



# Evolution of the dynamic susceptibility of paradigmatic glass formers below the critical temperature $T_c$ as revealed by light scattering

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## Abstract

We have studied the molecular glass formers toluene and picoline in the liquid and glassy state by quasi-elastic light scattering applying tandem Fabry–Perot interferometry. Also, we perform an analysis of the susceptibility spectra of the inorganic glasses calcium potassium nitrate (CKN) and silica at  $T < T_g$ . At high temperatures the evolution of the susceptibility minimum in toluene and picoline is described by mode coupling theory (MCT), and the critical temperature  $T_c$  can be determined consistently. Below  $T_c$  the asymptotic laws of MCT fail, and the excess wing of the  $\alpha$  process is rediscovered in a similar way as in the dielectric spectra. The wing contribution controls the evolution of the susceptibility minimum and is attributed to a secondary relaxation process different from the slow  $\beta$  process. The fast dynamics spectra exhibit a trend to white noise at low frequencies ( $T < T_c$ ). We conclude, regarding the fast dynamics, that MCT predictions hold also below  $T_c$ . In CKN the relaxation mechanism changes below  $T_g$  and the fast relaxation in both CKN and silica is reproduced by assuming thermally activated transitions in asymmetric double well potentials, while this is not possible for picoline or toluene.

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## 1. Introduction

The molecular slowing-down in simple glass forming liquids has been investigated by several techniques such as neutron and light scattering (LS) as well as dielectric spectroscopy (DS) [1]. Mode coupling theory (MCT) reproduces well the evolution of the dynamic susceptibility above the

critical temperature  $T_c$ , the latter being about 20% above the glass transition temperature  $T_g$ . Also, molecular dynamics simulations strongly support the MCT scenario [2]. Below  $T_c$  the situation is less clear. Concerning the temperature dependence of the non-ergodicity parameter  $f(T)$ , which is expected to show a cusp-like behavior at  $T_c$ , agreement is a matter of debate [1,3]. Related to this is the prediction of a ‘knee’ in the fast dynamics spectrum upon cooling ( $T < T_c$ ), i.e., a crossover to white noise spectrum at low frequencies. However, up to now no such behavior has been confirmed.

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The available frequency window of LS technique is restricted to frequencies  $\nu > 0.3$  GHz, and one can argue that the knee can not be identified unambiguously within this narrow range. Recent MCT analyses have shown that the experimental data are compatible with the presence of a knee [4]. In addition, below  $T_c$  the analysis of spectra is hampered by the fact that the time constant  $\tau_\alpha$  of the main relaxation ( $\alpha$  process) actually does not diverge at  $T_c$  as presumed by the idealized MCT. Hence, contributions from the  $\alpha$  process have also to be taken into account when the analysis is extended to  $T < T_c$ . However, there is no straightforward way to do this. MCT assumes a hopping mechanism but in the analyses presented so far, the interpretation of the hopping parameter is not clear and no identification with otherwise testable quantities has been proposed [4,5]. Clearly, further experimental data is needed to explore the features of the susceptibility spectrum below  $T_c$ .

Below  $T_g$ , due to structural arrest, only secondary relaxation processes are identified but here, concerning fast dynamics, experimental facts are rare, since most of the LS investigations cover temperatures above  $T_g$ , only. The slow dynamics ( $<1$  MHz) of glass formers have been thoroughly probed by many different methods and attributed to the slow  $\beta$  process, i.e., of Johari–Goldstein (JG) type [6–9]. On the other hand, starting from very low temperatures ( $T < 1$  K) mostly inorganic glass formers have been studied in order to understand the low temperature anomalies of glasses [10]. Commonly structural defects are assumed which can be described by dynamics in asymmetric double well potentials (ADWP). Below a few K tunneling is observed which leads to quite universal relaxation behavior. At higher temperatures, some authors have extended the model to include also thermally activated crossing of the barrier [10]. The validity of this extension is debated, and it is by no means clear how the dynamics observed in inorganic glasses compare to those of organic systems and whether the dynamics probed by LS below but close to  $T_g$  may be attributed to such relaxation processes. Again, progress may become only possible when experimental data covering several decades in frequency are obtainable even below  $T_g$ .

The aim of this contribution is to present susceptibility spectra of paradigmatic organic as well as inorganic glass formers as obtained by tandem Fabry–Perot interferometry (TFPI) in the temperature range  $2.5T_g$  to  $0.05T_g$ . For comparison, dielectric data are shown. We want to address the question how the susceptibility spectra evolve below  $T_c$  and even below  $T_g$ . We will demonstrate that the so-called excess wing of  $\alpha$  process, identified in dielectric experiments, is rediscovered in the LS spectra at  $T_c > T > T_g$ , and we think that the emergence of the excess wing determines the evolution of the susceptibility minimum in the GHz range at low temperatures. Thus, including a hopping mechanism, is not sufficient to understand the dynamics below  $T_c$ . Analyzing the LS data below  $T_g$ , we want to demonstrate that quite different relaxation behavior is observed, in particular, when organic and inorganic glass formers are compared.

## 2. Experimental

We study the molecular glass formers toluene and  $\alpha$  picoline on the one hand, and the inorganic glasses calcium potassium nitrate (CKN) and silica on the other. Picoline has the same molecular weight as toluene but a much stronger dipole moment, and it exhibits less tendency to crystallize allowing us to compile more LS spectra at  $T_g < T < T_c$  [11]. To our knowledge no LS and DS data are reported on this system. Part of the data on toluene, CKN and silica together with experimental details have been reported previously [11–14]. In the case of silica we have compiled now LS data up to 1600 K.  $T_g$  is found by DSC to be 133 K in picoline and 117 K in toluene. We have applied additional interference filters in order to suppress parasitic contributions from higher orders in the TFPI spectra. Only recently, it has become clear that such filters are needed to get reliable broad band LS spectra [14–16].

## 3. Theoretical background

For proceeding the predictions of the idealized MCT are briefly summarized [1]. Due to the

interplay of fast dynamics and  $\alpha$  process a minimum is found in the susceptibility at  $T > T_c$  which can be interpolated by

$$\chi'' = \chi''_{\min} [b_{\text{MCT}}(v/v_{\min})^{a_{\text{MCT}}} + a_{\text{MCT}}(v/v_{\min})^{-b_{\text{MCT}}}] / (a_{\text{MCT}} + b_{\text{MCT}}), \quad (1)$$

where  $v_{\min}$  and  $\chi''_{\min}$  are the frequency and the amplitude of the minimum. The exponents  $a_{\text{MCT}}$  and  $b_{\text{MCT}}$  determine the low-frequency behavior of the fast dynamics and the high-frequency part of the  $\alpha$  process, respectively; and are connected via the exponent parameter  $\lambda$ . The temperature dependence of  $v_{\min}$ ,  $\chi''_{\min}$  and of the time scale  $\tau_\alpha$  is given by  $\chi''_{\min} \propto \sigma^{1/2}$ ,  $v_{\min} \propto \sigma^{1/2a}$ ,  $\tau_\alpha \propto \sigma^{-\gamma}$ ,  $T > T_c$  (2)

with  $\sigma = T - T_c$ . The exponent  $\gamma$  is related to the exponents  $a_{\text{MCT}}$  and  $b_{\text{MCT}}$  via  $\gamma = 1/(2a) + 1/(2b)$  and  $a$  and  $b$  are fixed by the exponent parameter  $\lambda$ .

From  $\chi''_{\min}(T)$ ,  $v_{\min}(T)$  and  $\tau_\alpha(T)$  the critical temperature  $T_c$  should follow. Below  $T_c$  the fast relaxation spectrum is expected to change its shape. A crossover from a power law with an exponent  $a_{\text{MCT}}$  at high frequencies to a white noise spectrum ( $a = 1$ ) at low frequencies is expected. As a consequence of the appearance of this ‘knee’ a decrease of the relaxation strength of the fast dynamics is expected upon cooling. This constitutes the anomaly of the non-ergodicity parameter, a generalized Debye–Waller factor.

Concerning the low temperature dynamics in glasses we will recourse to the ADWP model of Theodorakopoulos and Jäckle [17] and Gilroy and Phillips [18]. These authors extended the standard tunnel model to include also thermally activated transition of the barrier. Following the assumption that the distribution  $f(\Delta)$  of the asymmetry parameter  $\Delta$  is flat the LS susceptibility is related to the distribution of barrier heights  $g(V)$  by a simple expression

$$\chi''_r \propto 1/kT \int \int \frac{\omega\tau}{1 + (\omega\tau)^2} \sec h^2(\Delta/2kT) \times f(\Delta)g(V) d\Delta dV \cong \int \frac{\omega\tau}{1 + (\omega\tau)^2} g(V) dV. \quad (3)$$

The correlation time  $\tau$  is determined by an Arrhenius law, explicitly  $\tau = \tau_0 \exp(V/kT)$ .

#### 4. Results and discussion

Fig. 1 displays the LS susceptibility spectra of toluene, and Fig. 2 shows the picoline spectra as obtained by LS (Fig. 2(a) and (b)) and by DS (Fig. 2(c)). Picoline is a type A glass former since no slow  $\beta$  process can be identified in the DS data whereas toluene is a type B system with a strong slow  $\beta$  process [7,19]. For type A systems a kind of wing at high frequencies shows up close to  $T_g$  which degenerates to a simple power-law contribution with an exponent  $c \cong -0.1$  at  $T < T_g$  (cf. Fig. 2(c)). Inspecting the LS spectra of both toluene and picoline it is clear that quasi-elastic contributions are also observed below  $T_g$ . Even at temperatures around 10 K, in addition to the boson peak a crossover to a relaxational contribution is found.

Figs. 1 and 2(a) also displays the MCT interpolation of the susceptibility minimum by Eq. (1) with a fixed exponent parameter  $\lambda = 0.689$  leading to  $a_{\text{MCT}} = 0.331$  and  $b_{\text{MCT}} = 0.66$  in the case of toluene, and  $\lambda = 0.70$  leading to  $a_{\text{MCT}} = 0.327$  and  $b_{\text{MCT}} = 0.641$  in the case of picoline. Clearly, at high temperatures the MCT interpolation repro-

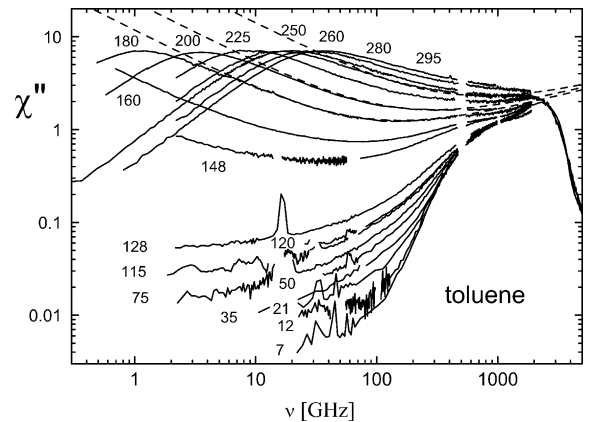
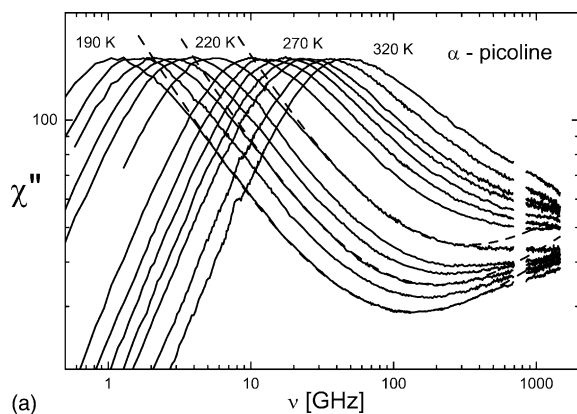
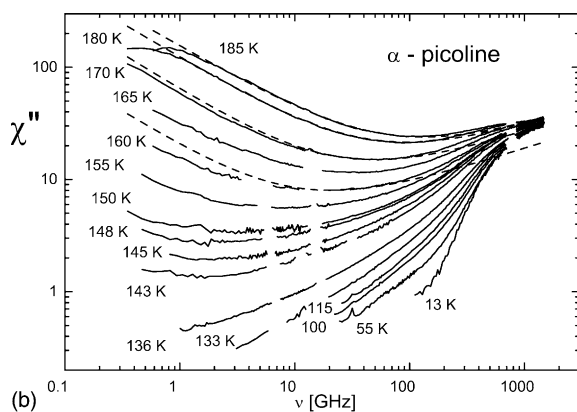


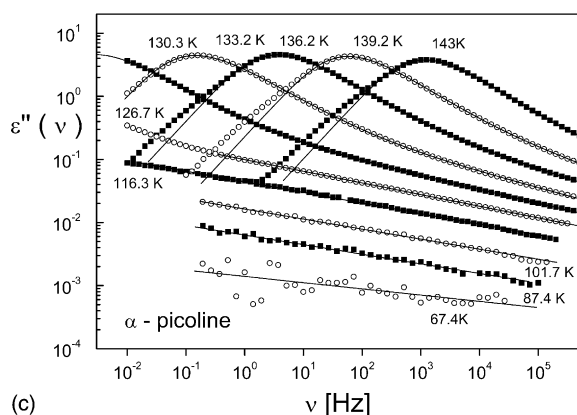
Fig. 1. LS spectra of toluene; (---) interpolation of the susceptibility minimum by MCT (Eq. (1)). Numbers indicate temperature in K.



(a)



(b)



(c)

Fig. 2. (a) LS spectra of picoline at high temperatures ( $T \geq 190$  K); (---) MCT interpolations (Eq. (1)). (b) LS spectra below 190 K; (---) MCT interpolation; note that MCT interpolation becomes worse at lower temperatures. (c) DS spectra; (—) fit interpolating both  $\alpha$  relaxation peak and excess wing.

duces well the minima whereas at low temperatures the agreement becomes worse (cf. Fig. 2(b),

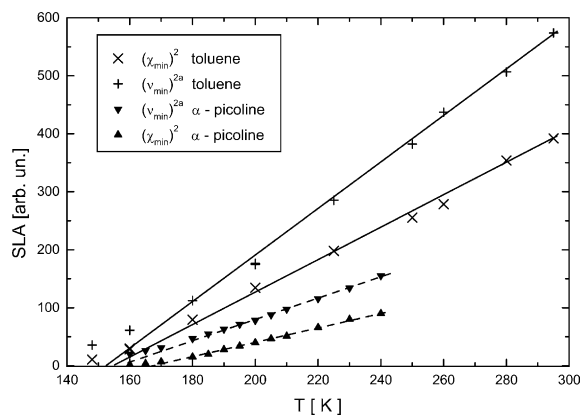


Fig. 3. Test of MCT scaling laws: temperature dependence of  $\chi''_{\min}$  and  $v_{\min}$  in toluene and picoline, plotted in a way to reveal linear behavior; SLA: scaling law amplitude.

dashed line). The parameters  $\chi_{\min}$ ,  $v_{\min}$  and  $\tau_{\alpha}$  [11] all exhibit the behavior expected from MCT at  $T > T_c$  (cf. Eq. (2) and Fig. 3): extrapolating linearly the scaling laws, one obtains consistently  $T_c = 153 \pm 3$  K for toluene, and  $T_c = 162 \pm 5$  K for picoline. Thus, the idealized MCT provides a very satisfying interpolation at high temperatures. However, first deviations from the scaling laws appear already around 180 K in both systems (cf. Fig. 3).

In order to unravel the origin of the deviations from the MCT scaling laws we present in Fig. 4 the LS data of toluene and picoline in a way to provide a master plot for the  $\alpha$  relaxation peak, i.e.,  $\chi''$  is plotted as a function of  $\nu\tau_{\alpha}$ . The time constant  $\tau_{\alpha}$  has been taken from analyzing the  $\alpha$  process as revealed by NMR, DS and LS data [7,11]. In this presentation, following MCT, the loci of the minimum are expected to obey the relationship  $\chi_{\min}(\nu) \propto (v_{\min}\tau_{\alpha})^{-b_{\text{MCT}}}$  as is indicated by the dashed line. This is observed at  $T > 160$  K in the case of toluene, and at  $T > 180$  K for picoline. Below this crossover temperature (from now on called  $T_x$ ) deviations show up. It appears that the minimum is determined not any longer by the high-frequency power law of the  $\alpha$  process (with exponent  $b_{\text{MCT}}$ ) but rather by some spectral feature which only appears below  $T_x \cong 180$  K. Since picoline does not exhibit a slow  $\beta$  process, the only candidate for a change in the susceptibility is the excess wing

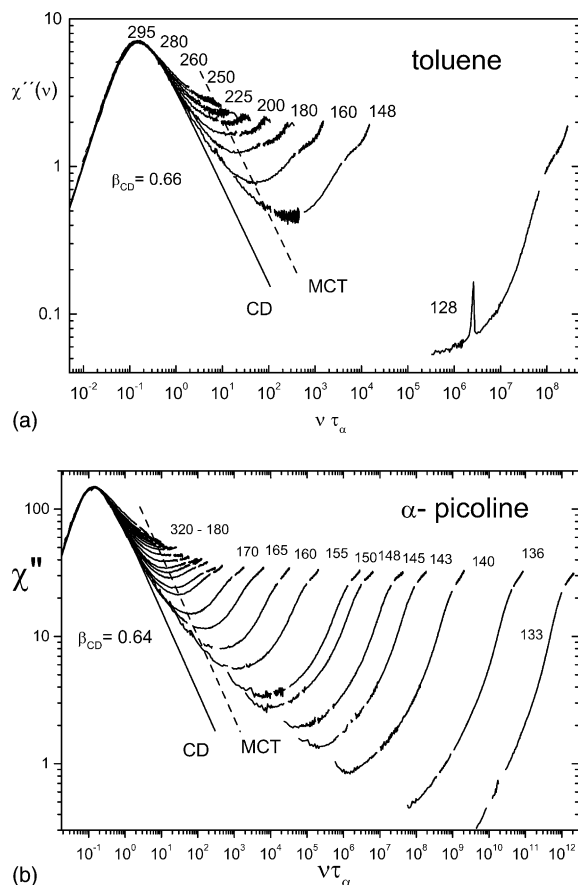


Fig. 4. LS spectra of toluene (a) and of picoline (b) plotted as a function of the reduced frequency  $\nu\tau_\alpha$ ; (—) interpolation of the  $\alpha$ -peak by a Cole–Davidson susceptibility; (---) loci of the susceptibility minimum according to MCT.

which clearly shows up in the DS spectra (cf. Fig. 2(c)). Comparing the LS data for toluene and picoline it is obvious that they are quite similar; in particular, the same deviations from the high temperature MCT scenario show up at low temperatures (cf. Fig. 4). Thus, one may speculate that the  $\beta$  process is actually not probed by LS experiments, a conclusion also drawn for the glass former Epon [20]. This supports the view that the wing contribution is a distinct phenomenon as compared to the JG process [7]. Inspecting the DS spectra of glycerol and propylene carbonate [7,21], which have been measured to higher frequencies, it is found that the excess wing indeed disappears at high temperatures. We conclude that below

the temperature  $T_x$  the high temperature evolution of  $\chi''(\nu)$  being well described by MCT changes due to the appearance of the excess wing of the  $\alpha$  process.

The excess wing is observed by DS in all supercooled liquids provided that no distinct  $\beta$  process is present. Its similar spectral shape has been stressed by several authors and may be described by a power-law contribution with exponent  $c < \beta_{CD}$  [7,22,23]. Applying a recently introduced distribution of correlation times [7] we can fully interpolate the high-frequency part of the DS spectra of picoline including both the  $\alpha$  peak and the wing (cf. Fig. 2(c), solid line). At very low frequencies some deviations from the fit occur which are usually not found in low molecular weight glass formers; the reason is yet unclear. The wing is believed to be a kind of precursor of the  $\alpha$  process or a secondary relaxation process [7,24]. Recent NMR experiments favor the picture that the wing is a highly restricted reorientation similar to the slow  $\beta$  process [8,9] though the first is LS active and the latter not. We think it cannot be attributed to a kind of hopping mechanism. We note that in the glass ( $T < T_g$ ) the wing contribution degenerates to an almost constant loss behavior, a behavior reported for many systems [25]. Consequently, a crossover to apparent constant LS susceptibility is expected and indeed found [26].

Inspecting further the LS data of picoline below  $T_x$  (cf. Fig. 2(b)) it can be recognized prior to any fit procedure that the exponent of the fast dynamics contribution (fast process) becomes a strong function of temperature in the interval  $T_g < T < T_x$ . In order to describe this phenomenon we introduce the exponent  $a$  which is the exponent attributed to fast dynamics spectrum below  $T_x$ . It is obvious that  $a > a_{MCT}$ , e.g.,  $a = 0.72$  is found at 115 K. For this low temperature ( $T < T_g$ ) the exponent can directly be read from the data since no interference with the wing contribution is expected. It appears that  $a$  is essentially constant at  $T < T_g$  but drops above  $T_g$  and finally reaches the level of  $a_{MCT}$  close to  $T_x$ . This may be taken as indication that below  $T_x$  the exponent  $a$  of the fast dynamics contribution increases towards 1. In other words, the susceptibility spectrum crosses over to a white noise spectrum, a behavior

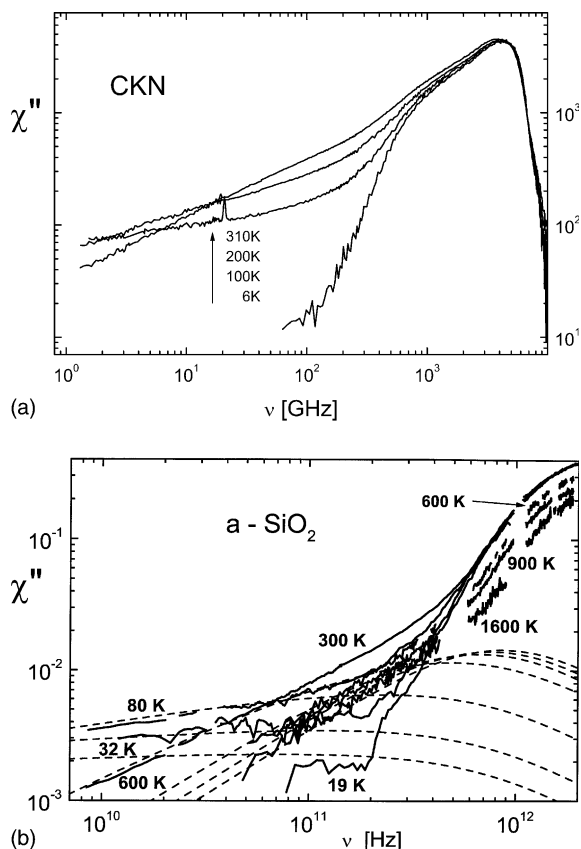


Fig. 5. LS spectra of CKN (a) and of silica (b) below  $T_g$ ; (---) interpolation of the silica spectra within the ADWP model taking the distribution of barrier heights  $g(V)$  from Fig. 7; note that spectra for  $T > 300$  K of silica do not change their spectral shape.

similarly predicted by MCT. This trend is impossible to follow to a temperature at which  $a = 1$  holds since the glass transition at  $T_g$  interferes leading to a temperature independent exponent  $a$  at  $T < T_g$ . We note that the situation is less clear for toluene due to the weak signal-to-noise ratio. Such crossover to white noise may be also anticipated by inspecting DS data of propylene carbonate [21] as well as LS data of CKN (cf. below and Fig. 6). Thus, we think that also at  $T_c > T > T_g$  the fast dynamics are still described by MCT.

Next we consider two inorganic glass formers CKN and silica which both have been studied below  $T_g$  only (cf. also Ref. [12–14]). Fig. 5 displays

the LS susceptibility data. Since data are presented well below  $T_g$ , secondary relaxation contributions are observed only. Obviously, the latter are characterized by power-law contributions with an exponent  $a$  showing a strong temperature dependence. Even a ‘crossing’ of the power laws is found. This behavior we take as a clear indication that the quasi-elastic contributions are indeed originating from relaxation processes. For these systems the fast dynamics spectra exhibit completely different behavior as compared to the organic glass formers discussed above, for which the exponent  $a$  is essentially temperature independent below  $T_g$ . The temperature dependence of the exponent  $a$  is plotted in Fig. 6. Below 300 K the exponent  $a$  for both CKN and silica is proportional to temperature. In the case of silica, for which we compiled data up to 1600 K, a crossover to  $a \cong 1$  is observed at highest temperatures, i.e., the spectra of silica above 400 K are virtually indistinguishable.

In order to describe the data of silica and CKN we apply the ADWP model. Assuming as the most simple case an exponential distribution of barriers,  $g(V) = V_0^{-1} \exp(-V/V_0)$ , the model predicts for the low-frequency part of the susceptibility,  $\nu \ll (2\pi\tau_0)^{-1}$ , a power-law contribution with an exponent  $a = kT/V_0$  at low temperature,  $T \leq V_0/k$ . At higher temperatures the exponent is expected to crossover to  $a = 1$ . Exactly, this behavior is

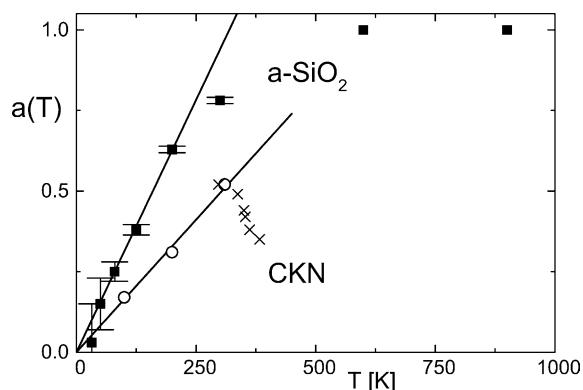


Fig. 6. The exponent  $a$  as a function of temperature for silica and CKN, above  $T_g$  data for CKN (crosses) are taken from Ref. [15].

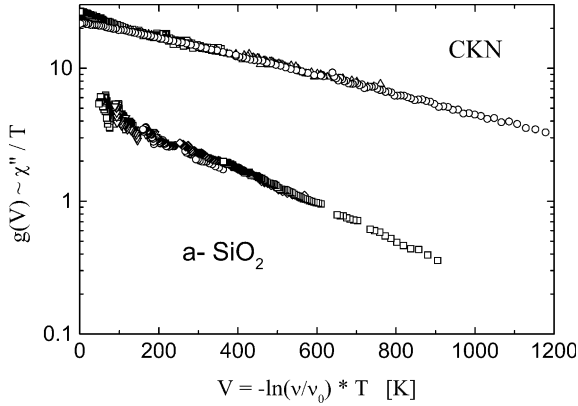


Fig. 7. Distribution of barrier heights  $g(V)$  obtained from the LS spectra of Fig. 5 by rescaling the axes; note that barrier heights are on the order of  $T_g$ .

observed. Thus, in the systems studied the dynamics are well reproduced by the ADWP model. In Fig. 6 we added data for CKN at  $T > T_g$  ( $= 333$  K) taken from Ref. [15]. Above  $T_g$  the ADWP model breaks down, and  $a(T)$  starts to decrease upon heating. As we know that MCT reproduces the LS data of CKN at high temperatures [27] and an exponent  $a_{\text{MCT}} \cong 0.3$  is found above  $T_x$ , one can anticipate as in the case of picoline that a crossover to white noise spectrum is found when cooling the liquid below  $T_c$ .

As we have shown in previous work [12–14] the LS spectra directly allow to extract the distribution  $g(V)$  by rescaling the axes with  $T$  and  $1/T$ , respectively, and, indeed an exponential distribution is revealed with barrier height on the order of  $T_g$  (cf. Fig. 7). Equally well, the LS spectra may be interpolated quantitatively by the ADWP model applying Eq. (3) and taking  $g(V)$  from Fig. 7 as input [14]. The results for silica are included in Fig. 5(b). Clearly, a very satisfying agreement is found at low frequencies. For high frequencies the boson peak obscures the relaxation contribution.

## 5. Conclusions

The goal of this contribution is to review our results on the evolution of the LS susceptibility

while cooling a glass former from high temperatures ( $T > T_c$ ) to temperatures well below  $T_g$ . In the case of the organic glass formers toluene and picoline above  $T_c$ , the susceptibility minimum is described by the asymptotic laws of MCT allowing to identify consistently the critical temperature  $T_c$ . Below  $T_c$  the locus of the susceptibility minimum does not follow the MCT prediction any longer. Instead, it is controlled by the appearance of the excess wing which is attributed to some secondary relaxation process different from the JG process. There are indications that the susceptibility spectrum crosses over to a white noise spectrum as predicted by MCT when temperature is decreased below  $T_c$ . This trend is impossible to follow to a temperature at which actually  $a = 1$  holds since below  $T_g$  the exponent  $a$  becomes independent of temperature or, in the case of CKN (and silica), an anew decrease of  $a(T)$  is observed. In other words, new relaxational features appear below  $T_g$ . We conclude that concerning the fast dynamics MCT holds even below  $T_c$  down to  $T_g$ , though the appearance of the excess wing governing the low-frequency part the susceptibility minimum in the GHz regime is not accounted for by the theory. We note that the failure of the high temperature laws of MCT is first observed at  $T_x$  which is somewhat higher as compared to  $T_c$ , the latter being obtained from extrapolating the scaling laws (cf. Fig. 3). This discrepancy may be the reason that for some glass formers a broad range of  $T_c$  values is reported.

Below  $T_g$ , our experiments demonstrate that quasi-elastic contributions show up in the LS spectra down to low temperatures and that they can also be resolved by the tandem Fabry–Perot technique. In the case of the inorganic glasses silica and CKN the ADWP model assuming thermally activated dynamics in double well potentials describes the dynamics at  $T < T_g$ . The ADWP approach leads to the same (simple) expression for the susceptibility as applied for the JG process [7]. However, the typical barrier heights involved in the case of the ADWP dynamics are on the order of  $T_g$ , and it appears that no low-energy cut exists, whereas for the JG process barrier heights of some  $10T_g$  are found within a Gaussian distribution. Furthermore, all molecules participate in the JG

process [7,19] while this is not assumed for the ADWP which are commonly attributed to rare defects in the glass [10]. Finally, we note that in the case of boron oxide the ADWP model fails [28]. Here, a similar behavior is found as, e.g., in picoline, i.e., the exponent  $a$  is essentially temperature independent ( $T < T_g$ ). In contrast, for polystyrene we find that the ADWP model works in first approximation [12]. Hence, up to now no definite understanding of the fast dynamics in glasses ( $T < T_g$ ) can be given. In particular, the temperature independence of the exponent  $a$  in picoline or toluene is not well understood. For example, it may be explained by assuming the dominance of quasi-elastic contributions originating from anharmonicity effects [29].

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