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The light scattering relaxation function of glass-forming molecules: a general probabilistic approach

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Abstract

Depolarized photon correlation spectra of two fragile glass-forming epoxy molecules of different molecular weights are reported. The correlation function of the simplest molecule shows stretched-exponential behaviour, while deviations from this form have been observed for the larger molecule, suggesting the presence of a second, long-time power law. The relaxation dynamics is suitably described for both systems in terms of a probabilistic approach based on the tool of the limit theorems of probability theory. On the basis of this theory, the presence of the long-time power law can be related to the existence of intermolecular interactions, while the stretched-exponential behaviour of simple molecules can be obtained in the case of negligible long-range interactions.

(Some figures in this article are in colour only in the electronic version; see www.iop.org)

1. Introduction

The dynamics of supercooled systems is a fascinating and still not completely solved problem of condensed matter physics. The supercooled liquid, i.e. a system which remains liquid below the melting temperature for a long time before crystallization, is characterized by a dramatic increase of its viscosity for temperature approaching the glass transition, accompanied by nonexponential (stretched) relaxation processes [1]. Instead of the pure Debye response the stretched-exponential decay law, described by the time-domain Kohlrausch–Williams–Watts (KWW) relaxation function

$$\phi_{KWW}(t) := \exp \left[- \left(\frac{t}{\tau_{KWW}} \right)^\gamma \right] \quad (1)$$

(where $0 < \gamma < 1$ is the stretching parameter and $\tau_{KWW} > 0$ is the relaxation time) is often suggested to represent the experimental data (see [2] and references therein). However, this

law is not universally valid since it appears that the supercooled systems usually follow the two-power-law response [3], i.e. that the susceptibility function

$$\chi(\omega) = \chi'(\omega) - i\chi''(\omega)$$

associated with the time-domain relaxation function $\phi(t)$ by the relation

$$\chi(\omega) = \int_0^{\infty} e^{-it\omega} \left(-\frac{d\phi(t)}{dt} \right) dt \quad (2)$$

exhibits two fractional power laws in frequency, namely,

$$\begin{aligned} \chi'(\omega) \propto \chi''(\omega) \propto (\omega\tau)^{n-1} & \quad \text{for } \omega\tau \gg 1 \\ \chi'(0) - \chi'(\omega) \propto \chi''(\omega) \propto (\omega\tau)^m & \quad \text{for } \omega\tau \ll 1 \end{aligned} \quad (3)$$

with exponents $0 < n, m < 1$ and relaxation time τ . Let us add that the KWW function (1) leads to the high-frequency power law only, with

$$n = 1 - \gamma.$$

Different microscopic models have been proposed to explain the origin of the observed nonexponential response; for example, the diffusion and trap depletion model [2] which describes the KWW behaviour of both electronic and molecular relaxation, and the mode-coupling theory [4] which gives a rationale for the dynamics of slightly supercooled systems including the high-frequency (von Schweidler) power law of the structural relaxation.

In section 2 we present briefly a probabilistic approach to modelling relaxation phenomena in complex systems, introduced and studied recently [5–12]. In this approach a general formalism, applied to the analysis of the first passage of a system, allows us to treat relaxation processes regardless of the precise nature of the interactions in a particular system—rather as a result of some statistical rules that the large system follows as a whole. On the other hand, the model relates the local random characteristics of complex systems to the relaxation laws observed on the macroscopic level and hence it can provide a clue to better understanding of the physical mechanism of relaxation by yielding strict conditions leading to a particular response.

The formula for a time-domain relaxation function $\phi(t)$, derived in the framework of the approach to relaxation phenomena presented [6], on the basis of limit theorems of probability theory [13–17], may lead, for properly chosen parameters, to the two-power-law property (3) [8]. The low-frequency power law appears when large-scale correlation is taken into account. Its exponent m depends on both long- and short-range interactions while the high-frequency power-law exponent n is determined by the latter only. When one neglects the long-range interactions, the model yields the stretched-exponential decay law. The probabilistic approach considered results, hence, in a formula for $\phi(t)$ that is a generalization of KWW function (1) and, moreover, can be applied as a time-domain counterpart of the Havriliak–Negami (HN) fitting function [18], commonly used for susceptibility data (however, the functions are not exactly related by (2)).

In sections 3 and 4 we report the results of a depolarized light scattering study of monoepoxide and diepoxide systems. Epoxy systems are particularly appropriate for the study of the supercooled state of matter. They do not crystallize on cooling the system below the melting point, give a strong depolarized signal and are available with different molecular structures that allow us to study the influence of the structure on the dynamics of the system. Here, to fit the experimental data obtained by the photon correlation technique, the formula derived in the approach presented in section 2 has been used. A detailed analysis of the shape of the relaxation function detected in the fitting procedure gives evidence of the stretched-exponential decay-law response of monoepoxide and of the two-power-law response

of diepoxide liquids that, on the basis of the theoretical considerations, suggests the presence of long-range interactions in the relaxation dynamics of larger molecules.

2. Theoretical background

It is a well-known fact that the time-domain relaxation function has the meaning of a survival probability of the nonequilibrium initial state of the system [3, 19–23]. Its value $\phi(t)$ is equal to the probability that the irreversible transition of the system as a whole from its initial state will take place after the time instant t ; that is,

$$\phi(t) = \Pr(\tilde{\theta} \geq t) \quad (4)$$

where $\tilde{\theta}$ is a waiting time of the system for the transition from the initial state. The waiting time $\tilde{\theta}$ of the system as a whole is determined by the first passage of the system from the initial state [5, 9] and hence equal to

$$\tilde{\theta} = \min(\theta_{1N}, \dots, \theta_{NN})$$

where N denotes the size of the system and the θ_{iN} , $i = 1, \dots, N$, are waiting times for the transitions of particular objects taking part in the relaxation process. The distribution of the waiting time θ_{iN} of the i th object may depend on both short- and long-range interactions. In the model presented, the dependence is expressed in terms of the conditional distribution [6]

$$\Pr(\theta_{iN} \geq t | \beta_i = b, \eta_{i,N} = s) = \exp\left(-\frac{b}{A_N} \min\left(t, \frac{s}{K}\right)\right). \quad (5)$$

Here the positive random variable β_i is the relaxation rate representing the influence of the local environment on the i th object, while

$$\eta_{i,N} = a_N^{-1} \max(\eta_1^i, \dots, \eta_{i-1}^i, \eta_{i+1}^i, \dots, \eta_N^i)$$

where η_j^i is the time that the i th object waits for to get information about the relaxation process of the j th one. This information can influence the process for the i th object, so $\eta_{i,N}$ describes the long-range interactions. The parameters A_N , a_N and K are positive constants. We assume additionally that the waiting times θ_{iN} , as well as the relaxation rates β_i and the times η_j^i , form sequences of independent and identically distributed random variables and that the families $\{\beta_i\}$ and $\{\eta_j^i\}$ are independent of each other. Note that as a consequence of (5) one gets

$$\Pr(\theta_{iN} = \infty) = \left\langle \exp\left(-\frac{\beta_i \eta_{i,N}}{K A_N}\right) \right\rangle \neq 0$$

which reflects the fact that some objects do not undergo the transition (i.e. are somehow ‘frozen’ in the initial state) or that sometimes the initial state is an equilibrium state for the object. (Here $\langle \cdot \rangle$ denotes the expected value.)

If we consider a relaxing system of large size N (in practice, it is enough to take $N \sim 10^7$), the relaxation function $\phi(t)$ in equation (4) is approximately equal to the following first-passage limit:

$$\phi_{FP}(t) := \lim_{N \rightarrow \infty} \Pr(\min\{\theta_{1N}, \dots, \theta_{NN}\} \geq t). \quad (6)$$

At first sight the probability considered in (6) seems to be arbitrarily close to 0 for large N . However, the waiting times θ_{iN} satisfying (5) may significantly alter with increase of the system size N and therefore the limit in (6) can provide a nontrivial relaxation function for some specific choices of sequences β_i , η_j^i and the parameters of the model. Moreover, on the basis of limit theorems of probability theory the possible nontrivial forms of the limit $\phi_{FP}(t)$

in equation (6), as well as the conditions (for the β_i , η_j^i and the parameters) leading to them, are strictly determined [10]. Among the formulae obtained, only

$$\phi_{FP}(t) = \exp\left(-\frac{1}{k} \int_0^{k(t/\tau)^\alpha} \left(1 - \exp\left(-\frac{1}{s}\right)\right) ds\right) \quad (7)$$

with parameters $\tau, k > 0$ and $0 < \alpha < 1$ can be chosen to describe the supercooled (equilibrium) systems since (as was shown in [6]) the relaxation function $\phi_{FP}(t)$ given by (7) is the only one which tends to 0 as $t \rightarrow \infty$ and also has asymptotical behaviour of the form of two fractional power laws in time:

$$-\frac{d\phi}{dt} \propto \begin{cases} (t/\tau)^{-n} & \text{for } t \ll \tau \\ (t/\tau)^{-m-1} & \text{for } t \gg \tau \end{cases}$$

with power-law coefficients equal to

$$n = 1 - \alpha \quad \text{and} \quad m = \frac{\alpha}{k}.$$

In the case $k > \alpha$ one gets that $0 < n, m < 1$ and, hence, that the susceptibility function $\chi_{FP}(\omega)$, associated with relaxation function (7) by means of relation (2) satisfies the two-power-law condition (3) in frequency [8]. So, the asymptotical behaviour of $\chi_{FP}(\omega)$ is like that of the HN function, one of the most popular empirical functions used to fit the response of supercooled systems [24]. $\chi_{FP}(\omega)$ and the HN functions were compared numerically in [25] and it appeared that their shapes resemble each other, so $\chi_{FP}(\omega)$ should fit reasonably well any susceptibility data that can be represented by the HN formula. Consequently, $\phi_{FP}(t)$ of the form (7) is worth taking into considerations as a fitting function in the case of time-domain experiments; the more so since applying the HN method in this case needs some special, rather complicated numerical procedures (see e.g. [26, 27]) while formula (7) can be used in a more direct way. Let us add that the model presented (originally introduced to describe dielectric relaxation [6]) was adopted recently to explain the scaling laws observed in financial data and formula (7) was successfully used to fit positive daily returns of different indices [28], which shows its wide application.

The conditions for relaxation rates β_i and waiting times η_j^i providing the form (7) of the limit in (6) can be formulated as the following scaling properties [6]:

$$\lim_{b \rightarrow \infty} \frac{\Pr(\beta_i \geq xb)}{\Pr(\beta_i \geq b)} = x^{-\alpha_1} \quad \text{for any } x > 0 \quad (8)$$

and

$$\lim_{s \rightarrow \infty} \frac{\Pr(\eta_j^i \geq xs)}{\Pr(\eta_j^i \geq s)} = x^{-\alpha_2} \quad \text{for any } x > 0 \quad (9)$$

for $0 < \alpha_1, \alpha_2 < 1$ such that $\alpha_1 = \alpha_2 = \alpha$. Moreover, one obtains the following relations for the parameters: $k = K^\alpha$, $A_N = N^{1/\alpha} L(N)$ with a function L slowly varying at ∞ [13] and $a_N = \inf\{t: \Pr(\eta_j^i \geq t) \leq 1/(N-1)\}$.

One should add that the case of different exponents α_1 and α_2 in conditions (8) and (9) was discussed in [6] and it was shown that the limiting function obtained in this case either tends to some positive value at ∞ or does not possess the long-time (low-frequency) power-law property. Therefore this case does not relate to the supercooled systems.

The parameter K in (5), and so $k = K^\alpha$ in (7), reflects the intensity of the influence of long-range interactions. It is easy to derive analytically that as the parameter k tends to 0, formula (7) gives KWW function (1) with $\gamma = \alpha$ and $\tau_{KWW} = \tau$. On the other hand, $k \approx 0$ corresponds to $K \approx 0$ and, consequently, to the right-hand side of (5) of the exponential form $\exp(-bt/A_N)$. The distribution of θ_{iN} does not depend on η_j^i in this case,

so the model presented with parameter K close to 0 can be used to describe systems with negligible intermolecular interactions. Moreover, relaxation function (6), introduced for large systems, can be expressed then in terms of relaxation rates β_i only, namely as

$$\phi_{FP,k=0}(t) = \left\langle \exp(-\tilde{\beta}t) \right\rangle \quad \text{where } \tilde{\beta} = \lim_{N \rightarrow \infty} \frac{1}{A_N} \sum_{i=1}^N \beta_i \quad (10)$$

and it can be shown in the framework of probability theory that the relaxation function in (10) cannot take a form other than

$$\phi_{FP,k=0}(t) = \exp(-(At)^a)$$

for some $A > 0$ and $0 < a \leq 1$. For $a = 1$ the function obtained corresponds to the classical exponential response while for $a < 1$ it can be recognized as KWW function (1) with $\tau_{KWW} = 1/A$ and $\gamma = a$. This latter result, which is in agreement with the analytically obtained limit of function (7) for $k \rightarrow 0$, is a consequence of a nontrivial limit theorem derived in the theory of Lévy-stable probability distributions [13–16] and it appears if (and only if) the scaling condition (8) for the relaxation rates β_i is fulfilled with $\alpha_1 = a$. The classical exponential form ($a = 1$) is the limiting case if the expected value of β_i is finite, which follows from the law of large numbers.

3. Experiment

Two commercial samples of epoxy compounds have been used for this study, namely, the monoepoxide phenyl-glycidyl ether (PGE) produced by Aldrich with molecular weight of $150.18 \text{ g mol}^{-1}$ and the diepoxide diglycidyl ether of bisphenol-A (DGEBA) produced by Shell Company with a molecular weight of 380 g mol^{-1} . The samples were filtered through a $0.22 \text{ }\mu\text{m}$ Millipore filter and distilled into dust-free light scattering round cells (10 mm inner diameter), which were flame sealed afterwards. These samples kept at room temperature for several months showed no trace of crystallization. A Brookhaven BI-9000AT Correlator was used for depolarized photon correlation spectroscopy (PCS) [29] measurements in the range 10^{-6} –10 s. The 90° scattering configuration was exploited for both samples. Back-scattering (180° configuration) measurements were previously taken for the DGEBA sample that were consistent with the 90° ones and the results were reported and discussed in [30]. We use here the 90° configuration since it gives a better signal-to-noise ratio, which is important for the detailed study of the shape of the relaxation function.

Table 1. Relaxation times and stretching parameters calculated by fitting the PGE spectra of figure 1 by means of KWW formula (1).

T (K)	τ_{KWW} (μs)	γ
197.7	17490 ± 40	0.61 ± 0.01
200.6	2927 ± 6	0.61 ± 0.01
203.4	547.4 ± 1.6	0.62 ± 0.01
206.2	125.6 ± 0.8	0.61 ± 0.01
209.0	29.4 ± 0.7	0.61 ± 0.01
211.5	8.6 ± 1.1	0.59 ± 0.04

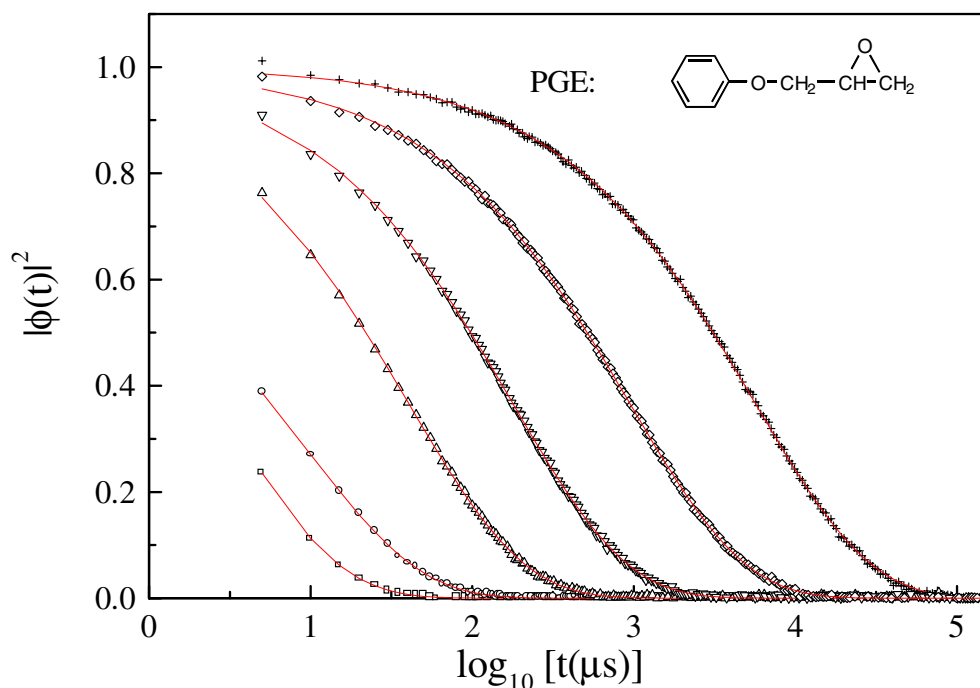


Figure 1. Photon correlation spectra of PGE. The temperatures, from left to right, are 211.5, 209, 206.2, 203.4, 200.6 and 197.7 K. The full lines are obtained by fitting the spectra by means of KWW formula (1). In the inset, the chemical structure of PGE is shown.

4. Results and discussion

Figure 1 shows spectra taken at different temperatures for PGE. The full lines are obtained by fitting the spectra by means of KWW relaxation function (1). The values of the parameters obtained by the fit are reported in table 1. In the present case of DLS spectra of PGE, the KWW function is revealed to be completely adequate to fit the spectra, within the experimental error. This is better shown in figure 2(a) for $T = 203.4$ K, where the log scale emphasizes the small-amplitude long-time behaviour. Moreover, in figure 2(b) the residuals are reported, together with the expected error. The residuals do not show any marked trend as a function of time and their values lie, on average, within the expected error. It is worth noting the utility of the estimated error for checking the goodness of the fit. In the case of PCS measurements, the determination of this error requires particular care, due to the presence of correlated error coming from the photon correlation procedure. For this work, we have adopted the procedure outlined in [31].

For the sake of the comparison, the function of form (7) was also used for fitting the PGE spectrum of figure 2(a) under discussion. We have obtained $\alpha = 0.617$, $\tau = 547$ and $k < 0.1$ with residuals comparable with those of the KWW fitting (see figure 2(c)). Let us note that for such a small value of parameter k , function (7) is close to the KWW function, as was shown in section 2, and hence the two fitting procedures are consistent in indicating the stretched-exponential decay law to be followed by PGE.

In figure 3 the spectra taken from DGEBA are reported at five different temperatures together with the fitting curves of form (7). It has to be stressed that KWW function (1),

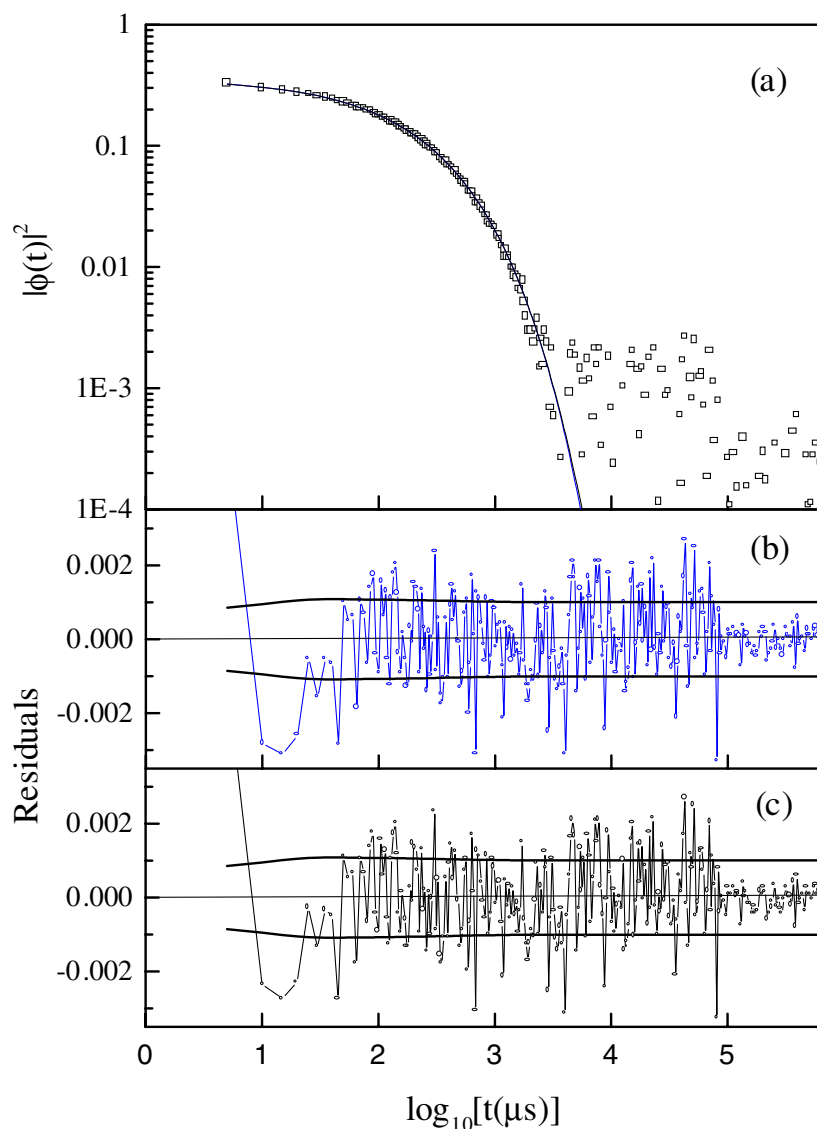


Figure 2. (a) The PCS spectrum of PGE at $T = 203.4$ K, where the log scale emphasizes the small-amplitude long-time behaviour. The full line is obtained by fitting the spectrum by means of KWW formula (1). (b) Residuals of the fit by KWW formula (1) (thin line) together with the expected error (thick lines). (c) Residuals of the fit by formula (7) (thin line) together with the expected error (thick lines).

although it represents the main features of the measured spectra, fails in a more quantitative comparison. In contrast, function (7) is able to represent, within the experimental error, the spectra over the whole time and temperature region investigated. In order to provide evidence of the difference between the fitting functions (1) and (7) discussed here, we have reported in figure 4(a) the spectrum at $T = 267.1$ K on a log scale with both fitting curves. The analysis of the residuals shows a well-defined oscillatory behaviour which reveals that the KWW formula does not adequately represent the response function (see figure 4(b)). Moreover the long-time

part of the spectrum appears to be more stretched than the KWW function, suggesting the presence of the second power-law regime. In this case, function (7), which explicitly takes into account the intermolecular interactions, is more adequate to describe the data (see the residuals plot in figure 4(c)). The parameters of the fit are presented in table 2.

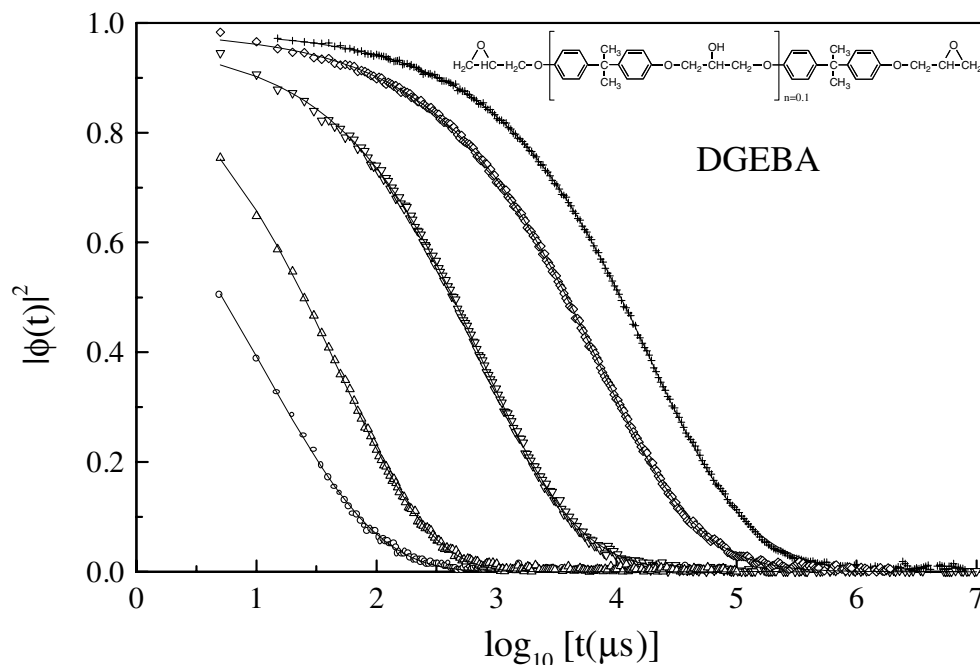


Figure 3. Photon correlation spectra of DGEBA. The temperatures, from left to right, are 274.9, 272.2, 267.1, 262.8 and 261.2 K. The full lines are obtained by fitting the spectra by formula (7). In the inset, the chemical structure of DGEBA is shown.

Table 2. Relaxation parameters of DGEBA calculated by fitting the spectra of figure 3 by means of formulae (1) and (7).

T (K)	τ_{KWW} (μ s)	γ	τ (μ s)	α	$m = \alpha/k$
261.2	80770	0.53	71400 ± 1500	0.57 ± 0.01	0.91 ± 0.02
262.8	30246	0.52	26600 ± 600	0.57 ± 0.01	0.90 ± 0.04
267.1	3000	0.54	2730 ± 70	0.61 ± 0.01	0.98 ± 0.02
272.2	169	0.55	154 ± 9	0.69 ± 0.02	0.98 ± 0.03
274.9	52	0.51	50 ± 12	0.66 ± 0.02	0.95 ± 0.02

The two-power-law response, obtained for DGEBA, is typical of macromolecules and of polymeric systems, where usually the HN empirical function is used to fit the susceptibility data [24, 32]. The HN function is a phenomenological law which was introduced to interpolate the high- and low-frequency power laws experimentally observed in different (electric, mechanical etc) relaxation experiments. The existence of such power laws was tentatively explained in terms of different molecular mechanisms. The more traditional one is that of independent relaxing species with random relaxation rates. A more recent approach [19] has proposed a cluster model where, in a hierarchical scheme, the faster degrees of freedom successively constrain the slower ones. On this basis, models more strictly related to the morphology and

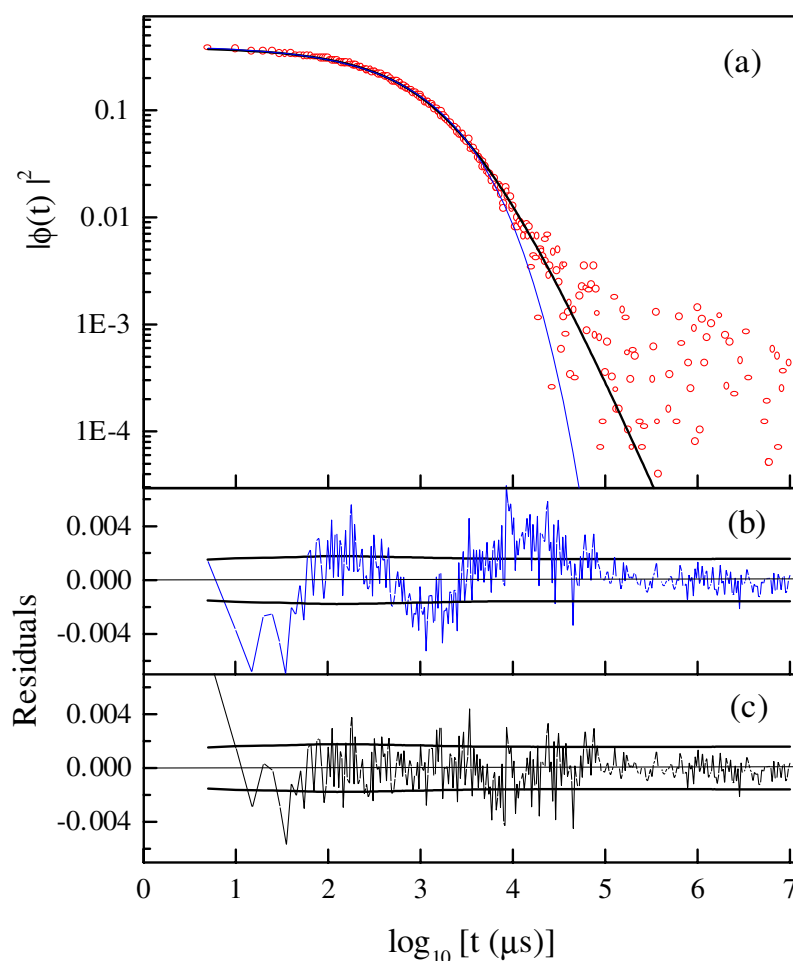


Figure 4. (a) The spectrum of DGEBA at 267.1 K. The thin line is the best fit of the data by KWW function (1). The thick line is the fit by means of formula (7). (b) Residuals of the fit by KWW formula (1) (thin line) together with the expected error (thick lines). (c) Residuals of the fit by formula (7) (thin line) together with the expected error (thick lines).

dynamics of macromolecules and polymers have been also proposed [32].

The probabilistic approach presented in this paper starts from more fundamental properties of disordered systems, considering the effect of the local environment on the relaxation rates β_i , and the intermolecular interactions on the waiting times η_j^i , both being random variables. Two scaling properties, equations (8) and (9), of the distributions of β_i and η_j^i indicate in the case of equal scaling exponents that the relaxation function takes form (7). In the framework of the proposed model, the long-time power-law behaviour of DGEBA can be attributed to the presence of long-range interactions. The origins of stronger intermolecular interactions in DGEBA with respect to PGE can be found in the more complex structure of the first molecule (see the insets of figures 1 and 3), and are probably related to the higher steric hindrance in the rotation of single molecules. A change of short- and long-range interactions is expected to be reflected in a change of the shape parameters α and k . In particular, a reduction of k (increase of the low-frequency exponent $m = \alpha/k$) is expected for increasing temperature,

when the role of interparticle interactions generally reduces. This effect can be seen both for DGEBA, comparing the results obtained by different light scattering techniques [30], and, more generally, for high-molecular-weight glass-forming systems [32]. These results help us to clarify the physical meaning of the coefficients α and k introduced by the theory, even if a more quantitative link between their values and molecular properties has still to be achieved.

In conclusion, light scattering from the two glass formers PGE and DGEBA has been successfully interpreted by means of a probabilistic approach based on the tool of limit theorems of probability theory. The transition from the KWW to the two-power-law behaviour has been evidenced for increased molecular complexity, which, in the proposed theory, has been related to an increase of long-range interaction.

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