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Coherent dynamic structure factor of orthoterphenyl around the mode coupling crossover temperature T_c

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The coherent dynamic structure factor of supercooled orthoterphenyl has been measured by time-of-flight neutron scattering. The data extend previous backscattering and spin-echo data towards shorter times. In the β relaxation regime the two-step decay of density correlations can be described by the same master function as found before for self-correlations. We show that the mode-coupling crossover temperature T_c can be determined from β relaxation data alone. The wave-number dependence of the relaxation amplitude follows, to some extent, that of the static structure factor. [S1063-651X(97)11107-2]

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I. INTRODUCTION

The molecular van der Waals liquid orthoterphenyl (OTP) is one of the best studied glass formers. It is representative of a whole class of glasses whose characteristic is an extremely non-Arrhenius behavior of the shear viscosity $\eta(T)$ [1]. The dynamics of viscous OTP was studied by many experimental methods, e.g., light scattering [2], dielectric relaxation [3], frequency-dependent specific heat [4], nuclear magnetic resonance [5], computer simulation [6], and reorientation measurements of host molecules [7].

In particular OTP has become a model system for tests of the mode-coupling theory (MCT) of the glass transition [8,9]. OTP consists of relatively small molecular units which interact only weakly via nondirectional van der Waals forces; intramolecular degrees of freedom seem to play no role on time scales that are relevant for the glass transition dynamics [10,11]. In a series of mainly incoherent neutron-scattering experiments it was possible to verify, even quantitatively, several main predictions of idealized MCT [12–14] and to determine a crossover temperature $T_c = 290 \pm 5$ K.

While incoherent scattering measures tagged-particle motion, additional information on interparticle correlations can be gained by coherent scattering from a deuterated sample. First measurements of deuterated OTP at a spin-echo and a backscattering spectrometer [15] confirmed that density correlations $\Phi(Q,t)$ decay in two steps and can be described by the same scaling laws as found for self-correlations. Around the intermediate plateau, $\Phi(Q,t)$ was compatible with the asymptotic law

$$\Phi(Q,t) = f_Q + H_Q g_\lambda(t/t_\sigma). \tag{1}$$

Using β relaxation times t_σ derived from the incoherent data as an input, the amplitude H_Q could be shown to follow the predicted power law and to extrapolate consistently to $T_c \simeq 290$ K. In an alternative approach, the Debye-Waller factor f_Q was determined from a scaling analysis of structural α relaxation or, below T_c, from the unresolved elastic peak alone. The onset of the fast localized β process showed up as a cusp in $f_Q(T)$ which could be fitted by the square root law $\sim (T_c - T)^{1/2}$, confirming again $T_c \simeq 290$ K. However, the Q resolution was insufficient to resolve the wave-number dependence of parameters.

In the framework of MCT the Q dependence of f_Q and H_Q is determined solely by the static properties of the system: The coupling coefficients are written in terms of the static structure factor $S(Q)$. So far they have been calculated for systems like hard spheres [16], Lennard-Jones spheres [17], and binary mixtures of hard spheres [18]. In these simple liquids f_Q and H_Q are found to oscillate in phase and out of phase with the static structure factor $S(Q)$, respectively; the line-shape parameter λ and the crossover time t_σ are Q independent. This behavior has been verified with excellent accuracy in a mesoscopic system [19].

On this background, we performed additional neutron-scattering experiments in order to study the β relaxation in more detail and especially to investigate the Q dependence of intermolecular correlations. The work presented here extends our earlier experiments to shorter times using the time-of-flight technique.

After some experimental considerations and a first look at the raw data in Sec. II, in Sec. III we work out a factorization property of $S(Q,\omega)$ in the β relaxation regime. We combine our intermediate scattering functions $\Phi(Q,t)$ with back scattering results for selected Q and T to achieve an enlarged dynamic range which allows us to determine the temperature dependence of the MCT parameters as well as to check the Q independence of the line-shape parameter λ and the time t_σ. These results will be used in Sec. IV for the analysis of the Q dependence of the α and β relaxation amplitudes f_Q and h_Q. We close with a discussion in Sec. V.

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II. EXPERIMENT AND RAW DATA

Fully deuterated OTP ($C_{18}D_{14}$, $T_g=243$ K, $T_m=329$ K) was obtained partly from the Max-Planck-Institut für medizinische Forschung (Heidelberg, Germany) and partly from MSD-Isotopes (Pointe Claire, Quebec, Canada). It was purified several times by slow vacuum distillation at 120 °C and 1 mbar. The purity and deuteration ratio as determined by mass spectroscopy and high-resolution NMR (400 MHz) were better than 99%. The coherent cross section thus contributes about 85% to the total neutron cross section.

The OTP was sealed into soda lime glass capillaries with an inner diameter of 1.2 mm and a wall thickness of 10 μm (Fa. Hilgenberg, Malzfeld) which were arranged on a circle of 2 cm diameter approximating a hollow cylinder. The container scattering was lowered significantly compared to the capillaries used previously [15].

Before filling, the capillaries were flushed with distilled water to remove dust, and burned out at 850 K for several hours, a process which turned out to considerably decrease crystallization tendencies. After filling, the sample was tempered at 350 K for several days. When left at room temperature no crystallization occurred over more than 12 months. The sample could be undercooled down to 200 K without any crystallization, but not lower because an increasing number of capillaries started to crack.

The experiments were performed on the time-of-flight spectrometer IN5 at the Institut Laue Langevin in Grenoble, France. Two different incident wavelength λ_i were used: (i) 5.7 Å, leading to an elastic energy resolution $\Delta E=65$ μeV [full width at half maximum (FWHM)] and a Q range $0.25\leq Q_{\text{el}}\leq 1.95$ Å^{-1}, and (ii) 6.5 Å with $\Delta E=25$ μeV and $0.2\leq Q_{\text{el}}\leq 1.7$ Å^{-1}. The sample transmission was about 90 %. A vanadium standard was used for relative detector normalization.

The raw data set of 89 groups of ^{3}He detectors was condensed into a reasonable number of points by regrouping several dectectors and binning time channels, keeping the relative errors below a bound of typically 10^{-2}. The data were then interpolated to constant Q with step $\Delta Q=0.05$ Å^{-1}.

Figure 1 shows some representative experimental spectra $S(Q,\omega)$ in units which are arbitrary but the same for all Q and T. Spectra for several temperatures at $Q=1.45$ Å^{-1} (a) and for different wave numbers at $T=293$ K (b) are shown on a logarithmic scale. One recognizes a strong quasielastic broadening for the higher temperatures which vanishes for the lower ones where it gives way to a distinct inelastic scattering around 1 meV (boson peak). We notice that the spectral shapes of $S(Q,\omega)$ are different for different Q.

In Fig. 2 we show the static structure factor $S(Q)$ as measured on IN5 [$S(Q)\simeq 2\int_0^{\omega_{\max}(Q)} d\omega S(Q,\omega)$]. The double-peak structure is resolved and the temperature dependence of $S(Q)$ (Fig. 2, inset) agrees well with the one measured on the diffractometer D20 [20]. One recognizes particularly strong temperature variations around 0.85 Å^{-1}, a region not accessible to the earlier diffraction experiment [20].

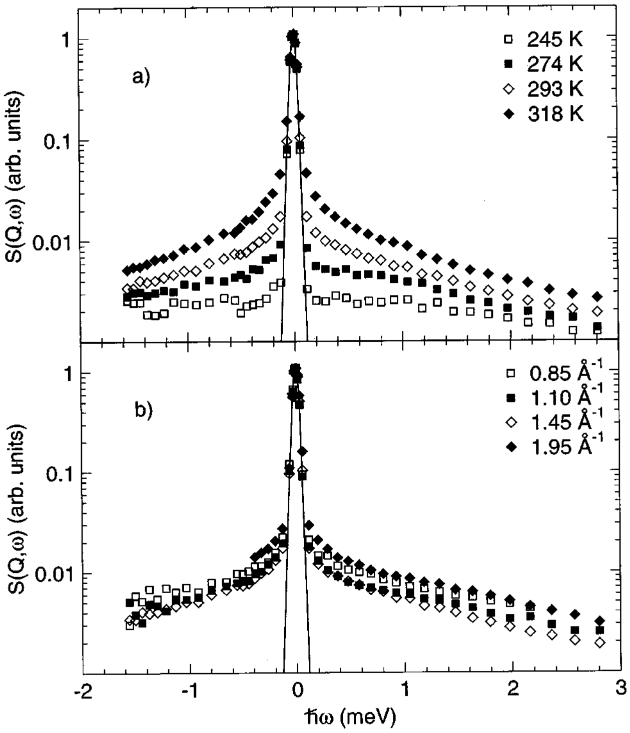

FIG. 1. Quasielastic spectra $S(Q,\omega)$ of deuterated OTP as measured on the time-of-flight spectrometer IN5, normalized to their values at $\omega=0$ (a) for selected temperatures T at $Q=1.45$ Å^{-1} and (b) for selected wave numbers Q at $T=293$ K. The line shows the experimental resolution of IN5 as measured on vanadium.

III. β SCALING REGIME

One of the most remarkable predictions of MCT is the factorization property of the β relaxation: In a certain frequency and temperature range, all observables are expected to have the same spectral distribution. This implies in particular that the neutron-scattering law $S(Q,\omega)$ factorizes into a Q- and a ω-dependent part:

$$S(Q,\omega)=S(Q)A_QG(\omega). \tag{2}$$

In Fig. 3 we show the rescaled intensities $S(Q,\omega)/S(Q)A_Q=G(\omega)$ for different wave numbers Q.

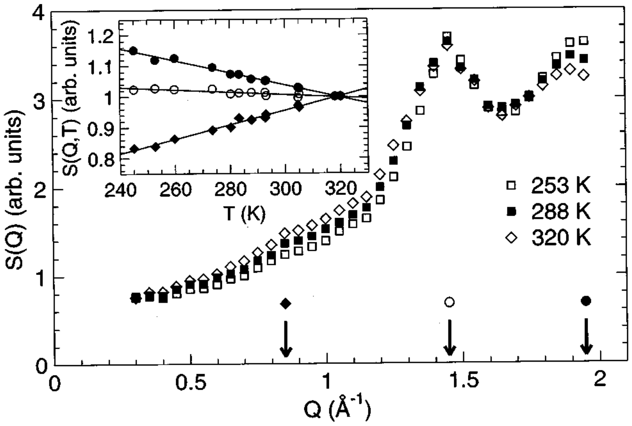

FIG. 2. Static structure factor $S(Q)$ of deuterated OTP for three different temperatures. The inset shows the temperature dependence of $S(Q)$ at selected values of Q which are indicated by the arrows in the figure. The $S(Q)$ have been scaled to their values at 320 K for clarity. Note the strong temperature variations of $S(Q)$ at $Q=0.85$ Å^{-1} and $Q=1.95$ Å^{-1}.

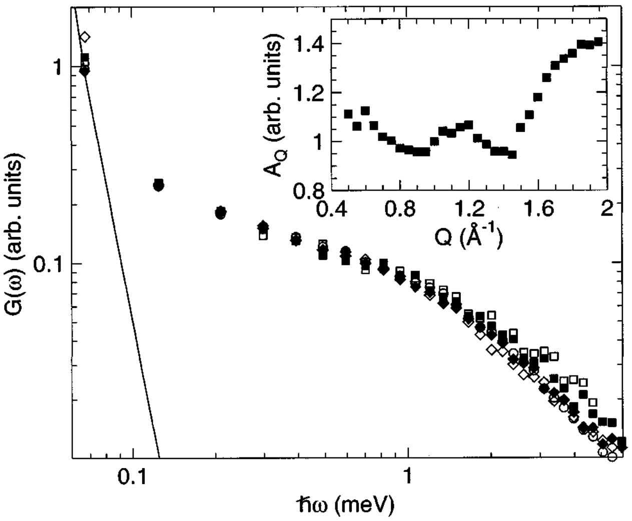

FIG. 3. Test of the factorization property (2) for the data at 293 K: The rescaled intensities $S(Q,\omega)/S(Q)A_Q = G(\omega)$ are shown for five wave numbers Q ranging from 0.85 Å^{-1} to 1.95 Å^{-1} on a logarithmic scale. For energies between 0.1 and 1 meV the spectral shape is the same. It starts to deviate more and more for energies higher than 1 meV. The line represents the measured resolution and sets the lower bound. The inset shows the scaling factor A_Q.

The factorization property (2) is found valid at least between 0.1 and 1 meV as the inelastic intensities can be scaled to a common frequency-dependent curve. The lower bound is due to elastic contamination. Above 1 meV the factorization breaks down, indicating that we are dealing with different processes. In fact 1 meV corresponds already to the boson peak (Fig. 1) and of course one must not [21] expect the β relaxation property (2) to hold for the vibrational spectrum above the boson peak maximum.

For a more quantitative analysis, we consider the normalized intermediate scattering function $\Phi(Q,t) = S(Q,t)/S(Q,t=0)$ obtained by Fourier deconvolution with the measured experimental resolution and by division with the static structure factor $S(Q)$.

We start our analysis by combining the new time-of-flight (IN5) with old backscattering (IN13) data for selected Q and T values. For $\lambda_i = 5.7$ Å the dynamic ranges of the two instruments overlap between 10 and 20 ps. As the back scattering data of IN13 of Ref. [15] could not be obtained on an absolute scale they have to be matched in the overlap region. Full $\Phi(Q,t)$ extending over more than two and a half decades in time are obtained for $Q = 0.85$, 1.2, 1.4, 1.6, 1.8, and 1.95 Å^{-1} at 293, 305, and 320 K.

For fits with Eq. (1) we used the tabulated [22] expansion coefficients of the scaling function $g_\lambda(t/t_\sigma)$ which is completely determined by one unique exponent parameter λ. In the incoherent case we got $\lambda = 0.77$ [12] from the long-time asymptote of $\Phi(Q,t)$. As meaningful four-parameter fits are not warranted by the precision of the data we profit from the fact that the line shape is not very sensitive to a variation of λ and fix $\lambda = 0.77$ for the rest of the analysis. As shown in Fig. 4 the two-step decay can be fitted according to the scaling law (1) in the range from 1 to 100 ps.

The temperature behavior of the β relaxation amplitude H_Q and the crossover time t_σ are predicted as

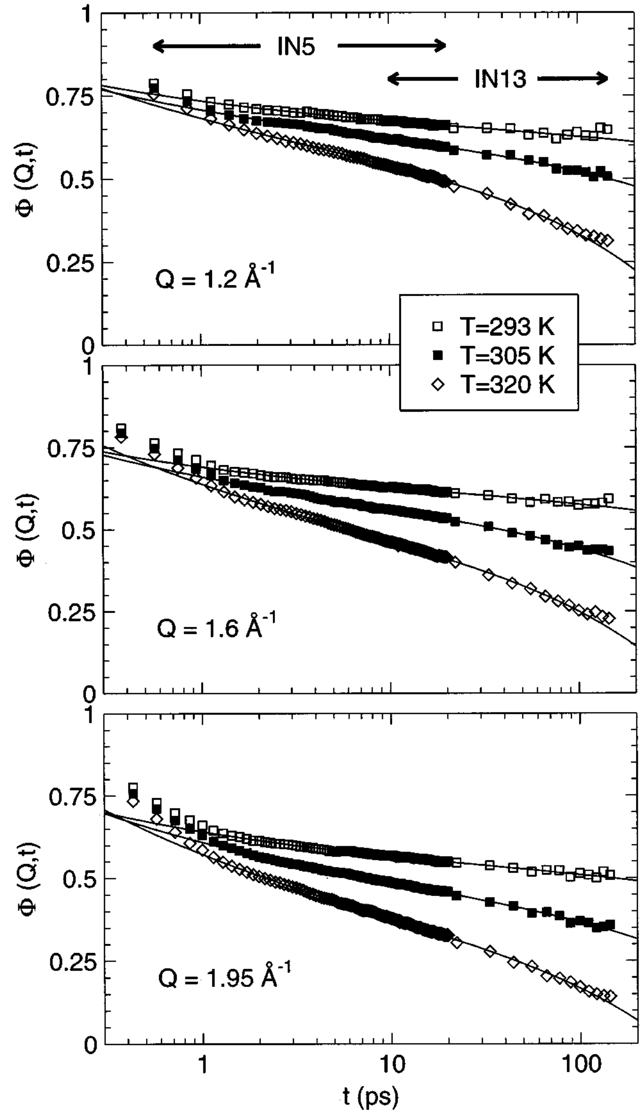

FIG. 4. Normalized intermediate scattering function $\Phi(Q,t)$ for different wave numbers and temperatures. IN5 and IN13 data are matched in their overlap region between 10 to 20 ps. Lines are fits with the MCT scaling law (1) with a fixed shape parameter $\lambda = 0.77$. Notice the lower plateau value of the correlation functions at $Q = 1.95$ Å^{-1} compared to $Q = 1.6$ Å^{-1}.

$$H_Q = h_Q c_0 |\sigma|^{1/2}, \quad t_\sigma = t_0 |\sigma|^{-1/2a}. \tag{3}$$

c_0 is a dimensionless constant and the critical exponent of the β relaxation, $a = 0.295$, is determined by λ. In Fig. 5 the amplitude and the crossover time are shown as H_Q^2 and $t_\sigma^{-1/2a}$ in order to test the predicted behavior. Extrapolation of the T dependence gives consistently $T_c \simeq 290$ K. The prefactor H_Q is found to depend on Q as already indicated by the earlier experiment [15]. t_σ is independent of Q within the statistics; i.e., all correlation functions are equally well fitted with a fixed Q-independent t_σ.

IV. WAVE-NUMBER DEPENDENCE OF AMPLITUDES

The combination of IN5 and IN13 data allowed us to obtain the intermediate-scattering function $\Phi(Q,t)$ over two and a half decades in time — at the expense of resolution in

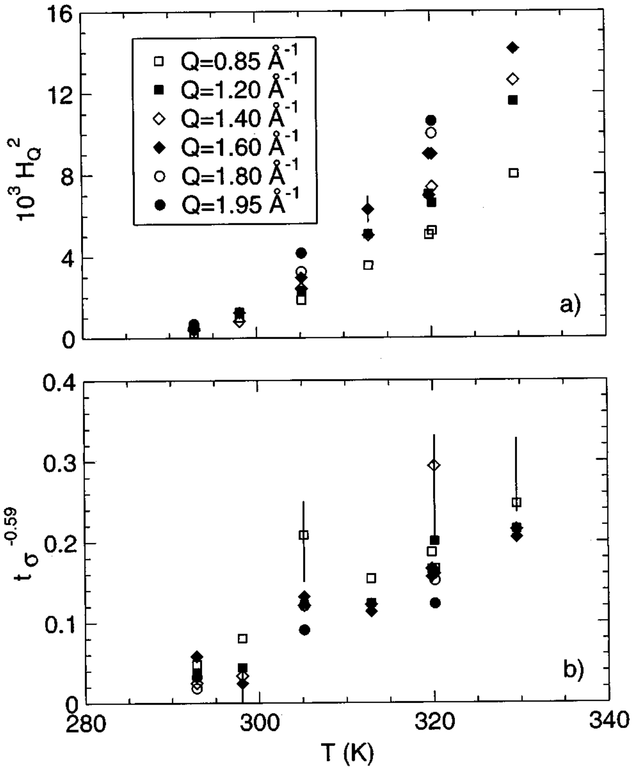

FIG. 5. Parameters H_Q (a) and t_σ (b) above 290 K as obtained from the fits with (1) for selected values of Q. The parameters are plotted such that a linearized T dependence is expected according to (3). Both extrapolate consistently to $T_c \simeq 290$ K.

Q. For analyzing the wave-number dependence of f_Q and H_Q it is preferable to use IN5 data alone. Below 288 K, $\Phi(Q,t)$ decays only due to the microscopic transient below 1 ps and stays constant in the range 2–20 ps within experimental errors [23]. The Debye-Waller factor f_Q is obtained directly from the time average over this region. Above 293 K, f_Q is extracted from fits with a fixed t_σ (Sec. III). The f_Q and H_Q are in very good agreement with the results obtained from the combined IN5/IN13 analysis, thereby supporting the correctness of our approach.

The temperature evolution of the Debye-Waller factor $f_Q(T)$ was measured with spin-echo IN11 and backscattering IN13 and has already been reported in [15] for $Q \geqslant 1.2$ Å^{-1}. MCT predicts

$$f_Q(T)=f_Q^c+h_Q\sqrt{\sigma}+O(\sigma), \qquad T\leqslant T_c, \tag{4a}$$

for the glass and

$$f_Q(T)=f_Q^c+O(\sigma), \qquad T>T_c, \tag{4b}$$

for the liquid.

In Fig. 6 the so-obtained Debye-Waller factors are plotted as a function of temperature for some selected wave numbers. Even without fitting with Eq. (4a), one recognizes a cusp at about $T_c \simeq 290$ K, more or less pronounced for different Q. Above T_c, f_Q shows a smooth variation which is compatible with a linear dependence on σ.

In order to extract the critical Debye-Waller factor $f_Q^c = f_Q(T_c)$ and the β relaxation amplitude h_Q we apply Eq. (4a). The result is shown in Fig. 7 and can now be compared to the static structure factor $S(Q)$ of OTP.

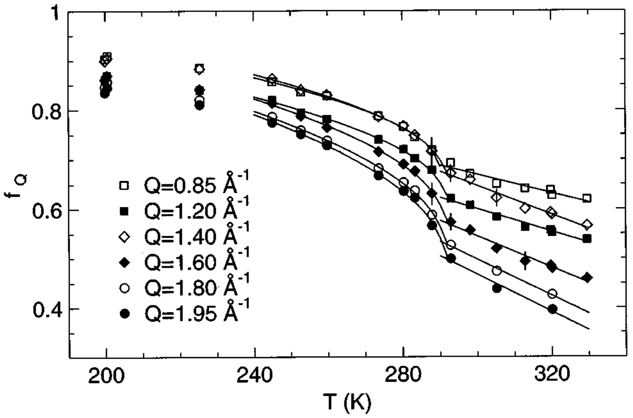

FIG. 6. Temperature dependence of the Debye-Waller factor for selected values of Q. For $T > 290$ K the results from the fits with Eq. (1) are shown. Values below 290 K were obtained as described in the text. Lines are fits with Eq. (4a), leading to a critical temperature $T_c \simeq 290$ K. Above T_c a linear temperature dependence is assumed.

The Q dependence of the nonergodicity parameter f_Q^c shows features similar to that of the static structure factor $S(Q)$ at least for $Q \leqslant 1.6$ Å^{-1}. Even the tiny shoulder in the static structure factor $S(Q)$ around $Q \simeq 0.8$ Å^{-1} (Fig. 2) is reflected in f_Q^c. The low-Q feature in $f_Q(T)$ is found at all temperatures, and so it is clearly significant. However, the pronounced second maximum of $S(Q)$ at about 1.9 Å^{-1} seems to be missing in f_Q^c, except for an eventually small shoulder (unfortunately, in this high-resolution measurement we had to choose an elastic Q range that does not extend beyond the second structure factor maximum).

The amplitude h_Q oscillates in antiphase with $S(Q)$. Two minima are discernable, the first in the region of the prepeak at $Q = 0.8$ Å^{-1} and the second in the vicinity of the first structure factor maximum at $Q \simeq 1.35$ Å^{-1}. However, both minima in h_Q are slightly shifted to smaller Q values compared to $S(Q)$. There are no indications for a minimum at the second structure factor peak at $Q \simeq 1.9$ Å^{-1}.

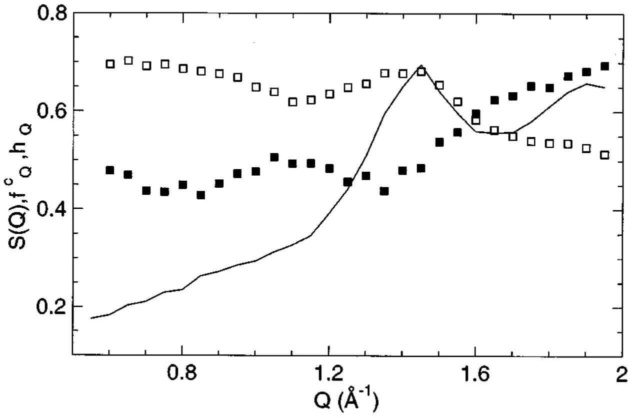

FIG. 7. Q dependence of the Debye-Waller factor $f_Q^c = f_Q(T_c)$ (open squares) and the amplitude h_Q (solid squares) from analysis of the data in Fig. 6 with Eq. (4a) compared to the static structure factor of OTP (line). Note the minima in h_Q are at lower Q values with respect to $S(Q)$.

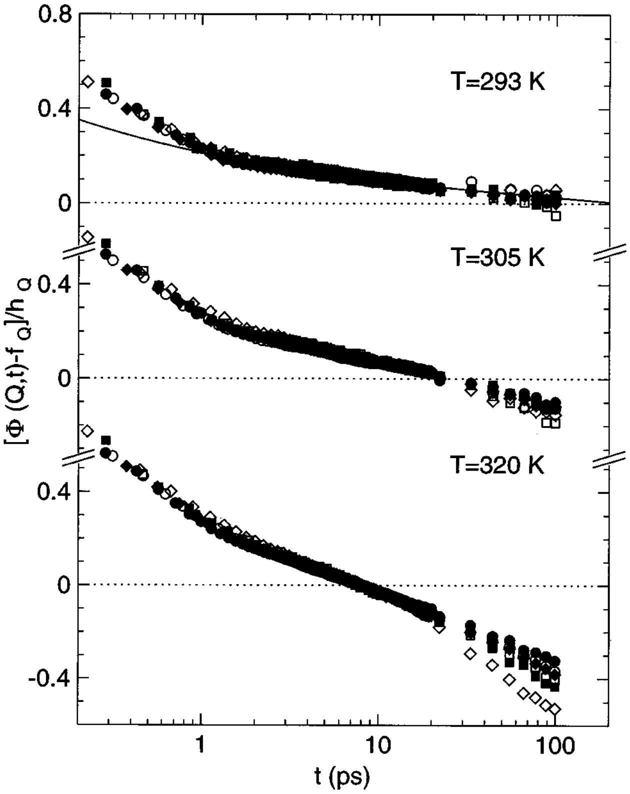

FIG. 8. Correlation functions for three different temperatures scaled according to Eq. (1). $[\Phi(Q,t)-f_Q]/h_Q$ is plotted as a function of the logarithm of time. The symbols are the same as in Fig. 5. The solid line is the β correlator $g_\lambda(t/t_\sigma)$ with $\lambda=0.78$.

According to mode-coupling theory the dynamics between the microscopic motions and that of the α process is governed by the β process. In our experiment this interval lies approximately between 1 ps and t_σ, the inflection point in $\Phi(Q,t)$ versus $\ln t$. Figure 8 demonstrates that the factorization property of the β process in this time interval is well satisfied; i.e., $[\Phi(Q,t)-f_Q]/h_Q$ is independent of Q. As expected, t_σ and the time range over which the factorization property is fulfilled decrease with increasing temperature. The small but systematic deviations seen at times beyond t_σ can be attributed to the α process. The apparently identical behavior for times lower than 1 ps should not be overinterpreted, since in the Fourier transform all higher frequencies are condensed into a few points and no details are visible. This factorization is only possible if the line-shape parameter λ and the crossover time t_σ of the β correlator are both independent of Q. Thus Fig. 8 strongly supports the results of Fig. 3 and the independence of both parameters. At 293 K a fit of $g_\lambda(t/t_\sigma)$ to the joined data gives $\lambda=0.78$, in excellent accordance with the value 0.77 determined previously [12].

V. DISCUSSION

In the undercooled liquid we could follow the density correlator over two and a half decades by combining the intermediate scattering functions from two spectrometers. $\Phi(Q,t)$ can be well described around the plateau by the scaling law of MCT. The amplitude H_Q and the crossover time t_σ follow power laws. Both parameters extrapolate consistently to $T_c \simeq 290$ K. The same critical temperature is obtained from the cusp of the Debye-Waller factor. We would like to stress that all parameters could be determined independently from our coherent data; their temperature dependence is in good accordance with results from incoherent neutron scattering [14].

The main motivation for coherent scattering experiments lies in the additional information about spatial correlations. In the present case, this information is contained in the wave-number dependence of the α and β relaxation amplitudes f_Q and h_Q; for the determination of the α relaxation time scale τ_Q, higher resolution or higher temperatures would be necessary. Results from previous measurements in three model liquids have been reviewed in Ref. [24]: generally, f_Q, h_Q, and τ_Q are found to reflect the oscillatory behavior of the static structure factor $S(Q)$. The α relaxation is particularly slow around the structure factor, just as expected from the well-known de Gennes narrowing in the normal liquid phase. Its amplitude f_Q oscillates in phase with $S(Q)$ while the β relaxation amplitude h_Q shows the opposite behavior.

The same phenomenon has also been observed in light-scattering experiments on a colloidal suspension of hard spheres [19]; the data were in quantitative agreement with numeric solutions of mode-coupling equations [16]. According to mode-coupling theory, relaxational dynamics is due to a backfeed effect expressed by a memory kernel which is governed by vertices which in turn depend only on the static structure factor $S(Q)$ [8]. The characteristic wave-number dependence of the relaxation amplitudes comes out as a direct consequence of this coupling.

So far mode-coupling equations have been worked out only for translational degrees of freedom. In a strict sense, the theory therefore applies only for systems of rigid spherical particles or mixtures of such [18]. In more complex molecular glass formers, there are additional rotational and intramolecular vibrational degrees of freedom which couple more or less strongly to the translational modes. The factorization property of the β relaxation makes it possible that in spite of these complications a generic frequency dependence is observed.

The static structure factor $S(Q)$ of a molecular system does not only describes molecule-molecule correlations but contains also intramolecular contributions. There is no obvious way to relate the structural quantity to the Q-dependent dynamical quantities and, it is left open to further experiments how far characteristic features of $S(Q)$ reflect themselves in the relaxation amplitudes f_Q and h_Q. In orthoterphenyl, our older dynamic measurements could not fully separate the two maxima of $S(Q)$ at 1.45 and 1.9 Å^{-1} and the region of the prepeak at 0.8 Å^{-1} was not accessible [15]. In the present experiment the Q resolution was sufficient to resolve variations of both the Debye-Waller factor f_Q^c and the amplitude h_Q. The first peak of $S(Q)$ at 1.45 Å^{-1} and even the prepeak at 0.8 Å^{-1} are clearly seen in f_Q and h_Q; there is only a weak, if at all, indication for a shoulder at 1.9 Å^{-1}. The reflection of the prepeak of $S(Q)$ in both parameters comes up as a novel feature and underlines the highly cooperative nature of the glass transition. We note in particular that the temperature dependence of $S(Q)$ is most pronounced around its second maximum (Fig. 2, inset, and Ref. [20]), and that α relaxation times at 1.9 Å^{-1} were found to scale with viscosity, determined without deconvoluting any intramolecular modes [10]: It remains surprising that these features might have no counterpart in the relaxation amplitudes.

Results for the relaxation strengths have been found in the polymer polybutadiene (PB) [25,26] and the ionic mixture CKN [$\mathrm{Ca}_{0.4}\mathrm{K}_{0.6}(\mathrm{NO}_3)_{1.4}$] [27,28].

In polybutadiene, the first structure factor maximum around 1.4 Å^{-1} is mirrored in f_Q and h_Q [25,26]. However, for f_Q no values for $Q<1.1$ Å^{-1} were reported and the β relaxation amplitude has only been calculated as the inelastic scattering fraction. The dynamics around the second maximum at 2.7 Å^{-1}, which can be ascribed to innerchain correlations, seems to be dominated by localized jump processes [29]. In orthoterphenyl, there is no such simple way to separate inter- and intramolecular correlations.

In the ionic mixture $\mathrm{Ca}_{0.4}\mathrm{K}_{0.6}(\mathrm{NO}_3)_{1.4}$ there is not only a structure factor maximum at 1.8 Å^{-1} but also a prepeak around 0.8 Å^{-1} which corresponds, however, to the electrostatically stabilized cation-cation distance. These two peaks have only weak counterparts in the amplitudes f_Q and h_Q [27,28], which have only been estimated: f_Q^c was extrapolated from temperatures above T_c; h_Q was calculated as the difference $f_Q(T)-f_Q^c$, where $f_Q(T)$ was measured well below T_g.

While the mode coupling theory is highly successful in explaining the frequency dependence of relaxational spectra, its present state does not allow us to fully exploit the additional information offered by the momentum resolution of coherent scattering.

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