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Scaling of broadband dielectric data of glass-forming liquids and plastic crystals

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Abstract. The Nagel scaling and the modified scaling procedure proposed recently by Dendzik *et al.* have been applied to broadband dielectric data on two glass-forming liquids (glycerol and propylene carbonate) and three plastic crystals (ortho-carborane, meta-carborane, and 1-cyano-adamantane). Our data extend the upper limit of the abscissa range to considerably higher values than in previously published analyses. At the highest frequencies investigated, deviations from a single master curve show up which are most pronounced in the Dendzik scaling plot. The loss curves of the plastic crystals do not scale in the Nagel plot, but they fall onto a separate master curve in the Dendzik plot. In addition, we address the question of a possible divergence of the static susceptibility near the Vogel-Fulcher temperature. For this purpose, the low-temperature evolution of the high-frequency wing of the dielectric loss peaks is investigated in detail. No convincing proof for such a divergence can be deduced from the present broadband data.

1 Introduction

Investigations of the dynamic response of glass-forming materials are of fundamental importance to describe glass transition phenomena [1]. Here the α - or structural relaxation is the most commonly investigated dynamic process. In the frequency dependence of the dielectric loss, $\varepsilon''(\nu)$, of dipolar systems the α -peak is the dominating spectral response, strongly slowing down with decreasing temperature. The microscopic origin of the spectral shape of the α -peak, which significantly deviates from the mono-dispersive Debye behaviour, is still unclear in many respects. Usually phenomenological functions as, *e.g.*, the Cole-Davidson (CD) function [2] or the Fourier transform of the Kohlrausch-Williams-Watts (KWW) function [3] are used to describe the α -peak. However, these empirical functions, which extrapolate to a power law, $\varepsilon'' \sim \nu^{-\beta}$, $\beta < 1$, at high frequencies, provide good fits of the experimental data at best up to 3 decades above the peak frequency, only. At higher frequencies an excess wing appears, which can be described by a second power law, $\varepsilon'' \sim \nu^{-b}$, with $b < \beta$ [4–7]. This excess wing was already noted in the early work of Davidson and Cole [2] and is a universal feature of glass-forming liquids [4–8], at least for all systems where a β -relaxation is absent (see below). Recently, we were able to show that this excess wing does not show up in various plastic crystals [9], a class of materials, whose properties resemble those of glass-forming liquids in many respects. In plastic crystals the centres of mass of the molecules form a regular crystalline lattice but

the molecules are dynamically disordered with respect to the orientational degrees of freedom. Due to similarities in the relaxation dynamics, orientationally disordered crystals are often considered as model systems for structural glasses, but are much simpler to be treated in theoretical approaches.

It has to be stated that until now there is no commonly accepted explanation of the microscopic origin of the excess wing in glass-forming liquids, although there are some theoretical approaches that are able to describe the wing at least partly [10]. In addition, it is possible to describe α -peak and wing by a superposition of two relaxation processes [4, 11–13]. A secondary relaxation process, usually termed β -process, was demonstrated to be present in a variety of glass formers by Johari and Goldstein [14]. Also recent theoretical developments within the coupling model [15] may point in the direction of a universal β -relaxation, closely connected to the α -process [16, 12] and it cannot be excluded that the wing is caused by a β -relaxation, whose peak is hidden under the dominating α -peak. Of course, as long as the occurrence of the excess wing and its evolution with temperature remains a mystery, also its absence in plastic crystals cannot be explained.

A very successful description of the wing is given by the so-called Nagel scaling [8, 17]. Nagel and coworkers found that the $\varepsilon''(\nu)$ curves for different temperatures and even for different materials, including the wing, can be scaled onto one master curve by plotting $Y_N := 1/w \log_{10}[\varepsilon'' \nu_p / (\Delta \varepsilon \nu)]$ vs. $X_N := 1/w(1 + 1/w) \log_{10}(\nu/\nu_p)$. Here w denotes the half-width of the loss peak normalized to that of the Debye peak, ν_p is the peak

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frequency, and $\Delta\epsilon$ the relaxation strength. During the past years some criticism of the Nagel scaling arose concerning its universality [18, 19] and accuracy [19, 20]. However, it is still commonly believed that the Nagel scaling is of significance for our understanding of glass-forming liquids and many efforts have been made to check for its validity in a variety of materials [9, 17–23]. Recently a modification of the original scaling procedure was proposed by Dendzik *et al.* [19] and reported to lead to a better scaling, especially at low frequencies. Here $Y_D := \log_{10}[\epsilon''\nu_s/(\epsilon_s\nu)]$ is plotted *vs.* $X_D := (1 + \beta) \log_{10}(\nu/\nu_s)$ with ν_s and ϵ_s read off at the intersection point of the two power laws $\epsilon'' \sim \nu$ and $\epsilon'' \sim \nu^{-\beta}$ below and above ν_p , respectively.

The universal scaling of α -peak and excess wing strongly suggests a correlation of both features. Especially, making certain assumptions, it was shown [24], that the wing exponent b should become zero for a limiting relative half-width, $w^* \approx 2.6$. It was argued that this constant loss behaviour could imply a divergence of the static susceptibility, supporting speculations about an underlying phase transition from the liquid to the glassy state. By a low-temperature extrapolation of $w(T)$ curves of various glass formers, Nagel and coworkers deduced that this constant-loss limit is reached near the Vogel-Fulcher temperature T_{VF} [24]. T_{VF} is determined from fits of the α -relaxation time τ with the Vogel-Fulcher law, $\tau = \tau_0 \exp[DT_{VF}/(T - T_{VF})]$. In addition, it was shown [6] that, if the scaling is valid and assuming $w = 1/\beta$, the exponents β and b are related by

$$\frac{b+1}{\beta+1} = \gamma, \quad (1)$$

where γ should be equal to the high-frequency limiting slope in the Nagel plot, γ' . Indeed, in [6] within the error bars a temperature-independent value $\gamma = 0.72(\pm 0.02)$ was found, in agreement with γ' read off from the master curve of the Nagel scaling. Finally, a “divergent” susceptibility was also deduced directly from an extrapolation of b to low temperatures, which for various glass formers indeed seemed to approach zero near T_{VF} [6].

Our group has made available dielectric loss data on glass-forming liquids and plastic crystals extending over the exceptionally broad dynamic range of up to 19 frequency decades [5, 25–29]. The present work has two objectives: First, to check if scaling still works for these extremely broadband data. Second, to look for evidence for a divergent dielectric susceptibility at low temperatures. We will apply both the original Nagel scaling [8] and the modified procedure proposed by Dendzik *et al.* [19] to broadband dielectric data on the low-molecular weight glass formers glycerol and propylene-carbonate (PC) and on the plastic crystals ortho-carborane (o-CA), meta-carborane (m-CA), and 1-cyano-adamantane (CNA). In addition, these data will be checked for a cross-over of the high-frequency wing to constant loss behaviour at low temperatures.

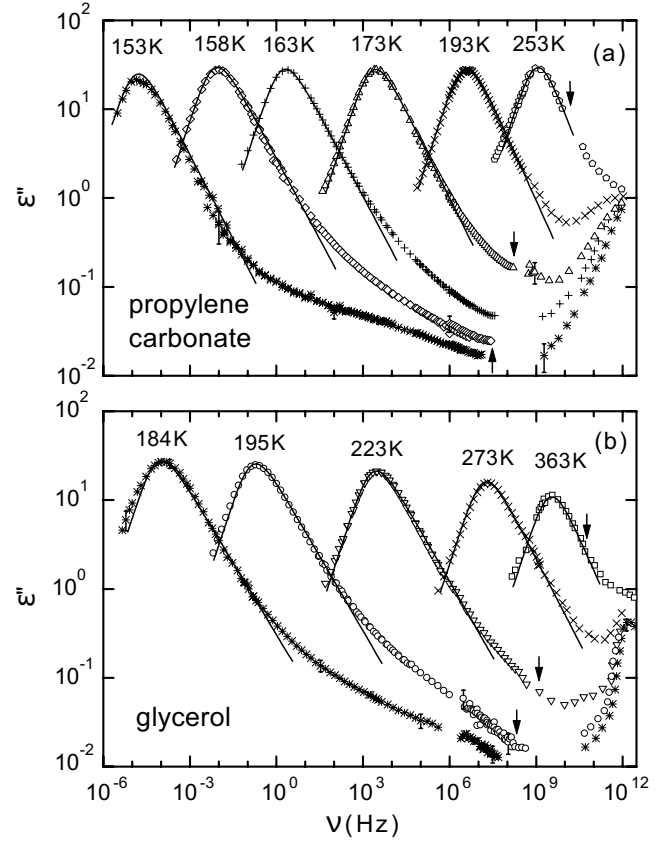


Fig. 1. Frequency dependence of the dielectric loss of PC (a) and glycerol (b) for various temperatures. For clarity reasons, only selected temperatures are shown and for frequencies $\nu > 1$ GHz only a part of the recorded data points are plotted (for the complete curves, see [28, 29]). The solid lines are fits with the CD function. The arrows indicate the cutoff frequency up to which the data have been used for the scaling plots of Figure 2.

2 Experimental

To collect data in a broad frequency range, various experimental setups were used, including a time domain spectrometer ($10 \mu\text{Hz} \leq \nu \leq 1 \text{kHz}$), frequency response analyzers and LCR-meters ($10 \text{mHz} \leq \nu \leq 30 \text{MHz}$), impedance analyzers (reflectometric technique, $1 \text{MHz} \leq \nu \leq 1.8 \text{GHz}$), a network analyzer (reflection and transmission, $100 \text{MHz} \leq \nu \leq 30 \text{GHz}$), a quasi-optic sub-millimeter wavelength spectrometer (transmission and phase measurement, $40 \text{GHz} \leq \nu \leq 1.2 \text{THz}$), and a Fourier-transform infrared spectrometer (transmission, $600 \text{GHz} \leq \nu \leq 3 \text{THz}$). For more experimental details the reader is referred to [30].

3 Results and discussion

Figure 1 shows the frequency dependence of the dielectric loss of PC and glycerol for various temperatures [28, 29]. $\epsilon''(\nu)$ exhibits the typical asymmetrically shaped α -relaxation peaks shifting through the frequency window with temperature. At high frequencies, in the GHz–THz

range, a minimum shows up, followed by another loss peak (the microscopic or boson peak). These high-frequency features are treated in detail in [25–29]. The solid lines in Figure 1 are fits of the α -peak region with the empirical CD function [2], which leads to good fits of the peak region. It was shown before [25–29], that the CD function describes the data clearly better than the Fourier transform of the KWW function. At frequencies about 2–3 decades above ν_p the excess wing shows up as a deviation of $\varepsilon''(\nu)$ from the CD fits, forming a second power law, $\varepsilon'' \sim \nu^{-b}$. The slope of the excess wing increases with increasing temperature as found previously for other glass-forming materials [6]. For high temperatures the excess wing seems to merge with the α -peak. At the lowest temperatures investigated, for both materials in the excess wing region a slight downward curvature of $\varepsilon''(\nu)$ may be suspected. This could be indicative for a β -relaxation process. The associated β -peak could be either superimposed to the excess wing, becoming important at low temperatures only [31], or it may even be the origin of the whole wing feature [4, 11–13]. However, the experimental errors are too large to allow for a definite conclusion concerning the presence of a β -peak and further experiments are in progress to clarify this question.

The data on glycerol and PC, after application of the Nagel [8] and Dendzik scaling [19] are shown in Figures 2(a) and (b), respectively. To maintain readability, for each material $\varepsilon''(\nu)$ curves for three different temperatures are shown only. For both materials the lowest temperature has been omitted, as here, due to the relatively high experimental errors near the α -peak region, an unequivocal determination of the scaling parameters is difficult. We also want to avoid contributions from a possible β -peak, superimposed to the wing at low temperatures¹ as, e.g., found for Salol [20]. The data have not been corrected for conductivity contributions, which only weakly affect $\varepsilon''(\nu)$ about 1–2 frequency decades below ν_p . The effect of the conductivity and the correct method of subtraction are a matter of controversy [22, 23] and therefore we decided to show the raw data. The scaling cannot be expected to work in the region of the ε'' -minimum in the GHz–THz region. Therefore only data points up to a cutoff frequency that is indicated by arrows in Figure 1 were used. The maximum value of X used at the different temperatures is indicated by the symbols. For the Dendzik scaling, the present plots extend to higher values of X_D than any previously published scaling plots. For the Nagel scaling, a plot up to $X_N = 12$ has been published in [24] but for one material only (glycerol). In addition, a Nagel plot up to $X_N = 16$, comparing different glass formers, was shown in [7]. However, this plot was obtained using data where the α -peak was missing and some crude assumptions for the determination of the scaling parameters had to be made to arrive at the scaling at high frequencies. Figure 2 reveals that both scaling procedures lead to good scaling of the

¹ Indeed, for PC, even after an optimization of the Nagel scaling parameters [23] within the experimental errors, the 153 K curve shows pronounced deviations from the curves at higher temperatures in the whole frequency range (not shown).

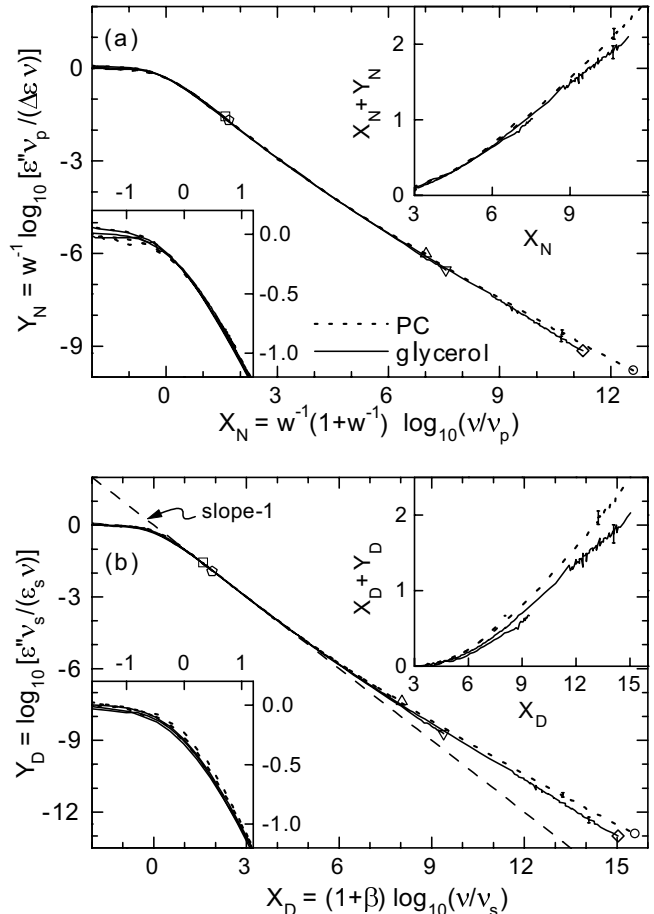


Fig. 2. (a) Nagel plot [8] of the dielectric loss results on PC at 158 K, 173 K, and 253 K and glycerol at 195 K, 223 K, and 363 K. The lower inset shows a magnified view of the region near $X = 0$. The upper inset shows the data for $X > 3$, with the ordinate replaced by the sum of ordinate and abscissa of the main frame. The symbols (same as in Fig. 1) indicate the highest value of X used for the different temperatures. (b) Same results as in (a), scaled according to Dendzik *et al.* [19]. The dashed line indicates the scaling curve for CD behavior at $\nu > \nu_s$. The insets are defined in the same way as for (a).

data at not too high values of X . There are reports of some shortcomings of the Nagel scaling in the region near $X = 0$ [18–20]. In contrast, the modified procedure proposed by Dendzik *et al.* was shown to lead to a better scaling in this region [19]. For the present data on PC and glycerol in this region (lower insets in Figs. 2(a) and (b)) indeed the quality of the Nagel scaling seems to be slightly worse, but the differences are very subtle. It was shown that the modified procedure proposed by Dendzik *et al.* is able to scale different CD curves onto one master curve [19]. At high frequencies this master curve is a straight line with slope -1 (straight dashed line in Fig. 2(b)). Then the excess wing should show up as deviations from the CD master curve at high X_D values, as is indeed seen in Figure 2(b). The most interesting result of these scaling plots is that, at high values of X , in both scaling approaches deviations

from a single master curve show up. These deviations are more pronounced in the Dendzik scaling. For example, in the Dendzik scaling the curves at 158 K for PC and at 195 K for glycerol start to significantly deviate from each other above an X_D value corresponding to 50 Hz for PC and 1 kHz for glycerol. In the Nagel plot this deviation occurs at 43 kHz for PC and at 3 MHz for glycerol. While for the Nagel scaling the deviations are almost within the experimental errors, this is not the case for the Dendzik scaling (see error bars in Fig. 2), which clearly fails at high frequencies. However, for both procedures the curves obtained at different temperatures for a single material scale (also including data at temperatures not shown in Fig. 2).

A general problem of the scaling plots is the fact that the Y value varies considerably, roughly following $Y = -X$, but the information of interest is the presence of small deviations from this variation. Therefore a much better resolution can be reached when correcting for this $Y = -X$ behaviour. This can be achieved by plotting the quantity $Z(X) = Y(X) + X$. In the upper insets of Figures 2(a) and (b) the data at $X > 3$ are plotted in this way. Clearly the resolution of the plots is enhanced and the deviations of the different curves from a universal scaling behaviour become more obvious.

Finally it may be mentioned that for high temperatures ($T \geq 253$ K in glycerol and $T \geq 203$ K in PC), the dielectric loss data can also be scaled onto each other by simply plotting $\varepsilon''/\varepsilon_p$ vs. ν/ν_p , with ε_p being the peak value of $\varepsilon''(\nu)$. Here the excess wing seems to have merged with the α -peak and β is nearly temperature-independent.

In Figure 3 both scaling plots are shown for the plastic crystals (o-CA, m-CA, and CNA). These plastic crystals have a rigid molecular structure and β -relaxation contributions from intramolecular degrees of freedom can be excluded. Their dielectric loss was shown to exhibit no or only a very weak wing [9]. In Figure 3, for clarity reasons, curves for two temperatures only are shown for each material. Clearly, the Nagel scaling (Fig. 3(a)) does not work for the plastic crystals investigated [9]. Again, in the upper inset of Figure 3(a) the deviations become more obvious. The deviations occur at much lower X_N values and are much stronger than for the glass-forming liquids (compare Fig. 2(a)). This can be rationalized considering that the $\varepsilon''(\nu)$ curves of the plastic crystals all follow a CD behaviour quite closely up to the highest frequencies [9] and that different CD curves do not scale in the Nagel plot [17]. In contrast, in the Dendzik plot (Fig. 3(b)) all curves for the plastic crystals scale onto the CD master curve (dashed line). It should be noted that these plastic crystals are relatively “strong” glass formers [26,32,33] within the classification scheme proposed by Angell and coworkers [34]. It cannot be excluded that the failing of the Nagel scaling in these materials is due to their high strength and not due to their plastic crystalline nature. This would also be in accord with Ngai’s ideas [16,12], concerning an explanation of the excess wing within the coupling model. Unfortunately, to our knowledge Nagel scaling was never applied to strong structural glass formers.

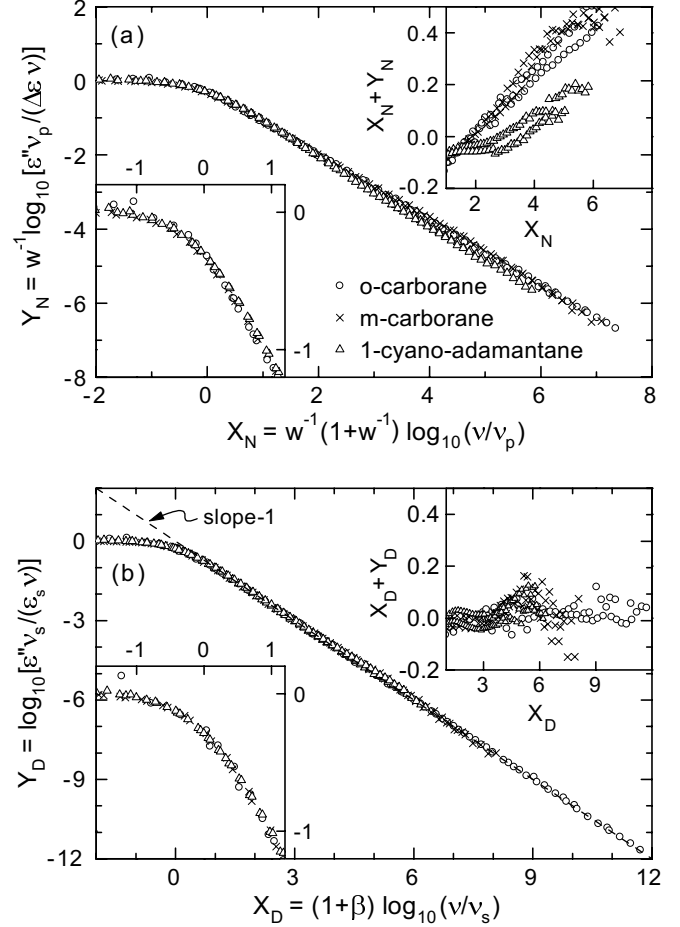


Fig. 3. Same plot as in Figure 2, but for the plastic crystals ortho-carborane at 130 K and 150 K, meta-carborane at 200 K and 210 K, and 1-cyano-adamantane at 280 K and 300 K [9].

Finally, we want to check if the excess wing approaches constant-loss behaviour at low temperatures in the samples investigated. Figure 4 shows the temperature dependence of β_{CD} for PC and glycerol, as resulting from the fits of the α -peak with the CD function (Fig. 1). For the CD function, the limiting half-width, $w^* \approx 2.6$, at which $\varepsilon'' = \text{const}$ [24] corresponds to $\beta^* \approx 0.18$. Inspecting Figure 4, it seems unlikely that $\beta_{CD}(T)$ will reach β^* at $T \approx T_{VF}$. From equation (1) and assuming $\gamma = \gamma' = 0.72$ [6], a different limiting value for the divergence of the static susceptibility, $\beta^{*'} \approx 0.38$, can be deduced [6]. While a linear extrapolation of $\beta(T)$ leads to a somewhat higher value of $\beta(T_{VF})$, an extrapolation to $\beta(T_{VF}) = 0.38$ at least is consistent with the data. The discrepancy between β^* and $\beta^{*'}$ arises due to the poor fulfilment of the assumption $\beta = 1/w$ made for the deduction of equation (1) from the Nagel scaling. In contrast to this, for the Dendzik scaling, equation (1) should be valid without such an assumption. However, while for the Nagel scaling a sample-independent value of $\gamma' = 0.72$ is consistent with our data (Fig. 2(a)), for the Dendzik scaling the value of γ' , read off from the high-frequency slope in the scaling plot, becomes sample-dependent. From Figure 2(b), $\gamma' = 0.79$ and

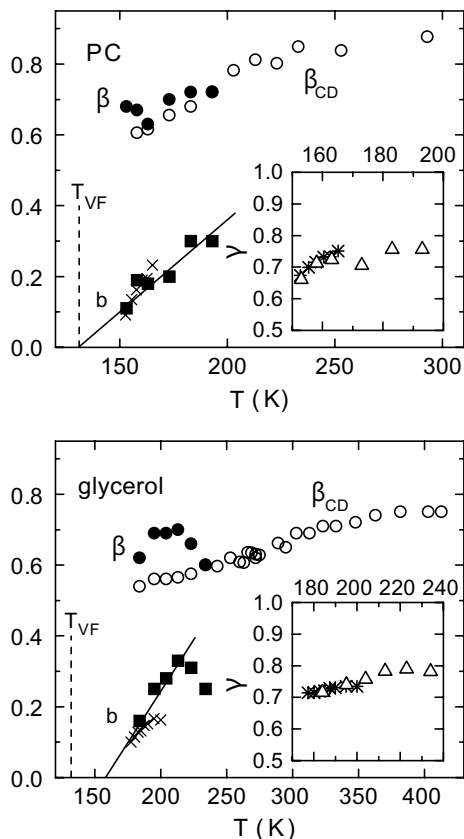


Fig. 4. Temperature dependence of β_{CD} (open circles) from fits of the α -peak with the CD function and of β (closed circles) and b (squares) from fits with a phenomenological ansatz (see text) for PC and glycerol. The crosses show the results reported in [6]. The straight solid lines demonstrate possible low-temperature extrapolations of $b(T)$. T_{VF} is indicated by the dashed line. The insets show γ as defined in equation (1) (triangles). The results from [6] are shown as stars.

$\gamma' = 0.715$ are determined. If $\gamma = \gamma'$ is assumed still to be valid, Equation (1) leads to $\beta^{*'} \approx 0.27$ and $\beta^{*'} \approx 0.4$ for glycerol and PC, respectively. The experimental data of $\beta_{CD}(T)$ may be consistent with these values (Fig. 4).

A more direct way to check for a divergent static susceptibility is the investigation of the temperature dependence of the exponent b . However, an unambiguous determination of b is difficult, especially at higher temperatures, where α -peak, wing, and the minimum are strongly superimposed to each other. For this purpose, a parameterization using the sum of two power laws was used in [6]. In order to take into account the additional contributions in the minimum region and the increase towards the boson peak, a constant loss and two power laws have to be added [35], leading to $\varepsilon'' = c_\beta \nu^{-\beta} + c_b \nu^{-b} + \varepsilon_c + c_3 \nu^{0.3} + c_n \nu^n$. This phenomenological ansatz was shown to provide good fits over a considerable frequency range [35]. In contrast to the scaling plots, for this analysis the lowest-temperature curves have been included as at least the $\nu^{-\beta}$ power law seems to be well defined. In addition, a possible β -peak, superimposed to the wing, may be expected to have only a small influence on the exponent b determined from the

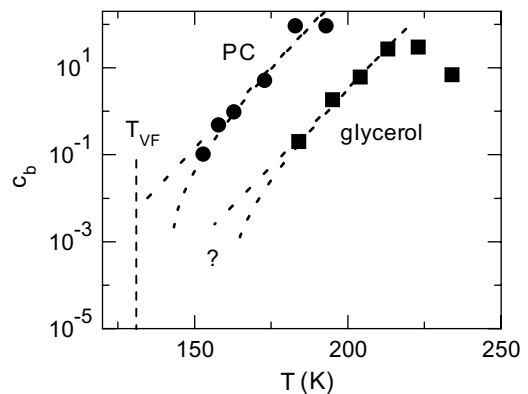


Fig. 5. Prefactor c_b of the power law $c_b \nu^{-b}$ used for the description of the excess wing in glycerol (squares) and PC (circles). The dashed line indicates $T_{VF} = 131$ K for glycerol [28]. For PC, $T_{VF} = 132$ K was determined [29]. The dotted lines demonstrate that it is not possible to draw conclusions concerning a non-zero value of c_b at T_{VF} .

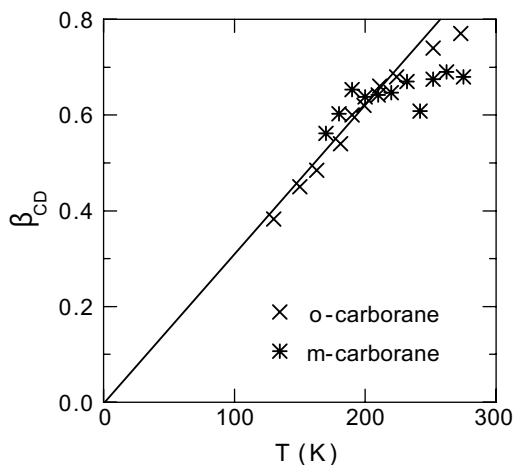


Fig. 6. Temperature dependence of β_{CD} from fits of the α -peak with the CD function for ortho- and meta-carborane. The solid line demonstrates a possible extrapolation to $b(T=0) = 0$.

fit. The resulting $b(T)$ and $\beta(T)$ are shown in Figure 4. $\beta(T)$ tends to assume somewhat higher values than β_{CD} due to the contributions of the wing power law ν^{-b} at low frequencies. In Figure 4 also the results of Leheny and Nagel [6] are included (crosses). Due to the larger frequency range available to us and the different evaluation procedure, we were able to extend the results of these authors to considerably higher temperatures. However, results of $b(T)$ could only be obtained up to 193 K for PC and up to 234 K for glycerol. Above these temperatures the excess wing has almost merged with the α -peak and an unequivocal determination of b is no longer possible.

In the insets of Figure 4 the obtained results for b and β are used to check the prediction $\gamma = \gamma'$ arising from the Nagel scaling. For both materials γ agrees roughly with the value of $\gamma' = 0.72(\pm 0.02)$, found from the high-frequency limiting slope of the Nagel scaling master curve [6], however, exhibiting a tendency to increase with temperature. Again, for comparison the results of Leheny and

Nagel [6] are included (stars). It is interesting that the slight temperature variation of γ , seen in the data of Leheny and Nagel [6], agrees with that detected in the present results. This indicates a systematic nature of this temperature dependence in contrast to the assumption of a constant $\gamma(T)$ within the error bars, made in [6]. The significant increase of $\gamma(T)$, detected for both materials, seems reasonable in light of the merging of excess wing and α -peak at high temperatures which can be suspected from Figure 1. This implies the approach of $\gamma = 1$ for high temperatures.

As demonstrated by the solid lines in Figure 4, $b(T)$ can be reasonably extrapolated to zero at T_{VF} for PC and to about $T_{VF} + 25$ K for glycerol. The result for PC indeed seems to support the approach of a constant loss at T_{VF} . However, even if b becomes zero, a “divergent” susceptibility will only be reached, if the pre-factor $c_b(T)$ does not approach zero for $T \rightarrow T_{VF}$. In Figure 5 $c_b(T)$ is shown for both materials. As indicated by the dashed lines the results do not allow for a definite conclusion concerning a finite value at T_{VF} .

For the plastic crystals, the situation is different, as here the wing is missing and a “divergent” susceptibility would only be reached if $\beta(T)$ approached zero at a finite temperature. Figure 6 shows the temperature dependence of β_{CD} for o-CA and m-CA [26, 33]. Indeed, $\beta_{CD}(T)$ weakly decreases with decreasing temperature. Due to the fact that these plastic crystals are relatively “strong” glass formers [26, 33], T_{VF} is difficult to determine. It can be expected to be located far below T_g where it is not possible to obtain significant information from an extrapolation of $\beta(T)$. The line in Figure 6 proposes a linear extrapolation of the low-temperature data to $\beta_{CD}(T = 0 \text{ K}) = 0$ but other extrapolations are also possible.

4 Conclusions

In summary, we have investigated the scaling of extremely broadband dielectric data using two different procedures. The spectra on two glass-forming liquids and three plastic crystals have been analysed, extending the range of the abscissa to higher values than in former analyses. For the glass-forming liquids, the approaches proposed by Nagel and coworkers [8] and by Dendzik *et al.* [19] both lead to a satisfactory scaling of the loss data at low frequencies. The extended frequency range available reveals a general failure of the Dendzik scaling at the highest frequencies investigated, while this effect is almost negligible for the Nagel scaling. More broadband data on glass-forming liquids are needed to check for the significance and universality of these deviations. If they were confirmed, this may indicate that the wing is not so intimately related to the α -relaxation as thought up to now. Then it could be caused by processes, other than the α -process (*e.g.*, a β -relaxation [11, 12]). But also a superposition of a very weak β -relaxation and the wing may be invoked to explain the failing of the scaling at high frequencies [20, 23, 31].

In addition, the broader frequency range of our data allowed for an extended check of the cross-over of the

wing to constant-loss behavior at a temperature near T_{VF} , claimed by Nagel and coworkers [6, 24]. The temperature dependence of the width parameter of the α -peak gives no evidence for such a behaviour. For PC and to a lesser extent also for glycerol, the temperature dependence of the wing exponent may be consistent with the proposed approach of a divergent static susceptibility. However, the development of the power law prefactor for low temperatures is still an open question. In our opinion the dielectric data reported in the literature and even our extended set of data do not provide a convincing experimental proof for a divergent susceptibility at low temperatures.

The data on the plastic crystals clearly cannot be scaled by the procedure proposed by Nagel and coworkers [9]. But using the Dendzik scaling, all curves from the investigated plastic crystals fall onto one master curve, different from the master curve of the glass-forming liquids. This master curve is identical with the master curve for the Cole-Davidson function. This result corroborates the recent finding that plastic crystals without intramolecular degrees of freedom do not show an excess wing but follow a CD behaviour up to the highest frequencies [9]. In light of the missing excess wing, for plastic crystals a divergence of the static susceptibility at finite temperatures seems unlikely, but cannot be fully excluded.

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