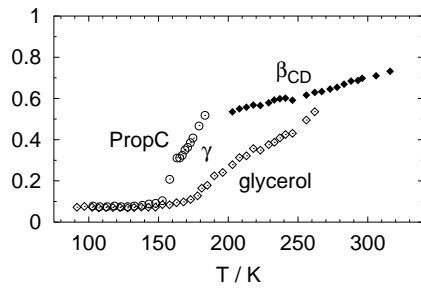
Figure 4 presents the dielectric loss  $\varepsilon''(\nu)$  of the glass former propylene carbonate (PropC). While cooling the  $\alpha$ -relaxation peak shifts to low frequencies and a high-frequency tail develops which cannot be described by a Cole–Davidson susceptibility, for example. Clearly, no  $\beta$ -process peak is discernible and we call these glass formers type A systems. The additional relaxation contribution can be interpreted as a crossover from a first power-law behaviour with exponent  $\beta_{CD}$  to another with an exponent  $\gamma$  [2, 3, 23–26]. In accordance with [6] we will call the second power law the von Schweidler law. Recently, we introduced a distribution of correlation times  $G(\ln \tau)$  [15], which allows us to interpolate the data (cf figure 4) and to extract the exponent  $\gamma$ . The temperature dependence of  $\gamma$  is displayed in figure 5. In addition we show the corresponding data for glycerol, and, for comparison, the temperature dependence of  $\beta_{CD}$  in the case of glycerol. Above  $T_g$  an essentially linear increase of  $\gamma$  is observed, whereas at  $T < T_g$  a crossover to a temperature independent value is found. Thus, it appears that below  $T_g$  the von Schweidler law survives covering a frequency range of at least seven decades ( $10^{-2}$  Hz  $< \nu < 10^5$  Hz). Because the exponent is found to be very small ( $\gamma \cong 0.07$  in PropC and glycerol) we have called this behaviour  $1/f$  noise [5, 15]. Similar conclusions were drawn by Nagel and collaborators [26]. Care has to be taken because at  $T < T_g$  the system is non-ergodic and the fluctuation dissipation theorem may not hold. However, Israeloff and Grigera directly probed  $1/f$  noise [27]. Comparing the temperature dependence of  $\gamma$  with that of  $\beta_{CD}$ , the trend is found that at high temperature  $\gamma$  and  $\beta_{CD}$  become equal. Thus, the high-frequency contribution seems to disappear at high temperatures. As discussed [4, 5, 25] this behaviour demonstrates that the scaling procedure introduced by Nagel and collaborators [2] does not hold at all temperatures.



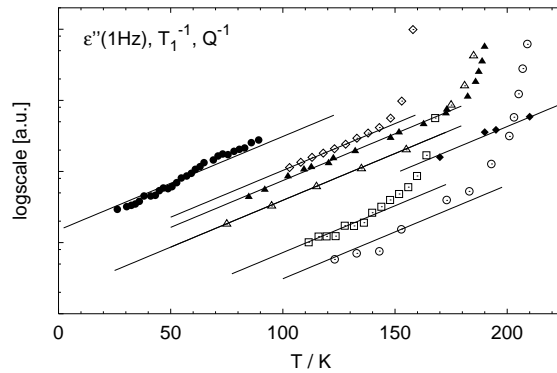
**Figure 4.** Dielectric loss  $\varepsilon''(\nu)$  of propylene carbonate as a function of frequency  $\nu$ . Solid lines:  $T > 150$  K interpolation by applying an appropriate distribution  $G(\ln \tau)$  [15].

The  $1/f$  loss shows a characteristic temperature dependence. In figure 6 we compiled the dielectric loss at  $\nu = 1$  Hz as a function of temperature for the type A glass formers glycerol, PropC, tricesyl phosphate (TCP) and propylene glycol (PG) together with the spin–lattice relaxation rate  $1/T_1$  of glycerol and triphenyl phosphite. The latter probes the spectral density at 55 MHz. In addition, we took mechanical loss data at  $\nu = 10$  kHz for polybutene [28]. In all cases a very similar temperature dependence is revealed, which can be described by

$$\varepsilon''(T) \propto \exp(T/T_f) \quad \text{with } T_f \cong 33 \text{ K.} \quad (3)$$



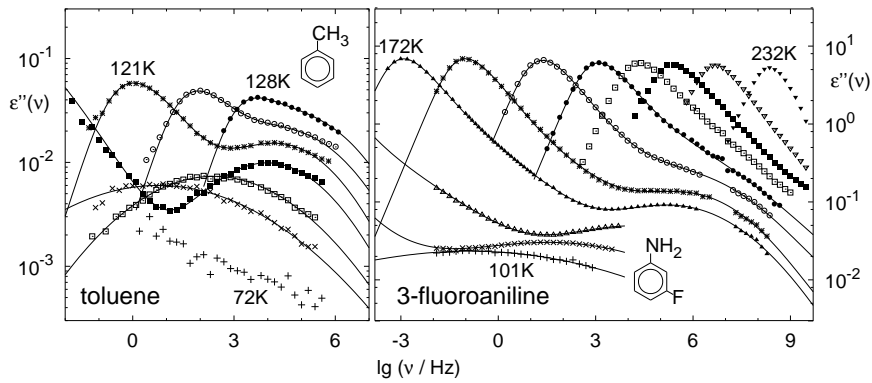
**Figure 5.** Exponents  $\beta_{CD}$  and  $\gamma$  characterizing the  $\alpha$ -process susceptibility at high-frequencies ( $\nu \gg \nu_{peak}$ ) as a function of temperature  $T$  for glycerol (open diamonds:  $\gamma$ , solid diamonds:  $\beta_{CD}$ ) and propylene carbonate (PropC; open circles:  $\gamma$ ).



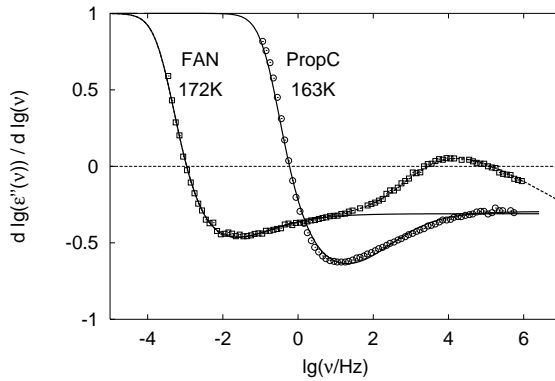
**Figure 6.** Comparison of the temperature dependence of the spin–lattice relaxation rate  $1/T_1$ , the dielectric loss  $\epsilon''$  and internal friction  $Q^{-1}$  data below  $T_g$  (vertical scale in arbitrary units):  $1/T_1$  ( $^2\text{H}$ ) for glycerol (full triangles),  $1/T_1$  ( $^{31}\text{P}$ ) for triphenyl phosphite (solid diamonds) and  $Q^{-1}$  at 10 kHz of polybutene (solid circles, [28]);  $\epsilon''(\nu = 1 \text{ Hz})$  for the glass formers propylene carbonate (open diamonds), glycerol (open triangles), propylene glycol (open squares) and tricesyl phosphate (open circles); solid lines represent an interpolation according to equation (3).

Similar behaviour was reported by Hansen and Richert [29]. Although different techniques probe different fluctuations it appears that the temperature dependence in type A glasses is similar over almost 11 decades in frequency. Of course, the results reported here need to be verified for further systems, and the origin of the constant loss is not understood. Here we note that recent NMR studies demonstrate that it can be regarded as a small amplitude process, which proceeds by the  $\alpha$ -process [5]. NMR [5] and dielectric relaxation studies [30] indicate that the dielectric loss at  $\nu > 10^5 \text{ Hz}$  exhibits some downward curvature. This may be interpreted by assuming an extremely broad  $\beta$ -process (cf below) [30] or it may indicate a cut-off of the von Schweidler law at high frequencies. Such a cut-off is discussed within extended MCT [31] and it constitutes the low-frequency knee which appears at  $T < T_c$  in addition to the high-frequency knee discussed above.

Next we turn to type B glass formers which exhibit a discernible  $\beta$ -relaxation peak. As seen in figure 7 toluene and fluoroaniline (FAN) are examples of this type. Both systems are dielectrically rigid, and a pronounced  $\beta$ -process is observed below as well above  $T_g$ . Hence, an intermolecular origin has to be assumed for the  $\beta$ -process as already promoted by Johari



**Figure 7.** Dielectric loss  $\varepsilon''(\nu)$  of toluene and 3-fluoroaniline; solid lines are fits by applying the Williams–Watts approach.



**Figure 8.** Derivative  $d \lg \varepsilon''(\nu) / d \lg \nu$  of PropC (open circles) as an example of a type A glass former and of fluoroaniline (FAN, open squares) as an example of a type B glass former; solid line: fit by  $G(\ln \tau)$  [15] as in figure 4; dashed line: complete convolution fit applying equation (4).

and Goldstein [11, 12]. The relative contribution of the  $\beta$ -process with respect to that of the  $\alpha$ -process increases significantly above  $T_g$ . In the case of FAN the merging of  $\alpha$ - and  $\beta$ -processes is observed at high temperatures.

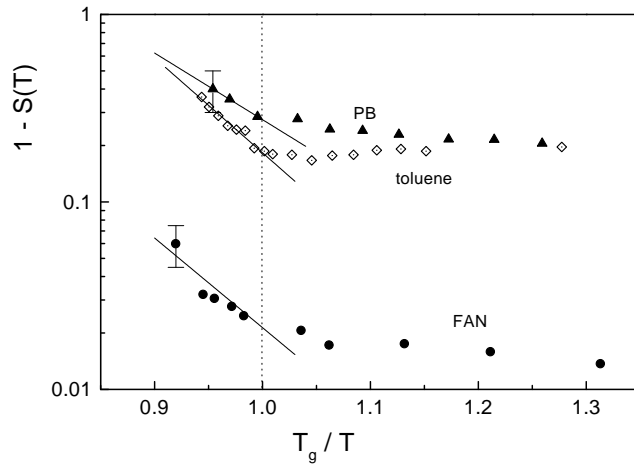
For carrying out a line-shape analysis of  $\varepsilon(\omega)$  including both  $\alpha$ - and  $\beta$ -processes we recourse to the Williams–Watts (WW) approach [13–15, 32–34]. There, both processes are taken to be statistically independent, and the  $\beta$ -process is assumed to relax only a part  $(1 - S)$  of the total polarization. Accordingly, we write for the normalized correlation function

$$\phi(t) = [(1 - S)\phi_\beta(t) + S]\phi_\alpha(t). \quad (4)$$

$\phi_\alpha(t)$  and  $\phi_\beta(t)$  are the normalized correlation functions of  $\alpha$ - and  $\beta$ -processes, respectively. The  $\beta$ -process is regarded as a small-amplitude reorientational process in which all molecules participate. Hence, the glass is assumed to be structurally homogeneous in the sense that no ‘islands of mobility’ exist [12, 35, 36].

The question rises of how to describe the  $\alpha$ -process in systems where the  $\beta$ -process is present. Figure 8 shows the derivative  $d \lg \varepsilon''(\nu) / d \lg \nu$  for FAN and PropC, i.e. the apparent





**Figure 9.** Relaxation strength of the  $\beta$ -process ( $1 - S(T)$ ) plotted against  $T_g/T$ : polybutadiene (PB, full triangles), toluene (open diamonds) and fluoroaniline (FAN, full circles); solid lines: guide for the eye.

exponent of  $\varepsilon''(\nu)$ . Concerning the high-frequency behaviour of  $\varepsilon''(\nu)$  at  $\nu \gg \nu_{peak}$  a crossover to a second power law can be observed in both systems. In the case of FAN deviations appear at highest frequencies due to the presence of the  $\beta$ -process. From these findings we conclude that the high-frequency wing is also found in type B glass formers and has to be taken into account when the  $\beta$ -process is analysed. Already, Haddad and Goldstein [37], Johari [38] and recently Hansen and Richert [29] presented indications for a ‘background loss’ presumably present in all glasses. We assume that at  $T < T_g$  it can be taken into account by choosing a power-law susceptibility with an exponent  $\gamma$  being a fit parameter.

The line-shape of the  $\beta$ -process is well described by assuming a Gaussian distribution of activation enthalpies [13–15, 34, 39, 40] and entropies [14]. By applying equation (4) we are able to interpolate the data in the entire frequency range below and also some degrees above  $T_g$  (cf figure 7) [15]. The mean logarithmic correlation time  $\langle \lg \tau_\beta \rangle = -\lg \nu_{\beta-peak}$  [13, 15] obtained is well fitted by an Arrhenius law with a mean activation enthalpy  $\langle \Delta H_\beta \rangle$  following the relationship  $\langle \Delta H_\beta \rangle \approx 24 RT_g$  [13]. Although some deviations from this behaviour are found we think it can be taken as a further hint that the  $\beta$ -process is an intrinsic feature of glasses.

We want to focus on the temperature dependence of the contribution of the  $\beta$ -process in terms of its relaxation strength ( $1 - S$ ). In figure 9 ( $1 - S$ ) is plotted as a function of  $T_g/T$ . In all cases a significant increase is observed at  $T > T_g$  whereas at  $T < T_g$  the  $\beta$ -process contribution is virtually temperature independent. Similar results were reported by others [34, 41]. However, the level of ( $1 - S$ ) is different in the various systems. We interpret ( $1 - S$ ) as measure of the spatial restriction of the reorientational motion involved in the  $\beta$ -process. As ( $1 - S$ ) increases above  $T_g$ , we conclude that the degree of spatial restriction is released upon heating. This interpretation offers a solution to the controversy of Richter *et al* [34] and Spiess [42] concerning the extent of spatial hindrance of the  $\beta$ -process. At  $T < T_g$  the reorientational angles are small, whereas at  $T > T_g$  the angles become large and can be detected by NS. Finally we note that the temperature dependence of ( $1 - S$ ) displayed in figure 9 may be interpreted in such a way that it is possible to decrease ( $1 - S$ ) at  $T < T_g$  by annealing the glass. Indeed this behaviour was observed by Johari [38] and recently by Olsen [30].

### 3. Discussion and conclusions

Combining results obtained from LS and from DS we cover the dynamic susceptibility in organic glasses ( $T < T_g$ ) in the range  $10^{-2}$  Hz  $< \nu < 10^{12}$  Hz. Three well distinguished relaxational processes can be identified, namely (i) fast dynamics described in some cases by thermally activated dynamics in ADWPs. To our knowledge our LS study presents the first survey of fast dynamics well below  $T_g$  extending down to frequencies  $\nu \cong 1$  GHz, and hence allows us to verify models for the fast dynamics. (ii) The  $\beta$ -process is found in many but not all glass formers. Its spectra reveal some universal properties; however, the relaxation strength varies significantly among the different systems. (iii) A background loss is observed, which extends over many decades.

In all cases small-amplitude motion has to be assumed at  $T < T_g$ ; however the corresponding barrier energies are significantly different. Whereas in the case of the fast dynamics typical barrier energies are on the order of some 100 K, the corresponding energies of the  $\beta$ -process are on the order of some 1000 K.

Clearly, the glassy state of simple organic systems provides a great variety of relaxational features which we are only beginning to understand and ascribing their physical origin remains a great challenge for the future.

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