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A study of α -relaxation in ortho-terphenyl by photon correlation spectroscopy

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Abstract. We studied the α -relaxation in ortho-terphenyl (OTP) by photon correlation spectroscopy (PCS) at temperatures ($250\text{ K} < T < 270\text{ K}$) below the crossover temperature T_c . We found the following. Contrary to the mode-coupling theory prediction, the non-ergodicity parameter f_c is temperature independent, which might be due to a strong orientational scattering in OTP. The stretched exponent β_k decreases as temperature decreases, and the relaxation time $\tau(T)$ obeys the Vogel–Fulcher law. In the short-time limit of PCS measurement ($t \sim 0.1\ \mu\text{s}$), no decaying process (neither the Johari–Goldstein β -relaxation nor the critical decay) was observed.

1. Introduction

The dynamics of molecules in a supercooled liquid is an interesting topic in condensed matter physics. Recently, mode-coupling theory (MCT) [1] has been developed to give a detailed description of the dynamics in a supercooled liquid. The hallmark of MCT is the prediction of β - and α -relaxations in a system's relaxation functions or susceptibility spectra. The β -relaxation is associated with a short timescale, and is related to the local dynamics. On the other hand, the α -relaxation is associated with a long timescale, and corresponds to the overall structural rearrangement of a system. The β -relaxation consists of two power-law-decay regions which show critical decay ($\sim t^a$) and Von Schweidler decay ($\sim t^{-b}$), and the two exponents a and b are related to the system-dependent exponent parameter λ by gamma function relations, $\lambda = \Gamma(1 - a)^2 / \Gamma(1 - 2a) = \Gamma(1 + b)^2 / \Gamma(1 + 2b)$. At the boundary of the β - and α -relaxations, the Von Schweidler decay is smoothly connected to the Kohlrausch–Williams–Watts (KWW) α -relaxation region where the relaxation is usually expressed by a stretched-exponential form, $f_c \exp(-(t/\tau)^{\beta_k})$, with three parameters. These are the non-ergodicity parameter f_c , the stretched exponent β_k , and the relaxation time τ . In the MCT, the crossover temperature T_c is defined as the temperature at which the transport properties of the system change from those for a strongly coupled liquid to those for a glass. Typically for a fragile glass-forming material, T_c is located between the calorimetric glass transition temperature T_g and the melting temperature T_m . The β -relaxation can be described by the scaled β -correlator both for $T < T_c$ and for $T > T_c$. However, the nature of the α -relaxation is quite different, depending on whether T is above or below T_c . When $T < T_c$, the MCT predicts that the non-ergodicity parameter will increase as $\sim (T_c - T)^{1/2}$. However, there

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is no prediction concerning the temperature dependence of β_k . When $T > T_c$, β_k and f_c^q are predicted to be temperature independent because of the time–temperature superposition principle.

Among glass-forming materials, ortho-terphenyl (OTP) is a good subject to study if one wishes to investigate the dynamics of a supercooled liquid because of its high glass transition temperature $T_g \simeq 243$ K [2]. With the aim of achieving an understanding of the α -relaxation in a glass-forming material, several experiments have been performed using different experimental techniques, which include dielectric spectroscopy [3], light scattering [4, 5], neutron scattering [6, 7], molecular dynamics simulations [8], and photon correlation spectroscopy (PCS) [4, 9, 10]. However, the results from different experiments are not unanimous, and no concrete conclusion has been reached yet.

In this paper, we report a PCS study of OTP for $T_g < T < T_c$. We found that the polarized component of the density correlation function contains an additional slow relaxation. Using the depolarized component, we found the following. Contrary to the MCT prediction, the non-ergodicity parameter was temperature independent. We speculate that this is due to a strong orientational scattering in OTP. The stretched exponent β_k decreases as the temperature decreases. In the temperature range considered in this study, $\tau(T)$ obeys the Vogel–Fulcher law with $T_0 \sim T_g - 60$ K. In the short-time limit of PCS measurement, no decaying process (neither the critical decay in MCT nor the Johari–Goldstein slow β -relaxation) was identified, even at temperatures below T_g .

2. Experiment

The ortho-terphenyl (OTP) used in this study was prepared in our laboratory. The 99% pure OTP was purchased from Aldrich Chemicals. To avoid crystallization due to impurities, the as-received OTP was filtered into a glass sample tube with a 15 mm diameter, and was kept under vacuum overnight before being flame sealed. The sealed sample was contained in a copper sample holder which is designed to be attached to the cold finger of an Oxford cryostat. The temperature range ($250 \text{ K} \leq T \leq 270 \text{ K}$) considered in this study was obtained using liquid nitrogen and an Oxford intelligent temperature controller (ITC-4) with 0.1 K precision.

A Spectra Physics Ar⁺-ion laser ($\lambda = 488$ nm) was used as the light source, with input power ~ 100 mW. The vertically polarized incident light was focused onto the OTP sample by lenses. The scattered light at 90° was collected by a micro-lens which is attached in front of a fibre-optical cable. Then the scattered light was transmitted to two photomultiplier tubes (PMT) by a fibre-optical cable. To avoid strong fluorescence light, a narrow-band ($\lambda = 488 \pm 10$ nm) blue filter was used before the scattered light was collected. The polarized (VV) and the depolarized (VH) components of the scattered light were selected by a Glan–Thomson analyser with a leakage factor of less than 1%. The scattering wavenumber q used in the present study was fixed as $q \sim 2.4 \times 10^5 \text{ cm}^{-1}$ ($\theta = 90^\circ$). The signal from the PMT was measured by the Brookhaven BI-9000AT digital correlator which can cover ten decades of time ($10^{-1} \mu\text{s} - 10^9 \mu\text{s}$). The finite size of the time window in our correlator limits the measurable temperature range to 250 K–270 K. The crossover temperature T_c of OTP is known to be ~ 290 K [6].

From the measured correlation function $G(t)$, the intensity–intensity correlation function $g_2(t)$ can be calculated. $g_2(t)$ is related to the intermediate-scattering function $\phi_q(t)$ by

$$\frac{G(t)}{G(\infty)} = g_2(t) = 1 + A|\phi_q(t)|^2 \quad (1)$$

where A is the instrumental coherent factor. $\phi(t)$ (since q is fixed in this study, the subscript

q is omitted), at different temperatures, was fitted to the stretched-exponential expression $\phi(t) = f_c \exp(-(t/\tau)^{\beta_k})$.

In our PCS measurement, several features are new and different from the case for a typical photon correlation apparatus. First, the use of fibre optics eliminates the pinholes and lenses. By virtue of this method, we can get an instrumental coherence factor of $A \sim 0.97 \pm 0.01$ with an aqueous suspension of polystyrene spheres. Second, the short-time relaxation was measured more reliably by measuring a cross correlation using two PMTs. The advantage of measuring cross correlation is that we can avoid the after-pulsing effect, which can be easily misunderstood as a decaying process in short-time PCS data.

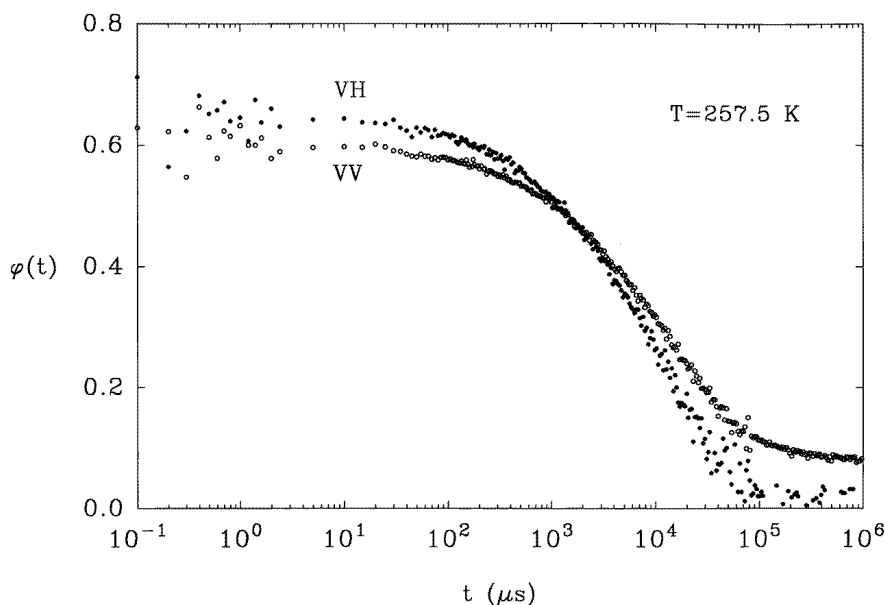


Figure 1. A semi-log plot of the intermediate-scattering function $\phi(t)$ versus t : VV (open circles) and VH (closed circles) components at the temperature $T = 257.5$ K.

3. Results and discussion

In a typical light scattering mechanism [11], density fluctuations, orientational fluctuations, and an interaction-induced scattering contribute to the polarized (VV) scattered light. On the other hand, only orientational fluctuations and an interaction-induced scattering contribute to the depolarized (VH) scattered light. In the low-frequency limit (typically less than 1 GHz), the scattered light mainly consists of the orientational scattering and interaction-induced scattering. Since the measurement time range in PCS corresponds to a low-frequency region (from 0.1 μ s to 1000 s), it is expected that the VV and VH components of $\phi(t)$ are the same unless there is an impurity or a cluster in the OTP sample. In our PCS measurement, however, as can be seen in figure 1, the VV and VH components of $\phi(t)$ appear to be different. The VV component of $\phi(t)$ does not decay to zero in the long-time limit, indicating that there is another slow relaxation. Because of the relaxation, the VV component of $\phi(t)$ is more stretched than the VH component of $\phi(t)$. In the VH component, there is no sign of an additional relaxation.

We also tested the scattered intensity during the PCS measurement. Figure 2 shows the

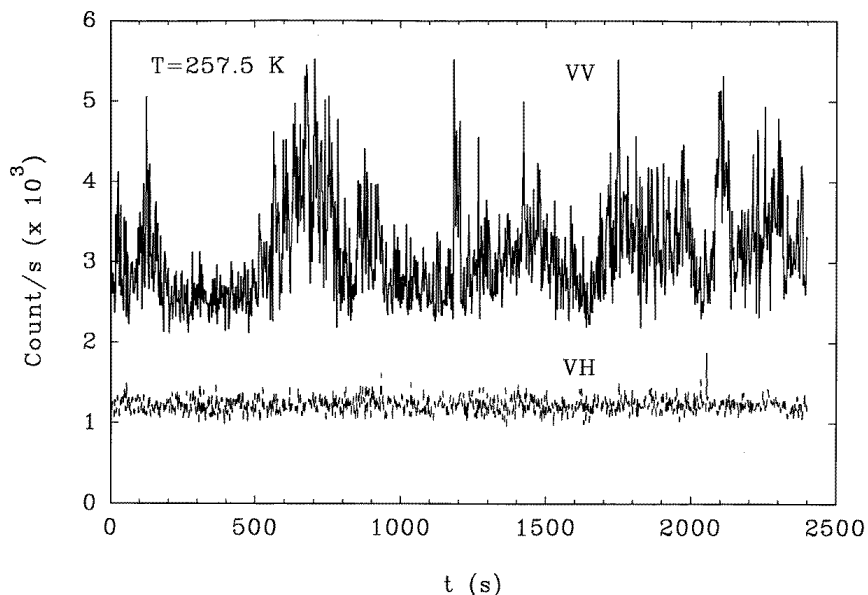


Figure 2. The count rate history of the VV (solid trace) and VH (dotted traces) components in the PCS experiment. The temperature for this measurement is the same as for figure 1.

count rate history for the PCS measurement (40 minutes) for both the VV and VH components. There are some differences between the VV and VH components. Firstly, the average count rate for the VH component is about a factor of 2 smaller than that for the VV component. This is to be expected, since the scattered intensity in the VH component is usually weaker than that in the VV component. Also, this is why the VH component of $\phi(t)$ in figure 1 looks noisier. Secondly, the count rate for the VV component fluctuates a lot, while the count rate for the VH component is more or less constant. At different temperatures, the overall differences between the VV and VH components are the same, but the amplitude of the fluctuations in the VV component gets bigger as the temperature decreases.

We can interpret these observations in two different ways. It could be that there are correlated domains which increase their size as the temperature decreases. On the other hand, it could be that there are impurities in the sample. The latter case is less probable, since no sparkling spot in our sample was identified during the measurement, and our sample was prepared with special care. An additional slow relaxation in the VV component was also observed in another PCS study of OTP, by Patkowski *et al* [12]. They explained that this additional slow-relaxation mechanism was due to the ‘dynamic clustering’ of the OTP sample. Our observation might support the idea of ‘dynamic clustering’. However, to confirm this, more careful inspection will be necessary.

Since the VV component appears to contain another slow relaxation, we focused our study on the VH component. Figure 3 shows $|\phi(t)|^2$ for different temperatures. The solid curves in this figure are the fits to the stretched-exponential form $|\phi(t)|^2 = f_c^2 \exp[-2(t/\tau)^{\beta_k}]$ with three fitting parameters, f_c , τ , and β_k . Instead of $\phi(t)$, $|\phi(t)|^2$ was used in the data fitting for the following reason. In the correlation data, $G(t)/G(\infty) - 1$ fluctuates around zero at long-time limits. To estimate $\phi(t)$, the square root of $G(t)/G(\infty) - 1$ is taken. Since taking a square root will remove all negative data, the data fitting at long-time limits will be biased. In our fitting process, the partial data range ($t \geq 10 \mu s$) was considered to avoid short-time

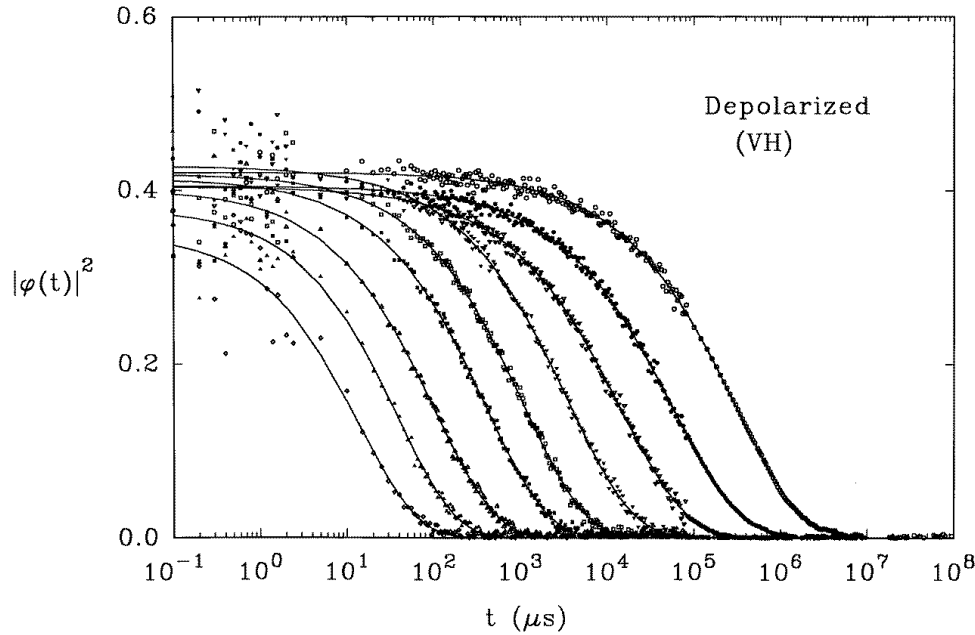


Figure 3. The VH component of the intermediate-scattering function over the temperature range $T = 250$ – 270 K. Different symbols represent different temperatures: $T = 250$ (open circles), 252.5 (closed circles), 255 (inverted open triangles), 257.5 (inverted closed triangles), 260 (open squares), 262 (closed squares), 265 (open triangles), 267.5 (closed triangles), and 270 K (open circles). The solid curves are fits to the stretched-exponential expression $\phi_q(t) = A \exp(-(t/\tau)^\beta)$.

noisy data. The difference between the results obtained using complete data and partial data was checked, and it appeared to be negligible for temperatures $T \leq 267.5$ K. However, the difference was quite big at the temperature $T = 270$ K, because we could not measure the early part of $\phi(t)$. As a result, at $T = 270$ K, partial data fitting gave us an unusually large f_c -value. Only for $T = 270$ K was the entire data range used to get the fitting parameters.

The temperature dependence of f_c is considered first. Figure 4 shows f_c for different temperatures. The function $f_c(T)$ obtained by the Mainz group is also plotted in this figure. The f_c -value obtained by the Mainz group is a little higher than our f_c . But within statistical errors, the two measurements are mutually consistent. We found that f_c is temperature independent, with the average values $\langle f_c \rangle = 0.639 \pm 0.020$ for our data and $\langle f_c \rangle = 0.679 \pm 0.020$ for the Mainz group's data. However, according to the MCT prediction, the non-ergodicity parameter is expected to increase as $(T_c - T)^{1/2}$ as the temperature decreases, because the temperature range used in this study was below T_c . Also, in other PCS studies of OTP [4, 9], no temperature dependence of f_c was observed over the temperature range $250 \text{ K} \leq T \leq 270 \text{ K}$. In an earlier PCS study [10], very small increases of f_c were observed when $T \leq 265$ K. But the amount of increase could be negligible. On the other hand, in neutron scattering [6] and in molecular dynamics simulation [8] studies, a square-root temperature dependence of f_c was observed. The observed temperature independence of f_c for PCS could be due to the small temperature range. To test this possibility, we compared the change in absolute value in different experiments over the same temperature interval. When the temperature changed from 290 K to 250 K, f_c increased from 0.7 to 0.9 in the neutron scattering study and from 0.3 to 0.5 in the molecular dynamics simulation study. In both studies, f_c increased by about

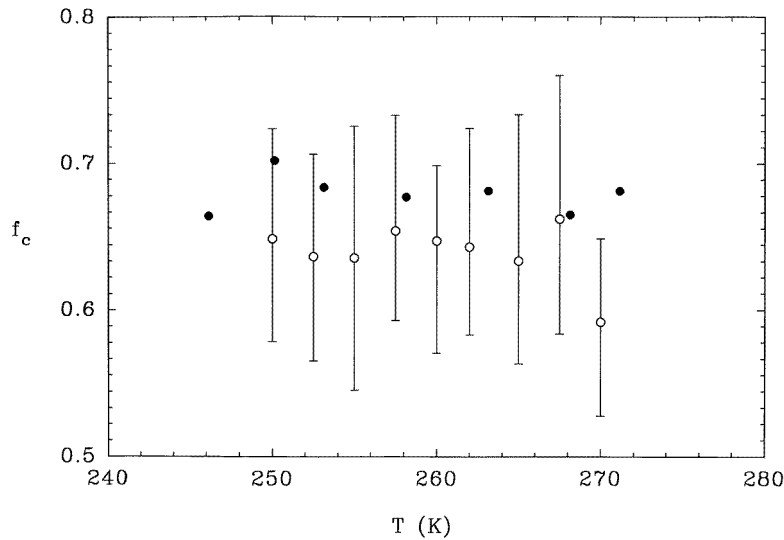


Figure 4. The temperature dependence of the non-ergodicity parameter from our PCS study (empty circles) and that obtained by the Mainz group (closed circles).

30–40%, with $\Delta t \sim 40$ K. However, in our PCS study the change of f_c was at most 10% (from 0.6 to 0.65), which is much less than the changes in the neutron scattering and molecular dynamics simulation studies. Therefore, it can be concluded that no square-root temperature dependence of f_c at $T < T_c$ was observed in the PCS measurement.

We may explain this observation in the following way. The time range used in a typical PCS measurement is much longer than a nanosecond. In other words, the frequency range in PCS measurements is much less than 1 GHz. As we mentioned earlier, in this frequency region the main scattering mechanisms for both VV and VH components are the orientation scattering and the interaction-induced scattering. In OTP, if the orientational scattering is stronger than the interaction-induced scattering, then the temperature dependence of f_c can be different from the MCT prediction. Recently, we estimated the relative importance of the orientational scattering and the interaction-induced scattering in the depolarized scattered intensity of Salol [13], and found that the orientational scattering is 30 times stronger than the interaction-induced scattering. Therefore, for OTP also, the orientational scattering can be a dominant scattering mechanism in the depolarized scattered light. To confirm this, we are planning to estimate the relative importance of the orientation scattering and the interaction-induced scattering in OTP.

To study the temperature dependence of the stretched exponent β_k , we plot $\beta_k(T)$ together with data from different experiments in figure 5. In this figure, $\beta_k(T)$ for the high-temperature region is the result from our depolarized light scattering study [5] in which the temperature dependence of β_k was obtained by analysing the α - and β -relaxation simultaneously using extended MCT [5]. In our light scattering data, β_k is temperature independent for $T > 300$ K ($T_c \sim 290$ K) which is consistent with the MCT time–temperature superposition principle. In another light scattering study of OTP by Steffen *et al* [4], however, β_k increases as temperature increases, and approaches unity at the high-temperature limit. This discrepancy may stem from the different ways of analysing the data. We believe that our method is more reliable for the following reason. The α -relaxation in susceptibility spectra cannot be analysed independently, since the high-frequency part of the α -relaxation peak is strongly affected by the β -relaxation.

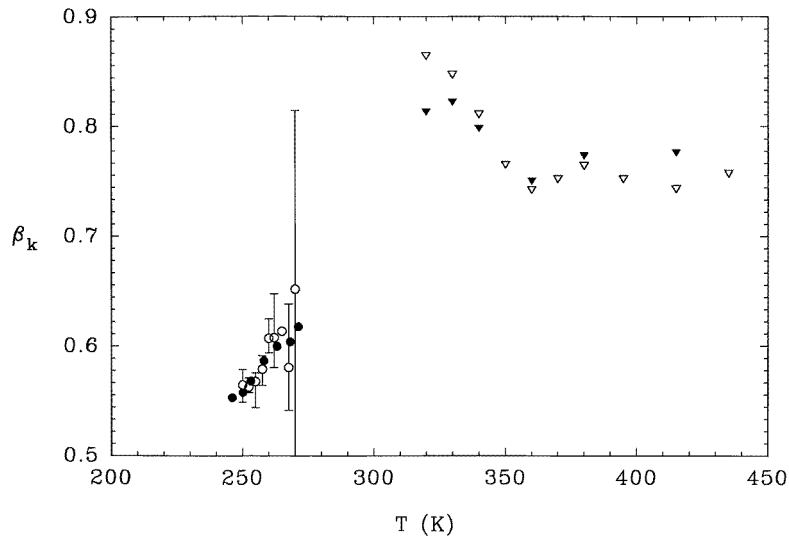


Figure 5. The temperature dependence of the stretched exponents β_k from different experiments. Closed circles, empty circles, and empty triangles represent data obtained from PCS by the Mainz group, from our PCS, and from depolarized light scattering, respectively.

As we found by analysis in our light scattering study [5], both relaxation types have to be analysed together to get a reliable $\beta_k(T)$. More detailed comparison between the two different data analysis methods is given in [14].

For temperatures less than 300 K, β_k decreases as temperature decreases. The data obtained by the Mainz group show the same trend in this temperature region. Considering the whole temperature range, the overall temperature dependence of β_k can be obtained. β_k is temperature independent when $T > T_c$, and decreases as temperature decreases when $T < T_c$. The same temperature dependence of β_k can also be observed for other glass-forming materials such as Salol [15], propylene carbonate [16], and CaKNO_3 [17]. In the case of Salol, as can be seen in figure 13 in reference [15], β_k was temperature independent when $T \geq 280$ K, and then decreased as T decreased when $T < 280$ K (T_c for Salol is ~ 260 K). In the cases of PC and CaKNO_3 , again the same temperature dependence of β_k was observed, i.e. β was temperature independent when $T > T_c$ and decreased as the temperature decreased when $T < T_c$ [16, 17]. As a result, we conclude that for fragile glass-forming materials, β_k is temperature independent for $T > T_c$ and decreases as the temperature decreases for $T < T_c$.

Now, we consider the temperature dependence of τ . $\tau(T)$ from the present study is plotted in figure 6. Usually, the temperature dependence of the relaxation time can be described by the Arrhenius law at low temperatures and the Vogel–Fulcher law at high temperatures. These two different temperature regions are separated by the Fisher–Stegel temperature T_{FS} , which appears to be very close to T_c [18]. We tried to fit our $\tau(T)$ with both the Arrhenius law, $\tau(T) = \tau_0^A \exp(E/kT)$, and the Vogel–Fulcher law, $\tau(T) = \tau_0^{VF} \exp(T_{VF}/T - T_0)$. From the Vogel–Fulcher fit, we found $T_0 \simeq 180$ K which is about 60 K below T_g . From the Arrhenius fit, we found $\tau_0^A = 3.25 \times 10^{-48}$ s, and $E = 4.7 \times 10^{-12}$ erg. As can be seen in figure 6, the Vogel–Fulcher law fits the data better than the Arrhenius law does over the temperature region $250 \leq T \leq 270$ K. This is different from what we expected, since the temperatures of this measurement are less than T_c . However, we do not want to lay too much stress on this result, since our relaxation time measurement only covers six decades of time, and the quality of the

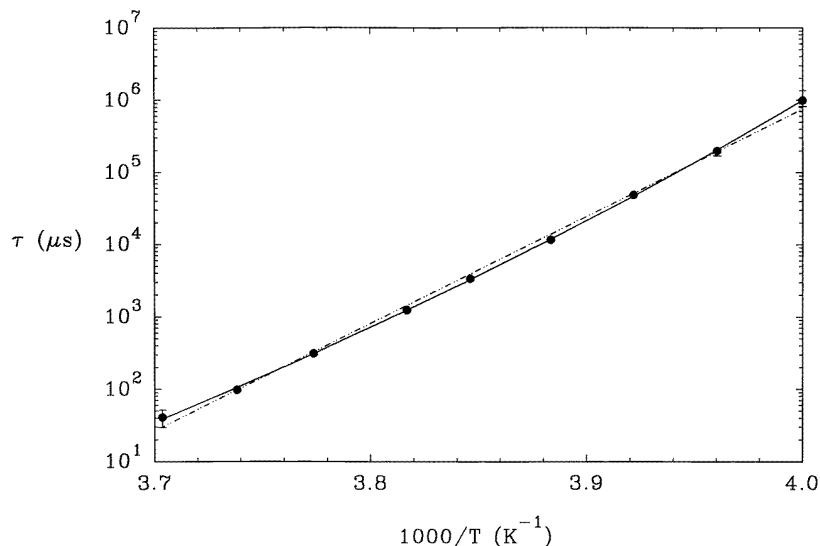


Figure 6. Relaxation times versus T . The dashed-double-dotted line is a fit to the Arrhenius law, and the solid curve is a fit to the Vogel-Fulcher law.

fit with the Arrhenius law is also good.

Now, let us move our focus to the boundary of the β - and α -relaxations. In the MCT, the β -relaxation precedes the α -relaxation, and consists of the critical decay and the Von Schweidler decay. These two decays are separated by the inflection point t_σ , where the non-ergodicity of a system is determined as $\phi(t_\sigma) = f_c$. The critical decay is related to $t < t_\sigma$ and decay up to t_σ . The Von Schweidler decay is related to $t > t_\sigma$ and is connected to the Kohlrausch-Williams-Watts (KWW) stretched-exponential relaxation region.

Recently, Torell *et al* [19] reported that some other relaxation occurs before the α -relaxation in a PCS study of B_2O_3 . This could be either a tail of the critical decay in the MCT or the Johari-Goldstein slow β -process. About 30 years ago, Johari and Goldstein observed another relaxation (secondary relaxation) which was faster than the α -relaxation in their dielectric spectroscopy study of several glass-forming materials (OTP, CKN, etc) [3]. The secondary-relaxation peak, at around 10 kHz–1 MHz, shows the Arrhenius temperature dependence. Nowadays, this process is known as the Johari-Goldstein (J-G) β -relaxation or the slow β -relaxation to distinguish it from the MCT β -relaxation. The origin of the J-G slow β -relaxation is not yet clear, but it is generally accepted that the source of the slow β -process is the branch motion in complex molecules, since this process can be relatively easily observed in complex materials.

There has been a question as to whether a short-time relaxation (either the critical decay or the J-G β -relaxation) can be observed in PCS experiments. We tried to answer this question in our PCS study. First, we would like to note a few things which are useful for short-time PCS data analysis. When we analyse the data based on the MCT, a $\log |\phi(t) - f_c|$ versus $\log t$ graph is used [19] to identify two power-law regions. From the two linear regions in the log-log plot, two exponents a and b are extracted and are tested to see whether they obey the exponent relation predicted in the MCT. However, there could be a problem in doing the data analysis this way. Since the log is taken after taking the absolute value of $\phi(t) - f_c$, the fluctuation of $\phi(t)$ around f_c is flipped over and takes only positive values. This flipping makes the noisy

short-time $\phi(t)$ obey a power-law decay such as that of the critical decay. Therefore, it is important to test for the existence of an additional relaxation before taking the absolute value of $\phi(t) - f_c$. In addition, it is also possible to misinterpret short-time correlation data caused by the after-pulsing effect from a photomultiplier tube as a power-law decay. As mentioned earlier, since our PCS apparatus measures cross correlation to avoid the after-pulsing effect, we can test for the existence of a short-time relaxation in a PCS measurement more reliably.

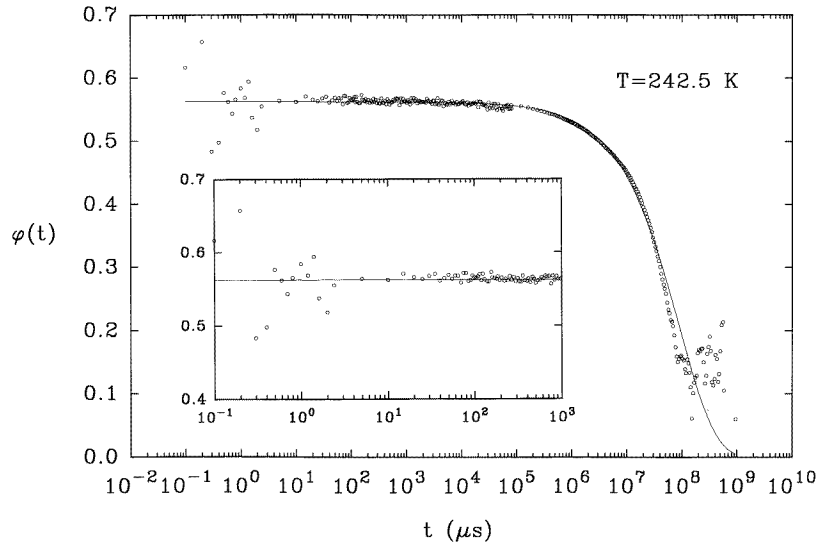


Figure 7. The intermediate-scattering function at the temperature $T = 242.5$ K. The solid curve is the fitting result obtained using the data for $t \leq 10 \mu\text{s}$. Inset: the short-time data fitted with the critical decay power law $\phi(t) - f_c = h(1/t)^a$ with the fitting parameters h and a .

Since we are more likely to observe the short-time relaxation in density correlators at low temperatures, we measured $\phi(t)$ at $T = 242.5$ K which is very close to T_g , and the results are shown in figure 7. Since the relaxation time at this temperature is close to the limit of our measurement window, fully relaxed $\phi(t)$ was not observed. In view of this, the fitting range was chosen as $t \geq 100 \mu\text{s}$ to avoid biased data analysis. As can be seen in figure 7, $\phi(t)$ fluctuates around f_c in the short-time region, but there is no sign of a short-time relaxation. We tested the possibility of observing the tail of the MCT β -relaxation. We fitted data with the power law $\phi(t) = f_c + h(1/t)^a$, with fixed $f_c = 0.623$. The fitting results are also shown, in the inset of figure 7. The fitting parameters h and a appear to be of the order of 10^{-10} , indicating that no power-law regions exist in PCS data. From the above observations, we conclude that we do not see any short-time relaxation in our PCS data.

For a more comprehensive study of the α -relaxation, it is important to study both α - and β -relaxations simultaneously. However, it is difficult to measure both β - and α -relaxations by either PCS or interferometric spectroscopy when the border of the two relaxations lies between ~ 1 ns (1 GHz) and $\sim 0.1 \mu\text{s}$ (10 MHz). In PCS measurements, the smallest accessible timescale is of the order of $0.1 \mu\text{s}$. In a typical light scattering experiment (e.g., with a Tandem interferometer), the smallest accessible frequency range is of the order of 1 GHz. Therefore, PCS and light scattering experiments share the difficulty of measuring both β - and α -relaxations simultaneously. To explore this hidden range of time (frequency), we are planning to use a confocal interferometer in a future study.

4. Conclusions

We studied the α -relaxation of glass-forming ortho-terphenyl by photon correlation spectroscopy at $T < T_c$. We performed a more reliable measurement by means of PCS using cross correlation and fibre-optical cables. The polarized (VV) and the depolarized (VH) components of the scattered light were measured for a 90° scattering geometry. We found that an additional slow relaxation exists in the VV component, possibly due to the clustering of samples. The temperature dependence of f_c , β_k , and τ was studied using the VH component. Contrary to the MCT prediction, no square-root temperature dependence of f_c was observed, i.e. $f_c(T)$ was temperature independent with the average value $\langle f_c \rangle = 0.639 \pm 0.02$. We suggest that this might be due to a strong orientational scattering in OTP. To obtain a more reliable temperature dependence of β_k , we compared our $\beta_k(T)$ with other experimental results, and found that β_k is constant and decreases as the temperature decreases at $T > T_c$ and $T < T_c$, respectively. Over the temperature range $250 \text{ K} < T < 270 \text{ K}$, the relaxation time can be described by the Vogel–Fulcher law with $T_0 \simeq T_g - 60 \text{ K}$. We also found that no additional short-time relaxations exist in our PCS experiment.

Acknowledgments

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