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# Excess wing in the dielectric loss of glass formers: further evidence for a $\beta$ -relaxation

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## 1. Introduction

Dielectric spectroscopy, with its extremely broad dynamic range accessible, has proven an important tool for the investigation of the dynamic response of glass-forming materials. Aside of the  $\alpha$ -relaxation, broadband dielectric spectra reveal a rich variety of additional dynamic processes, whose explanation is commonly felt as prerequisite to understand the glass transition and glass state in general [1]. Fig. 1 represents a schematic plot of broadband loss spectra demonstrating the most common contributions for two typical cases of glass-forming materials [1,2]. In many glass formers, in addition to the dominating peak cor-

responding to the structural  $\alpha$ -process, a slow  $\beta$ -relaxation is observed as indicated in Fig. 1(b). While in some materials, especially polymers, an explanation in terms of intramolecular degrees of freedom is possible, Johari and Goldstein (JG) [3,4] demonstrated that these processes seem to be inherent to glass-forming materials in general, even if an intramolecular origin can be excluded. However, until now no consensus has been reached concerning the microscopic origin of these so-called JG  $\beta$ -relaxations. At higher frequencies, contributions from a fast process in the minimum region (sometimes termed fast  $\beta$ -process), the boson peak near THz, and microscopic infrared excitations are observed, all of which are not further considered in the present work (see [1,2] for a treatment of fast process and boson peak).

In some glass formers, a second peak or shoulder due to a JG  $\beta$ -relaxation is not observed and instead an excess wing (also called ‘high-frequency

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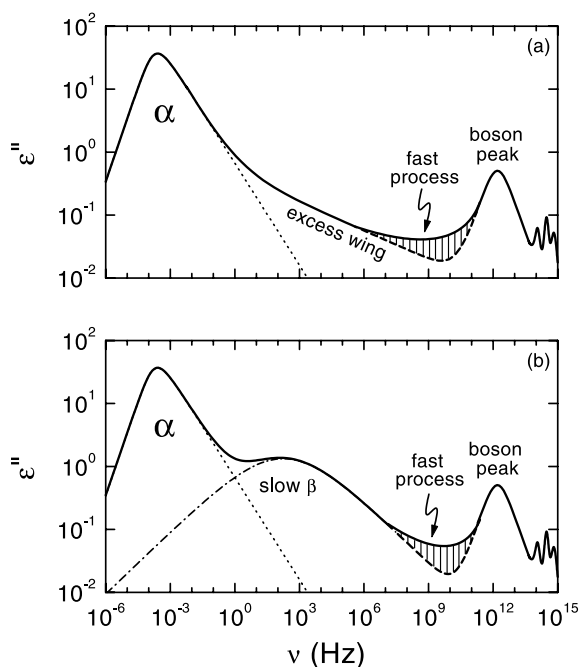


Fig. 1. Schematic view of the frequency-dependent dielectric loss in glass-forming materials as observed in broadband measurements [1,2]. Two typical cases are shown, namely the response of (a) type A[8] glass-formers showing an excess wing and (b) type B glass-formers with a well-resolved  $\beta$ -relaxation peak. Contributions of an additional fast process in the minimum region preceding the boson peak are indicated by the hatched area.

wing<sup>2</sup>) shows up as an excess contribution to the high-frequency flank of the  $\alpha$ -peak (Fig. 1(a)) [1,5–8]. It can be reasonably well described by a second power law,  $\varepsilon'' \sim \nu^{-b}$  with  $b < \beta$  in addition to the power law  $\nu^{-\beta}$ , commonly found above the  $\alpha$ -peak frequency  $\nu_p$  [6,7]. Some intriguing scaling properties of  $\alpha$ -peak and excess wing were found by Nagel and co-workers [5] leading to a collapse of the  $\varepsilon''(\nu)$  curves, including the wing, for different temperatures and materials onto one master curve. This led to the notion that the excess wing is part of the  $\alpha$ -relaxation; however, one has to mention that the universality and accuracy of the so-called Nagel scaling has been controversially discussed during recent years [9–12].

Until recently, it was commonly assumed that excess wing and  $\beta$ -relaxations are different phenomena and the existence of two classes of glass-

formers was proposed – ‘type A’ without a  $\beta$ -process but showing an excess wing and ‘type B’ with a  $\beta$ -process [8,13].<sup>1</sup> However, by inspection of Fig. 1 it is suggestive to assume a  $\beta$ -relaxation to be also present in type A glass formers. In these materials the  $\beta$ -relaxation time  $\tau_\beta$  may be so close to the  $\alpha$ -relaxation time  $\tau_\alpha$  that the main part of the corresponding relaxation peak becomes submerged under the dominating  $\alpha$ -peak and only its high-frequency flank remains visible, thereby appearing as excess wing. This notion, which was considered already in some earlier works [6,15–20], recently was strongly corroborated by the results of dielectric aging experiments below  $T_g$ , performed in our group for two typical type A systems, glycerol and propylene carbonate (PC) [21]. These low-temperature measurements were motivated by the well-established fact that the  $\alpha$ - and  $\beta$ -peak frequencies separate successively when the temperature is lowered. However, in glass formers without a well-resolved  $\beta$ -peak, very low temperatures, below  $T_g$ , may be necessary to observe this separation. But below  $T_g$ , aging effects start to play a role. Aging occurs when the sample has fallen out of thermodynamic equilibrium, i.e. simply spoken, when after cooling the molecules move so slowly that they do not reach a new equilibrium position during reasonable observation times. Therefore in those materials, only after very long aging times below  $T_g$  the  $\alpha$ -peak has shifted to sufficiently low frequencies for a  $\beta$ -peak to become detectable. In the framework of this picture, the above-mentioned two classes of glass formers may be marked just by a different temperature evolution of the  $\beta$ -dynamics: In type B materials,  $\tau_\beta(T)$  has a much weaker temperature dependence than  $\tau_\alpha(T)$ , leading to a clear separation of  $\alpha$ - and  $\beta$ -peak at low temperatures. In contrast, in type A materials  $\tau_\beta$  may more closely follow  $\tau_\alpha$  and only the high-frequency flank of the  $\beta$ -peak – the excess wing – is visible. In the present work we

<sup>1</sup> These terms should not be confused with those introduced earlier in the discussion of dielectric properties of polymers [14]. While this classification may be somewhat oversimplifying as there certainly are examples that cannot be clearly assigned as types A or B, it is useful to quickly denote systems with clear excess wing or well pronounced  $\beta$ -relaxation.

summarize our aging results, which since the original work [21] have been extended to propylene glycol (PG) and Salol, giving further evidence for a  $\beta$ -relaxation as the origin of the so-far mysterious excess wing of glass-forming liquids.

## 2. Experimental details

For the measurements, parallel-plane capacitors made from stainless steel with an empty capacitance up to 100 pF were used. High-precision measurements of the dielectric permittivity in the frequency range  $100 \mu\text{Hz} \leq \nu \leq 1 \text{ MHz}$  were performed by frequency response analysis, employing the Novocontrol alpha-analyzer. It allows for the detection of values of  $\tan \delta$  as low as  $10^{-4}$ . The response of the empty capacitor was measured and in the most relevant region of  $10 \text{ Hz} \leq \nu \leq 100 \text{ kHz}$  the corresponding loss was found to be smaller than  $1.5 \times 10^{-4}$ . For PC and glycerol additional frequency sweeps at  $20 \text{ Hz} \leq \nu \leq 1 \text{ MHz}$  were performed with the autobalance bridges Hewlett-Packard HP4284 and HP4285. For more details the reader is referred to [22,23].

To keep the samples at a fixed temperature for up to five weeks of aging time, a closed-cycle refrigerator system was used. The samples were cooled from a temperature about 20 K above  $T_g$  with the maximum possible cooling rate of about 3 K/min. The final temperature was reached without any temperature undershoot. As zero point of the aging times reported below, we took the time when the desired temperature was reached, about 200 s after passing  $T_g$ . For PC, PG, and glycerol during the complete aging process the time-dependent development of the loss spectra was monitored by performing frequency sweeps at regular time intervals. For Salol, due to technical problems, at times  $10^3 < t < 10^5 \text{ s}$  no spectra could be collected. For short aging times a reduced number of frequencies was used to enable sweeps fast enough to avoid significant aging during the sweep time. The temperature was detected by a platinum sensor, inserted into one capacitor plate and kept stable within 0.02 K during the measurement time of up to five weeks. In general the maximum aging times were limited by technical problems, e.g.

power failures or breakdowns of cooling water for the compressor of the refrigerator system, which become quite likely for long waiting times. Power supplies with battery backup and cooling-water independent refrigeration are currently being set up, aiming at aging times of up to one year.

## 3. Results

In Fig. 2 the dielectric loss,  $\varepsilon''(\nu)$ , of PC ( $T_g \approx 159 \text{ K}$ ) is shown for various temperatures [1,21,24]. The  $\alpha$ -peaks have been fitted with the empirical Cole–Davidson (CD) function [25], which provides a good description up to two decades of frequency above  $\nu_p$  (dashed lines). Alternatively also the Fourier transform of the Kohlrausch–Williams–Watts (KWW) function [26] can be used. At higher frequencies deviations from the CD function are revealed and at  $T \leq 193 \text{ K}$  a typical excess wing develops, showing up as a second power law before the minimum is reached. The spectra at 155 and 152 K, were measured after sufficiently long aging times to ensure that thermal equilibrium is reached. Especially, at 152 K the sample had to be allowed to age over five weeks to achieve this goal.  $\varepsilon''(\nu)$  at 152 K clearly shows a shoulder, i.e. a downward curvature, thus indicating the presence of a  $\beta$ -relaxation, which can be presumed to cause the excess wing at higher temperature. The solid line in Fig. 2 demonstrates how the 152 K curve can be described by an additive superposition of  $\alpha$ -peak (dashed line) and  $\beta$ -peak (dash-dotted line). For the  $\beta$ -peak the Cole–Cole function [27], able to fit the  $\beta$ -peaks in most type B glass-formers, was used. Similar descriptions are possible also for the results at higher temperatures [20,28]. In Fig. 3 new results for glass-forming Salol ( $T_g \approx 218 \text{ K}$ ) are shown. As for PC, this material is known as typical type A glass former [6] and was one of the examples used by Dixon et al. [5] to demonstrate the scaling of  $\alpha$ -peak and excess wing onto one master curve. Indeed with decreasing temperature, a characteristic excess wing is revealed [5,6]. However, the curve at 211.5 K, which was obtained after an aging time of 6.5 days, in place of the excess wing shows a well-pronounced shoulder, again clearly indicative of a  $\beta$ -relaxation.

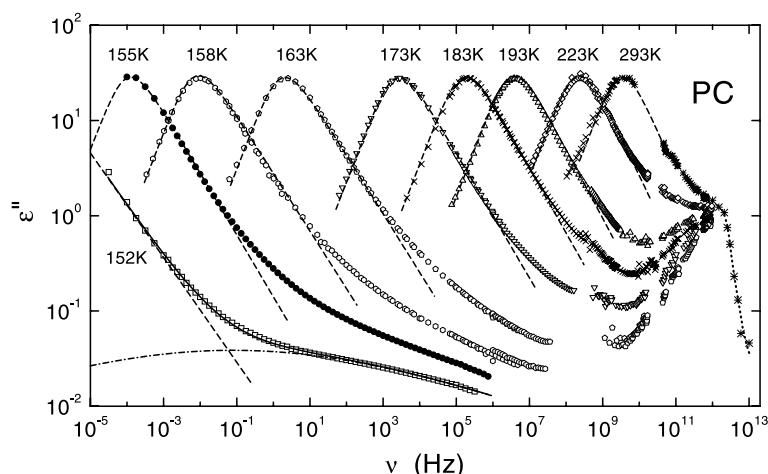


Fig. 2. Frequency-dependent dielectric loss of PC for selected temperatures [1,21,24]. All curves were taken in thermodynamic equilibrium. The dashed lines are fits of the  $\alpha$ -peak region with the CD function. The solid line at 152 K is a fit with the sum of a CD (dashed line) and a CC function (dash-dotted line). The dotted line in the THz region is drawn to guide the eye.

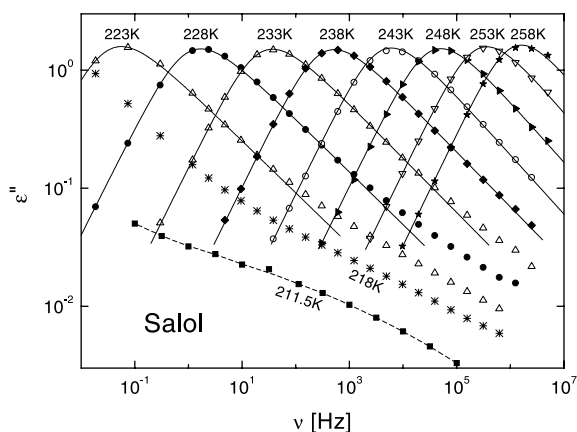


Fig. 3. Frequency-dependent dielectric loss of Salol for various temperatures. All curves were taken in thermodynamic equilibrium. The solid lines are fits of the  $\alpha$ -peak region with the CD function. The dashed line is drawn to guide the eye.

The time-dependent development of the spectra during aging is shown in Fig. 4 for PC [21], glycerol ( $T_g \approx 185$  K) [21], propylene glycol (PG,  $T_g \approx 168$  K) [28], and Salol for the lowest temperatures investigated. For PC and Salol the curves after maximum aging times are also shown in Figs. 2 and 3, respectively. The frequency range of Fig. 4 was chosen to cover the excess wing region only. The  $\alpha$ -peak is located at very low

frequencies and in  $\varepsilon''(\nu)$  is manifested in the somewhat steeper increase towards low frequencies. In Fig. 4 for PC, glycerol, and PG indeed the typical power law, characteristic of an excess wing, shows up for short aging times. However, with increasing time the amplitude of the excess wing diminishes and successively develops into a shoulder. For Salol the shoulder, present already for short times, becomes more pronounced during aging.

Admittedly, especially for glycerol (Fig. 4(b)) the curvature in  $\varepsilon''(\nu)$  is quite subtle. One should look at a small angle from the lower right corner of Fig. 4(b) towards the upper left, to see it more clearly. A more quantitative approach is given in Fig. 5, where the differences of the measured  $\varepsilon''(\nu)$  curves after full aging and the power laws  $c\nu^{-b}$ , represented as dashed lines in Fig. 4, are shown for all materials. Well-pronounced peaks are revealed with an amplitude that lies significantly above the experimental resolution limit.<sup>2</sup> This finding clearly proves the significance of the shoulders observed in the loss spectra. Further evidence has been given in [21] by plotting the derivatives of the

<sup>2</sup> One should note that the peaks in Fig. 5 do not represent the 'true'  $\beta$ -relaxation peaks (cf. the dash-dotted line in Fig. 2) but only the difference to a power law.

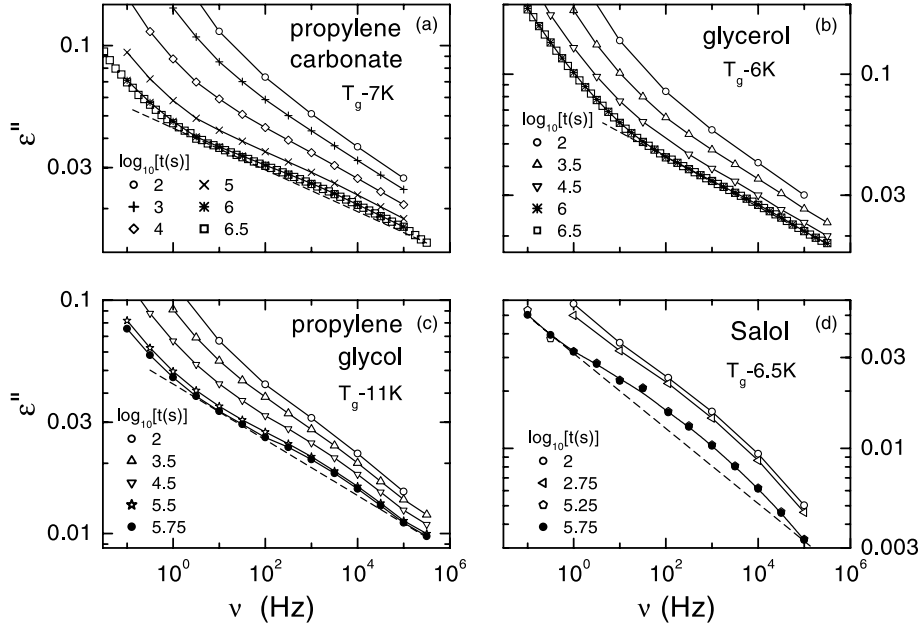


Fig. 4. Frequency-dependent dielectric loss of (a) PC [21], (b) glycerol [21], (c) PG [28], and (d) Salol for different aging times  $t$  (not all shown). The solid lines are drawn to guide the eyes. After maximum aging times of about five weeks, the spectra in PC and glycerol have been collected for an extended number of frequencies. The dashed lines represent power laws to demonstrate the significance of the shoulder after maximum aging (cf. Fig. 5).

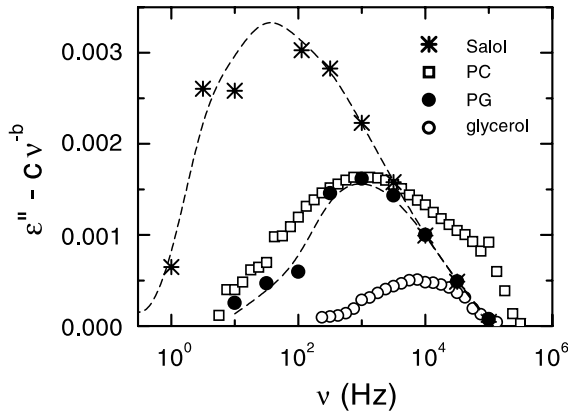


Fig. 5. Difference spectra of the dielectric loss for the longest aging times in Fig. 4(a)–(d) between  $\varepsilon''(\nu)$  and the power laws shown as dashed lines. The lines are drawn to guide the eyes.

$\varepsilon''(\nu)$  curves, where a maximum is observed. Finally, for PC and glycerol the measurements using the autobalance bridges (not shown) agree well with those obtained with the alpha-analyzer and

confirm the appearance of a shoulder after long aging times.

#### 4. Discussion

Using the concept of the fictive temperature [29], the results of Fig. 4 are interpreted in the following way: During aging after rapid cooling to a sub- $T_g$  temperature, thermodynamic equilibrium is successively being recovered. This is accompanied by a continuous decrease of the fictive temperature towards the ‘true’ temperature, leading to a strong shift of the  $\alpha$ -peak towards lower frequencies. It is well known that the relaxation times of  $\beta$ -processes usually exhibit a weaker temperature dependence than those of the  $\alpha$ -process. Therefore the secondary process causing the excess wing is less affected by the change of the fictive temperature during aging. The different aging dependences of  $\alpha$ -process and the  $\beta$ -process, responsible for the wing/shoulder feature, can also

easily be deduced from the fact that the spectra at different aging times cannot be scaled onto each other by a horizontal shift in Fig. 4. In addition, aging experiments reported by JG [3] on type B glass-formers revealed a negligible change of the  $\beta$ -peak frequency with aging time.<sup>3</sup> Therefore, with increasing aging time, respectively decreasing effective temperature, the time scales of  $\alpha$ - and  $\beta$ -process become successively more separated and the  $\beta$ -peak finally shows up in form of the observed shoulder.

The time necessary to reach thermodynamic equilibrium depends delicately on the temperature chosen for the aging experiments, reflecting the strongly increasing  $\alpha$ -relaxation time below  $T_g$  of most glass-formers. In PC and glycerol after an aging time of  $10^{6.5}$  s (about five weeks) the attainment of equilibrium could be stated. This can be clearly inferred from the agreement of the loss spectra for the two highest aging times in Fig. 4(a) and (b). However, in PG (Fig. 4(c)), where the maximum aging time was limited to 6.5 days, thermodynamic equilibrium was not reached. This can be deduced from the fact that the spectra at  $10^{5.5}$  and  $10^{5.75}$  s do not fall onto each other, i.e. there is still a significant time dependence. Currently experiments in PG are in progress aiming at longer aging times. For the collection of the present first results on Salol, the temperature of 211.5 K was chosen to reach equilibrium already after a maximum aging time of  $10^{5.75}$  s (about 6.5 days, Fig. 4(d)).

As discussed in detail in [21], further evidence supporting the explanation of the excess wing by a  $\beta$ -relaxation can be found in literature, both from dielectric experiments [6,11,19,24,30–32], mainly revealing vague indications of shoulders at low temperatures [6,11,24,30,31], and also from other experimental methods [33]. Also our recent results in ethanol corroborate this notion [34]. Here we want to discuss results reported for Salol: In [6] and very clearly in [11], shoulders in the excess wing region were observed at temperatures below  $T_g$  (208 and 204 K, respectively). At these tem-

peratures very long aging times would be necessary to reach equilibrium and both results can be assumed to be taken out of equilibrium. In the study of Wagner and Richert [19], performed on Salol, after quenching to temperatures below  $T_g$  with an extremely high cooling rate, a strong secondary peak appeared, which vanished after heating above  $T_g$ . The origin of this non-equilibrium feature is still a mystery, but it seems questionable if it is due to a ‘usual’  $\beta$ -relaxation, especially as its relaxation time is nearly temperature independent. Also it is unclear if the non-equilibrium secondary relaxations, observed in isochronal measurements of various quenched glass-formers [35] are manifestations of the relaxations causing the excess wing. From all these results the question arises if the indications for a  $\beta$ -relaxation in Salol seen below  $T_g$  in [6,11] may be pure non-equilibrium effects and if this relaxation may completely vanish in equilibrium. Only the present aging experiments could prove that the excess wing in Salol is due to a  $\beta$ -relaxation that is an equilibrium phenomenon. This statement of course is also valid for the other glass-formers investigated.

In Fig. 4, consistent with the findings in [19,35], the amplitudes of the  $\beta$ -peaks (only seen as excess wing at short times) seem to diminish with aging. Interestingly, in some type B systems with well-resolved  $\beta$ -relaxations such a decrease of the  $\beta$ -peak amplitude was reported, too [3,15]. Together with the findings in [19,35] and in light of the fact that  $\beta$ -relaxations are mostly detected below  $T_g$ , one could suspect that the  $\beta$ -relaxation in general is a non-equilibrium phenomenon and may vanish completely provided that (sometimes inaccessibly) long aging times are applied. However, the present results, showing a clear saturation of the time-dependent loss (Fig. 4) [23], clearly speak against this notion.

## 5. Conclusions

From the long-time aging results at  $T < T_g$  of four typical ‘type A’ glass formers shown in the present work we obtain clear evidence for a  $\beta$ -relaxation process causing the excess wing. Below  $T_g$ , after thermodynamic equilibrium is reached,

<sup>3</sup> In [15] even a small upward shift of the  $\beta$ -peak frequency was noted during aging.

$\alpha$ - and  $\beta$ -relaxation are sufficiently far separated to allow the detection of a significant shoulder developing out of the excess wing. Above  $T_g$  the loss-peaks caused by this  $\beta$ -relaxation are deeply merged under the  $\alpha$ -peaks and only their high-frequency flanks show up, leading to the power law known as excess wing.

The equilibrium sub- $T_g$  curves showing a shoulder can be well described with the sum of e.g. a CD and a CC function as demonstrated for PC in Fig. 2 [21,28]. At higher temperatures such a description of course is also possible [20,21,28] as was noted already in earlier works [6,16–18]. So far our results in PC, glycerol, and PG have been analyzed in this way, revealing a  $\beta$ -relaxation time  $\tau_\beta(T)$  that shows significant deviations from Arrhenius behavior [20,28]. Here it should be noted that  $\tau_\beta$  determined in this way certainly has a large uncertainty. For the lowest temperatures, where a shoulder shows up, a relatively precise determination is possible (cf. dash-dotted line in Fig. 2). However, for the higher temperatures there is quite a lot of freedom in the choice of parameters, due to the fact that for the description of the excess wing mainly the right flank of the  $\beta$ -peak is used. Especially the  $\beta$ -peak frequency ( $\sim 1/\tau_\beta$ ) can be almost arbitrarily shifted towards low frequencies, simultaneously increasing the peak amplitude, without considerably diminishing the quality of the fit. However, if the  $\beta$ -peaks are shifted too far towards high frequencies the fits become poor, i.e. it is possible to determine a lower limit of  $\tau_\beta$  at least. Taking this into account, it is not possible to describe the experimental data by fixing the values of  $\tau_\beta(T)$  to an thermally activated Arrhenius behavior.

This clear detection of a non-Arrhenius  $\tau_\beta(T)$  is astonishing as the  $\beta$ -relaxation times, determined in type B glass-formers are believed to follow an Arrhenius behavior. Thus one may have objections to using the term ‘ $\beta$ -relaxation’ for the relaxation causing the excess wing. In fact it could be possible that there are two different kinds of relaxations beyond the  $\alpha$ -relaxation, which both are inherent to glass-forming matter [36]: the JG  $\beta$ -relaxation and the relaxation causing the excess wing, both having different microscopic origins. Depending on amplitude and investigated fre-

quency and temperature range, only one of each may be visible in the spectra, leading to the apparently different behavior of types A and B glass formers. Indeed there are examples of glass formers that have three relaxations or show even an excess wing *and* a ‘ $\beta$ -relaxation’ [8] (in the other scenario this third relaxation would be denoted as  $\gamma$ -relaxation). While it is clear now that the excess wing is indeed caused by a relaxation process, from an experimental point of view it is difficult to decide whether or not this relaxation and the JG  $\beta$ -relaxations observed in type B glass formers originate from the same microscopic process.

However, it should be remarked that the notion of an Arrhenius temperature dependence of  $\tau_\beta(T)$  in type B glass formers to a large extent is based on non-equilibrium results, obtained below  $T_g$ . In addition, there is no principal reason that the  $\beta$ -process dynamics should always show Arrhenius behavior, especially as its microscopic origin can be regarded as unclarified yet. If the excess wing is indeed due to a  $\beta$ -relaxation, the difference between types B and A glass formers may be caused just by the temperature evolution of the  $\beta$ -dynamics: In the first materials it is rather weak, presumably Arrhenius, leading to a clear separation of both relaxation times at low temperatures which thus enables a clear detection of the  $\beta$ -peak. In contrast, in the latter materials the relaxation time of the  $\beta$ -process may more closely follow that of the  $\alpha$ -process (which is non-Arrhenius) and only the high-frequency flank of the  $\beta$ -peak – the excess wing – is visible (except at low temperatures after aging). Such an uncommon temperature dependence of  $\tau_\beta$  was already suspected by JG to explain the apparent absence of a  $\beta$ -relaxation in some glass-formers [3].

In this context we want to mention that the above scenario is consistent with the recently found correlation of  $\tau_\beta(T_g)$  and the Kohlrausch-exponent  $\beta_{KWW}(T_g)$  describing the width of the  $\alpha$ -peak [37]: Type A glass-formers have relatively large  $\beta_{KWW}$  which, within this correlation, implies a close vicinity of  $\tau_\alpha$  and  $\tau_\beta$  – the  $\beta$ -peak becomes sub-merged under the  $\alpha$ -peak. In [37] a possible explanation of this correlation within the framework of the coupling model [38] was proposed and even the Nagel scaling may be explained in this

way [17]. Finally, also a recent extension of the Weiss mean-field theory for finite systems using the model of dynamically correlated domains seems to be able to explain the present spectra, including the excess wing [39]. Interestingly, within this framework the occurrence of an excess wing is attributed to a superposition of peaks or shoulders, which are present in addition to the  $\alpha$ -peak, in good accord with the scenario developed above.

In summary, there is now compelling evidence that the excess wing found in type A glass formers is indeed the high-frequency flank of a second relaxation peak. Most likely it is the manifestation of the JG  $\beta$ -relaxation leading to a well-separated  $\beta$ -peak in type B glass formers, but being merged under the  $\alpha$ -peak for type A glass formers. While our dielectric measurements cannot provide information about the microscopic origin of this  $\beta$ -relaxation, by the present results two so-far mysterious phenomena in glass-forming materials – the excess wing and the  $\beta$ -relaxation have been unified into one. The knowledge of the true nature of the excess wing certainly is a prerequisite for any theoretical explanation of this phenomenon and it may be hoped that these results will further advance our understanding of the supercooled state of matter.

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