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# Three homeotropically aligned nematic liquid crystals: Comparison of ultrafast to slow time-scale dynamics

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The dynamics of two nematic liquid crystals, 4-(trans-4′-*n*-octylcyclohexyl)isothiocyanatobenzene and 4-(4-pentyl-cyclohexyl)-benzonitrile, are investigated as a function of temperature both in the homeotropically aligned nematic phase and in the isotropic phase using optical heterodyne-detected optical Kerr effect experiments, which measures the time derivative of the polarizability–polarizability-correlation function (orientational relaxation). Data are presented over a time range of 500 fs−70 μs for the nematic phase and 500 fs to a few hundred nanoseconds for the isotropic phase. The nematic dynamics are compared with a previously studied liquid crystal in the nematic phase. All three liquid crystals have very similar dynamics in the nematic phase that are very different from the isotropic phase. On the slowest time scale (20 ns–70 μs), a temperature-independent power law, the final power law, $t^{-f}$ with $f \sim 0.5$, is observed. On short time scales (∼3 ps to ∼1 ns), a temperature-dependent intermediate power law is observed with an exponent that displays a linear dependence on the nematic order parameter. Between the intermediate power law and the final power law, there is a crossover region that has an inflection point. For times that are short compared to the intermediate power law (≲2 ps), the data decay much faster, and can be described as a third power law, although this functional form is not definitive. The isotopic phase data have the same features as found in previous studies of nematogens in the isotropic phase, i.e., the temperature-independent intermediate power law and von Schweidler power law at short to intermediate times, and a highly temperature-dependent long time exponential decay that is well described by the Landau-de Gennes theory. The results show that liquid-crystal dynamics in the nematic phase exhibit universal behavior. © *2006 American Institute of Physics*. [DOI: 10.1063/1.2149867]

## I. INTRODUCTION

Dynamics of liquid crystals are complex and are the subjects of extensive experimental and theoretical studies because of the intrinsic interest in molecular systems that self-organize and because of the practical importance of liquid crystals. Remarkably, intricate dynamics have been observed around phase transitions even in the case of the simplest mesophase, the nematic phase. Like normal liquids, nematogens in the nematic phase[1] have no long-range correlation between their centers of gravity. Liquid crystals in the nematic phase flow like a normal liquid and have viscosities on the same order of magnitude as the corresponding isotropic phase. However, the nematogens in the nematic phase are, on average, aligned with their long axes having a net projection along a common direction, the macroscopic director.[1] This molecular ordering strongly affects the orientational relaxation behavior in the nematic phase and in the isotropic phase as the temperature is lowered toward the nematic-isotropic (*N-I*) phase transition from above.

Previous optical heterodyne-detected optical Kerr effect (OHD-OKE) experiments on nematogens in the isotropic phase observed two power law decays at short times (picoseconds to subnanoseconds) and an exponential decay at long times (nanoseconds to submicroseconds).[2–5] The long time-scale exponential decay is highly temperature dependent, and is well understood in terms of Landau-de Gennes (LdG) theory.[1] LdG theory predicts that in the isotropic phase there is short-ranged nematiclike order on a distance scale given by a correlation length, $\xi$. $\xi$ is highly dependent on temperature $T$, with

$$\xi(T) = \xi_0[T^*/(T-T^*)]^{1/2}, \quad T > T^*, \tag{1}$$

where $\xi_0$ has a value on the order of a molecular dimension and $T^*$ is the mean-field second-order phase transition temperature. $T^*$ is slightly below the phase transition, $T_{N\text{-}I}$, due to the weakly first-order character of the phase transition. The long time exponential decay is caused by the randomization of the pseudonematic domains. The relaxation time $\tau_{\mathrm{LdG}}$ of the exponential decay increases dramatically and diverges as the temperature approaches $T^*$ as a result of the growing correlation length $\xi$. $\tau_{\mathrm{LdG}}$ is given by

$$\tau_{\mathrm{LdG}} = \frac{V_{\mathrm{eff}}^* \eta(T)}{k_B(T-T^*)^{\gamma}}, \tag{2}$$

where $\eta(T)$ is the viscosity, $V_{\mathrm{eff}}^*$ is the effective volume, $k_B$ is the Boltzmann constant, and $\gamma$ has been shown to be 1, indicating the validity of mean-field theory.[1] In the pseudonematic domain picture, the short time-scale dynamics are dominated by intradomain relaxation. The observed short time-scale power laws have temperature-independent exponents and are not explained by the LdG theory. A number of theoretical studies[5–8] have addressed the short-time power laws, and a recent schematic mode coupling theory model of the orientational relaxation of nematogens in the isotropic phase has been able to reproduce the time- and temperature-dependent data for six nematogens.[9]

Although dynamics in the isotropic phase have been studied extensively,[3–5,10–18] experimental studies on the nematic phase have focused on the hydrodynamic regime (time scales much slower than microseconds).[19–23] The long time-scale dynamics have been explicated in a theory by Leslie-Ericksen.[24,25] Leslie-Ericksen hydrodynamic theory describes the dynamics of the changes in the director orientation while evolving from a nonequilibrium state to an equilibrium state. Time-resolved deuterium nuclear magnetic resonance (NMR) spectroscopy[21,22] has been applied to study the director reorientation dynamics of nematic liquid crystals. In these experiments, the director was first aligned along the direction of the magnetic field of the NMR spectrometer. Then, an electric field at a nonzero angle to the magnetic field was applied, during which the director moved from being parallel to the magnetic field to being at an angle to it. After the electric field was turned off, the director relaxed back to its initial direction, i.e., being parallel to the magnetic field. The change of the director orientation following the application and removal of the electrical field was recorded in the NMR spectra and has been shown to agree with the predictions of Leslie-Ericksen hydrodynamic theory. The tangent of the angle between the director and the magnetic field is predicted within the framework of Leslie-Ericksen hydrodynamic theory by solving the torque-balance equation,[26] to vary exponentially with the relaxation time determined by the strength of the magnetic and electric fields and the rotational viscosity coefficient.

A detailed examination of orientational dynamics in the nematic phase on short to moderate time scales was provided by recent OHD-OKE experiments applied on a homeotropically aligned nematic liquid crystal,4′-pentyl-4- biphenylcarbonitrile (5-CB).[27] The orientational dynamics in the nematic phase were shown to be very different from those observed in the isotropic phase.

To obtain a better understanding of the liquid-crystal dynamics and to investigate the universality of the nematic phase dynamics observed previously, OHD-OKE experiments have been applied to two additional homeotropically aligned nematic liquid crystals, 4-(trans-4′-*n*- octylcyclohexyl)isothiocyanatobenzene (8CHBT) and 4-(4-pentyl-cyclohexyl)-benzonitrile (5PCH). In this paper, the data for 8CHBT and 5PCH are presented and the results are compared to those of 5CB.[27] It will be shown that the data for all three liquid crystals can be fit extremely well on all time scales (500 fs to $>2\ \mu$s) over a wide range of temperatures by the same empirical function. All the features observed for 5CB, the temperature-independent final power law, the intermediate power law with an exponent that varies linearly with the order parameter, and the crossover region showing an inflection point that connects the two power laws, are also observed for 8CHBT and 5PCH. The results indicate that the nematic phase of liquid crystals has a universal behavior that is insensitive to the details of the molecular structures of the nematogens. In particular, the final power law observed from 2 ns to $>2\ \mu$s, within experimental errors, has an exponent $0.53\pm0.02$ for all three liquid crystals.

## II. EXPERIMENTAL PROCEDURES

OHD-OKE spectroscopy[17,28,29] was used to measure the orientational dynamics in the nematic phase of the liquid crystals, 8CHBT and 5PCH. The experiment measures the time derivative of the polarizability–polarizability-correlation function, which is equivalent to the orientational correlation function except on very short time scales ($\lesssim 1$ ps) where there can be contribution from collision-induced multiparticle effects.

To observe the full range of the dynamics, at each temperature several sets of experiments were performed with different pulse lengths and delays. For times $t<30$ ns, a mode-locked 5 kHz Ti:sapphire laser/regenerative amplifier system was used ($\lambda=800$ nm) for both the pump and the probe. For longer times, a continuous wave diode laser was used as the probe, and a fast digitizer (1 ns per point) recorded the data. The scans taken over various time ranges overlapped substantially, permitting the data sets to be merged by adjusting only their relative amplitudes. Additional experimental details have been published previously.[4,27,30]

8CHBT and 5PCH were purchased from Aldrich and used without further purification. 8CHBT and 5PCH have isotropic nematic transitions at 320–321 and 328–329 K, respectively. The samples were homeotropically aligned between two indium tin oxide-coated glass windows treated with surfactant dimethyloctadecyl[3-(tri-methyoxysilyl) propyl]ammonium chloride. The details of the sample preparation were presented previously.[27] An important aspect of these experiments is that the pump pulse propagates exactly along the director, which is normal to the faces of the windows. The electric field of the pump pulse is perpendicular to the direction of propagation, and it will disturb the angular distribution of the nematogens about the director without changing the orientation of the director. Following the pump pulses, the perturbed nematogen distribution will relax back to nematic alignment with the order parameter that corresponds to the sample temperature. Linear response theory[31] demonstrates that the relaxation from a sudden small perturbation of a system is directly related to the thermal equilibrium fluctuation of a system. Therefore, the dynamics observed in these experiments correspond to the thermal equilibrium orientational fluctuations of nematogens about the director. The experiments presented here are distinct from dynamical experiments performed on the nematic phase of liquid crystals on very long time scales.[19–23] In these slow experiments a perturbation is applied that changes the orientation of the director and then the time dependence of the establishment of the nematic order about the new direction of the director is observed.

## III. RESULTS AND DISCUSSION

OKE decay curves were measured for 8CHBT and 5PCH as a function of temperature from ~20 K above the

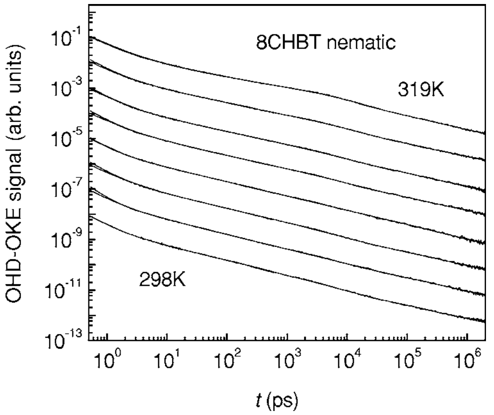

FIG. 1. Temperature-dependent nematic phase data for 8CHBT displayed on a log plot with fits using Eq. (3). The data sets have been offset along the vertical axis for clarity. Starting with the topmost curve, the temperatures for the data sets are 319, 316, 313, 310, 307, 304, 301, and 298 K, respectively. The final power law is temperature independent (see Fig. 4), but the intermediate power law is temperature dependent (see Fig. 3).

*N-I* transition to ~20 K below it. For the nematic phase, the data were taken from subpicoseconds to 2 $\mu$s and in some instances to 70 $\mu$s; for the isotropic phase, the data were taken from subpicoseconds to the delay times sufficiently long to accurately measure the final portion of the decay, the LdG exponential decay. The nematic phase data are displayed in Figs. 1 and 2. The temperatures are given in the figure captions. In the previous OKE studies on 5CB,[27] the nematic phase data were fitted to an empirical function that was shown to work very well over the entire time window. The fitting function, $F_N(t)$ ($N$ for nematic) was

$$F_N(t) = (at^{-s} + pt^{-z})\exp(-t/\tau) + gt^{-f}[1 - \exp(-t/\tau)]. \quad (3)$$

The fitting function has been discussed in detail previously.[27] Although there are a large number of parameters in Eq. (3), they can be determined quite accurately because most of them are relatively independent of the others. The fits for 8CHBT and 5PCH using Eq. (3) are also shown as the solid lines through the data in Figs. 1 and 2. The results of the fits using Eq. (3) are presented in Table I. Because data were taken at ~10 temperatures for each nematogen, the results for only three temperatures are presented in Table I: the highest, intermediate, and lowest temperatures. All fits span the time range 0.5 ps-2 $\mu$s. Equation (3) provides near perfect fits for both samples at all temperatures, which can be seen from the fact that the calculated curves are barely discernible from the data. The shapes of the decays for 8CHBT and 5PCH in the nematic phase are virtually identical to those of 5CB. For all three liquid crystals studied in the nematic phase, the very short time behavior (<3 ps) is modeled as a power law, $t^{-s}$. In contrast to the other power laws, this portion of the decay spans a relatively short time range. Therefore, its functional form cannot be definitively assigned as a power law.

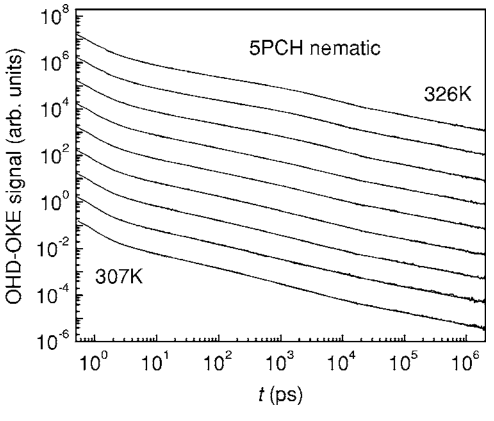

FIG. 2. Temperature-dependent nematic phase data for 5PCH displayed on a log plot with fits using Eq. (3). The data sets have been offset along the vertical axis for clarity. Starting with the topmost curve, the temperatures for the data sets are 326, 325, 323.8, 322, 319, 316, 313, 310, and 307 K, respectively. The final power law is temperature independent (see Fig. 4), but the intermediate power law is temperature dependent (see Fig. 3).

TABLE I. Parameters from fits to Eq. (3) for three nematogens in the nematic phase. Three temperatures, the highest, an intermediate, and the lowest, are presented for each nematogen. The results for 5CB are taken from previous work (Ref. 27).

| Nematogen | $T$ (K) | $s$ | $z$ | $f$ | $\tau$ (ns) |
|---|---|---|---|---|---|
| 8CHBT | 319 | 1.17 | 0.39 | 0.53 | 12.8 |
| | 310 | 1.49 | 0.52 | 0.53 | 7.26 |
| | 298 | 1.69 | 0.57 | 0.53 | 9.05 |
| 5PCH | 326 | 1.74 | 0.45 | 0.52 | 5.37 |
| | 319 | 2.14 | 0.55 | 0.52 | 4.39 |
| | 307 | 2.50 | 0.63 | 0.52 | 6.64 |
| 5CB | 306 | 1.68 | 0.38 | 0.54 | 203 |
| | 303 | 1.97 | 0.44 | 0.54 | 102 |
| | 298 | 2.10 | 0.47 | 0.54 | 80.0 |

The very short time decay is followed by another power law, the intermediate power law, represented by the term $t^{-z}$ in Eq. (3). The intermediate power law spans a few picoseconds to hundreds of picoseconds. The intermediate power law has been observed by previous OHD-OKE studies on supercooled liquids and nematogens in the isotropic phase, and it was found for these types of liquids that the power-law exponent $z$ is independent of temperature.[18] For liquid crystals in the isotropic phase, $z$ depends approximately linearly on the aspect ratio of the nematogens.[17] In the nematic phase, however, the power-law exponent $z$ depends on the temperature.[27] Figures 3(a) and 3(b) display the temperature dependence of $z$ for the two nematogens studied here, and the same results were found previously for 5CB.[27] As the temperature decreases, $z$ increases. $z$ varies from 0.39 to 0.57 for 8CHBT and from 0.45 to 0.63 for 5PCH, respectively, as the temperature is decreased ~20 K below the *N-I* transition. The OHD-OKE experiment measures the negative of the time derivative of the polarizability–polarizability (orientational)-correlation function. Associated with the intermediate power law $t^{-z}$, the correlation function decays with a functional form of $-t^{1-z}$. The increase of $z$ with decreasing temperature shows that the decay of the correlation function becomes slower as the temperature decreases. In the nematic phase, the microscopic order parameter $S$ is a measure of orientational order, i.e., the tendency of the nematogens to align along the director. $S$ monotonically decreases as the temperature increases in the nematic phase and becomes zero in the isotropic phase.[1,32] Although the functional form

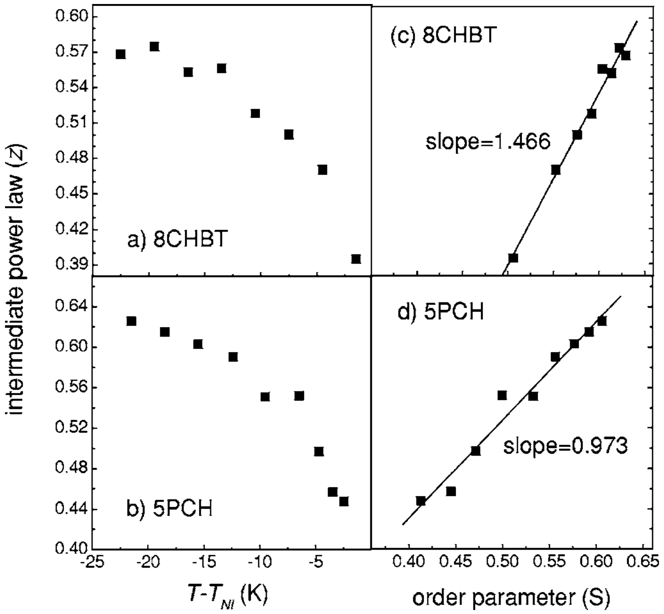

FIG. 3. (a), (b) The temperature dependence of the intermediate power law ($t^{-z}$) exponent $z$ obtained from the fits of the nematic phase data using Eq. (3). (c), (d) The exponent vs the nematic phase temperature-dependent order parameter, $S$. The order parameter data are taken from the literature (Refs. 3 and 34). The exponent and the order parameter appear to be linearly correlated.

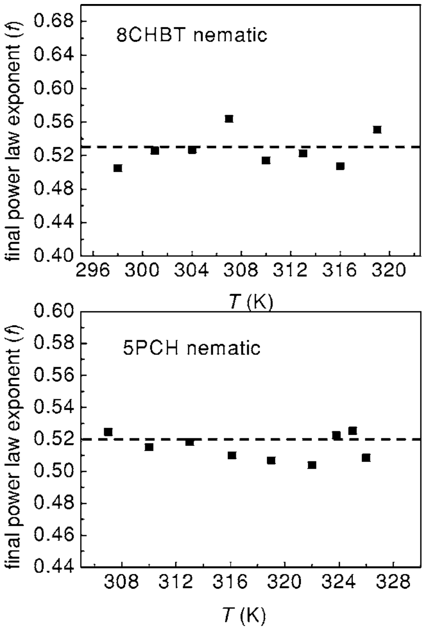

FIG. 4. The final power law ($t^{-f}$) exponent $f$ vs temperature obtained from the fits in Figs. 1 and 2. Within experimental error, the exponent is temperature independent. The dashed lines show the averaged values of $f$.

of the intermediate power-law exponent $z$ versus temperature is hard to determine, it is found, as displayed in Figs. 3(c) and 3(d), that, within experimental error, $z$ depends linearly on the change of the order parameter with temperature. Recently a schematic mode coupling model of nematogen dynamics in the isotropic phase has been presented.[9] This model was able to reproduce the full time and temperature dependence of the OHD-OKE data. However, there has been no detailed theoretical work done on the nematic phase dynamics other than the very long time-scale exponential hydrodynamic regime.[24,25] The linear correlation between the intermediate power-law exponent and the nematic order parameter provides an important touchstone for future theoretical treatments of the liquid-crystal dynamics.

Following the intermediate power law, the decay curves start to bend and cross over into a third power law, the final power law denoted by the term $t^{-f}$ in Eq. (3). The crossover region has been discussed in detail for 5CB in Ref. 27. The identical behavior is observed for the two nematogens studied here. In Figs. 1 and 2 the final power law spans from ~20 ns to 2 $\mu$s, appearing as straight lines on the log scale. Data were recorded to 70 $\mu$s, and there is no evidence of a change in the functional form. The most pronounced difference between the nematic phase data and the isotropic phase data (see Refs. 17 and 27 and Figs. 5 and 6) is the long time (ns to $\mu$s) behavior. In the isotropic phase, the decay at long time is an exponential [see Eq. (4) and Refs. 5 and 17]. The long time exponential decay is well described by LdG theory and is attributed to the pseudonematic domain randomization.[1] In the nematic phase over this time range, the decay is a power law, the final power law. Figure 4 shows the final power-law exponent $f$ as a function of temperature for both nematogens. Within experimental error, $f$ does not depend on the temperature. The averaged value of $f$ is 0.53 for 8CHBT and 0.52 for 5PCH, both of which are very close to the value obtained previously for 5CB, 0.54.[27] Within the experimental error, $f=0.53\pm0.02$. The results on three nematic liquid crystals indicate that there is a universal power-law decay, $t^{-f}$ with $f\sim0.5$. The exponent is not sensitive to the details of the molecular structures, at least for rodlike nematogens. At this time, there is no physical model to explain the existence and universality of this power law.

For all three liquid crystals, the transition region between the intermediate power law and the final power law has a mild "S" shape, that is, there is an inflection point. The slope in the later portion of the transition region is actually steeper than that of the final power law. The data progress from the intermediate power law to a decay that is steeper than the final power law, before becoming again less steep as the final power law. The mild S shape is more pronounced at higher temperatures because the intermediate power law has a smaller exponent so there is a larger difference in the slope between the starting and ending portions of the S shape. For those temperatures at which the intermediate power law has an exponent close to that of the final power law, the S shape is hardly perceivable. In Eq. (3), the crossover from the intermediate power law to the final power law is modeled as a decaying exponential that damps the intermediate power law and a growth in the exponential that turns on the final power law. To minimize the numbers of fitting parameters, the two exponential terms are given the same time constant, $\tau$. It is not clear if the decaying and growing exponentials that describe the crossover region have a physical significance or are simply a mathematical model of the behavior. Regardless, this form reproduces the mild S shape extremely well.

For comparison and completeness, data were also taken on the isotropic phase of 8CHBT and 5PCH, although the isotropic phase dynamics have been studied extensively on

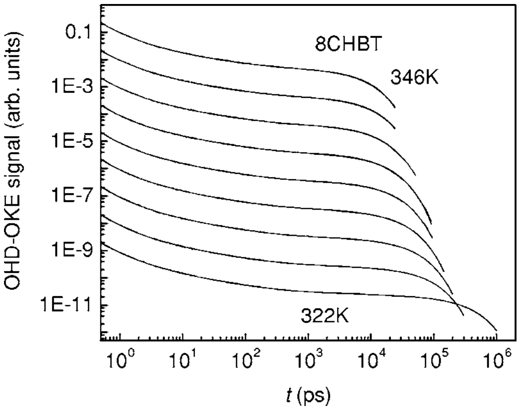

FIG. 5. Temperature-dependent isotropic phase data for 8CHBT displayed on a log plot with fits using Eq. (4). The data sets have been offset along the vertical axis for clarity. Starting with the topmost curve, the temperatures for the data sets are 346, 343, 340, 337, 334, 331, 328, 325, and 322 K, respectively.

several other liquid crystals.[5,17] Figures 5 and 6 show the isotopic phase data for 8CHBT and 5PCH, respectively. The temperatures for each data set are given in the figure captions. The isotropic phase data have been analyzed and reproduced with the schematic mode coupling theory model,[9] which requires numerical solutions to the mode coupling theory equations. For comparison to the nematic phase results, the isotropic phase data are described by the phenomenological function

$$F_I(t) = (at^{-s} + pt^{-z} + dt^{b-1})\exp(-t/\tau_{\mathrm{LdG}}). \tag{4}$$

Note the differences between this function and Eq. (3). The fit curves are shown as the solid lines through the data in Figs. 5 and 6. All fits start from 0.5 ps, before which the signal is obscured by the very strong electronic polarizability of the sample. If the $at^{-s}$ term is excluded, the rest of the function is identical to the phenomenological function that has been used in the previous analysis of the isotropic phase of nematogens and supercooled liquids.[18] The $at^{-s}$ term is added to model the decay at the very shortest times (0.5 ps to a few picoseconds). The $pt^{-z}$ term corresponds to the intermediate power law with $z$ found to depend on the aspect ratio of nematogens[17]; the $dt^{b-1}$ term is the von Schweidler power law with values of $b$ smaller than but close to 1 for liquid crystals; the exponential function with time constant $\tau_{\mathrm{LdG}}$ describes the LdG decay. The very short time power-law exponent $s$ was found to vary somewhat with temperature and fall between 1.2 and 2.5. However, the time window over which the very short time portion of the data can be observed is insufficient to determine its exact functional form. The intermediate power-law exponent $z$ and the von Schweidler power-law exponent $b$ are temperature independent for both 8CHBT and 5PCH, which is consistent with the earlier results found for other liquid crystals.[17,18] The values for $z$ and $b$ are found to be 0.62 and 0.99 for 8CHBT and 0.72 and 0.96 for 5PCH, respectively. The long time exponential decay obeys LdG theory [see Eqs. (1) and (2)] as displayed in Fig. 7. The LdG temperatures, $T^*$, are given in the plots. LdG theory has been shown many times to be able to describe the long time dynamics well. The short time dynamics, however, are not described by LdG theory. Previous studies on the isotropic phase of nematogens[4,5] and supercooled liquids[30,33] have shown that they have striking resemblances in the orientational dynamics.[18] The data for both types of liquids can be described accurately by the same empirical functional form described by Eq. (4). A modified mode coupling schematic model, which incorporates the aspects of the LdG description, has been recently applied to analyze the dynamics of nematogens in the isotropic phase. The new model was capable of reproducing the data extremely well from a very short time scale (a few hundred femtoseconds a very long time scale (hundreds of nanoseconds), where the LdG exponential decay completes the dynamics.[9]

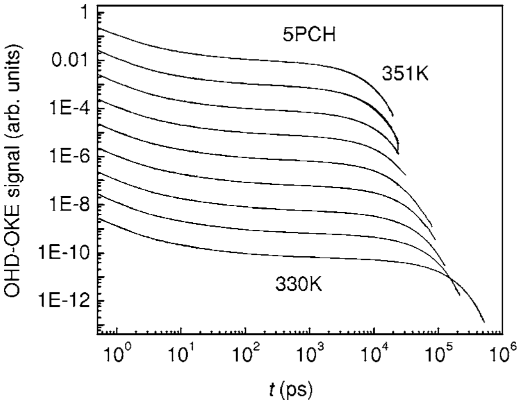

FIG. 6. Temperature-dependent isotropic phase data for 5PCH displayed on a log plot with fits using Eq. (4). The data sets have been offset along the vertical axis for clarity. Starting with the topmost curve, the temperatures for the data sets are 351, 348, 345, 342, 339, 336, 334, 332, and 330 K, respectively.

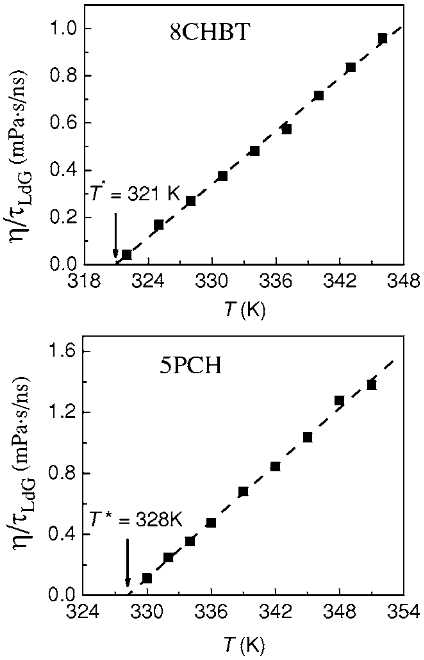

FIG. 7. The temperature dependence of the long time exponential decay constant ($\tau_{\mathrm{LdG}}$) (plotted as $\eta/\tau_{\mathrm{LdG}}$) of the isotropic data on 8CHBT and 5PCH. The dashed lines are the fits using LdG theory [see Eq. (2)] with $V_{\mathrm{eff}}$ and $T^*$ as adjustable parameters. The viscosity data are from the literature (Refs. 35 and 36).

As mentioned above, OHD-OKE experiments measure the time derivative of the polarizability–polarizability (orientation–orientation)-correlation function $\Phi(t)$. Integrals of the empirical functions, Eqs. (3) and (4), would give the decay of the correlation function. However, OKE experiments do not measure the absolute amplitude of the derivative of $\Phi(t)$. The OKE signal has to be scaled by a normalization factor to calculate $\Phi(t)$ correctly. This normalization factor is determined by the integral of the signal (the empirical fitting functions in this case) over $[0,\infty)$. Including the normalization factor, the calculated $\Phi(t)$ from the integral of Eq. (4) for the isotropic phase data sets presented here indicates that it has decayed substantially ($>90\%$) by the end of the data. Further, because the long time decay is the LdG exponential, it is clear that the final 10% of the correlation function will decay exponentially with the LdG decay constant.

The situation is quite different for the nematic phase even though the data have been recorded for times two to three orders of magnitude longer than in the isotropic phase. The integral of Eq. (3) over $[0,\infty)$ does not exist due to the slowly decaying component, the $t^{-f}$ term with $f\sim0.5$. This implies that an additional mechanism, which must decay faster than the final power law, must exist on time scales longer than the longest experiment (70 $\mu$s) to complete the relaxation. (The term final power law means that $t^{-f}$ is the last of the three power laws.) Leslie-Ericksen theory for the long time-scale relaxation of a nematic system to a new director direction following the application of a perturbation that changes the direction of the director[24,25] and experimental results[21,22] suggest that the reorientational dynamics will decay exponentially at very long times. To estimate how much the correlation function has decayed at the end of the experimental time window (70 $\mu$s), the final term in Eq. (3) was multiplied by an exponential, leading to the function

$$F_N(t) = (at^{-s} + pt^{-z})\exp(-t/\tau) + gt^{-f}[1-\exp(-t/\tau)]\exp(-t/\tau_L), \quad (5)$$

where $\tau_L$ is the time constant for the hypothetical exponential decay. The minimum value of $\tau_L$ can be determined. It must be slow enough that it does not influence the functional form of the final power law over the time scale of the experimental data within the signal-to-noise ratio (S/N) of the data. $\tau_L$ is estimated to be greater than 240 $\mu$s. The fraction of the correlation function that has decayed at the end of the time window ($\sim$70 $\mu$s) is estimated to be $<60\%$. Therefore, there is a substantial portion of the correlation function decay that has not been detected by these experiments in spite of the broad range of times covered. Experiments that bridge the gap between this work and those conducted in the hydrodynamic region[19–23] would provide valuable insights into the dynamics and determine if the correlation function decay is indeed exponential following the final power law.

## IV. CONCLUDING REMARKS

The nature of the experiments performed on nematogens in the isotropic phase and the nematic phase have fundamentally different characters. In the isotropic phase, the liquid is macroscopically isotropic with order parameter $S=0$. On a distance scale that is short compared to the correlation length $\xi$ [Eq. (1)], the nematogens form pseudonematic domains with local order parameter $S_L\neq0$. When the pump pulse $E$ field is applied, the nematogens experience a torque that produces a slight alignment with the field. When the field is removed, the macroscopic system is left with $S\neq0$. Field-free evolution will reestablish $S=0$. The restoration of $S=0$ can be roughly divided into two time regimes, the intradomain relaxation and the domain randomization (LdG relaxation). The intradomain relaxation is responsible for the very fast dynamics (less than hundreds of picoseconds to a few nanoseconds, depending on the temperature). Relaxation of the perturbed local order back to its equilibrium value occurs on the fast time scales but leaves the ensemble of local directors still slightly aligned with the direction defined by the $E$ field. This long-lived anisotropy can only decay by randomization of the domains and is responsible for the long time decay described by LdG theory.

In the nematic phase, the sample is macroscopically aligned and has an order parameter $S$ prior to the application of the laser $E$ field. The director is perpendicular to the sample cell window. Call this direction the $z$ direction, and the plane of the window the $x$-$y$ plane. The laser pump beam propagates along $z$, and has its $E$ field in the $x$-$y$ plane. Take the $E$-field direction to be the $x$ direction. The torque exerted by the laser $E$ field on the nematogens will pull them away from the $z$ axis, the director. This will change the order parameter. An anisotropy will develop in the $x$-$y$ plane. The probe $E$ field, which is also in the $x$-$y$ plane, will detect the anisotropy and its decay with time. In the isotropic phase, the system is initially macroscopically isotropic, and it decays (exponentially) to being macroscopically isotropic at a long time. In the nematic phase, the system is initially anisotropic with a well-defined order parameter $S$, and it decays back to its initial nematic ordering with order parameter $S$. Within the experimentally observed time window, the final decay is a power law with an exponent $\sim$0.5 for all three liquid crystals studied.

For experiments on nematogens in the isotropic phase on long time scales, LdG theory has shown the general nature of the observed exponential dynamics. The current experiments on the two nematogens in the isotropic phase combined with previous experiments on four other nematogens in the isotropic phase[5,17,18] demonstrate that the dynamics of nematogens in the isotropic phase display identical characteristics on all time scales. These characteristics have recently been reproduced by a schematic mode coupling model.[9] The main emphasis here has been the dynamics in the nematic phase over a very broad range of times, from subpicoseconds to tens of microseconds. With the presentation of data for two nematogens here, we now have three examples of the dynamics in the nematic phase that demonstrate the universal characteristics of orientational fluctuations about the director. These characteristics are a long time-scale temperature-independent power law with an exponent $\sim$0.5 (the final power law), a crossover region to a shorter time-scale power law (the intermediate power law) that is temperature dependent with an exponent that appears to be linearly dependent on the order parameter, and a very short time power law on time scales $<2$ ps. Analysis of the data suggests that the final power law does not lead to full relaxation back to the initial nematic structure, but rather it must be followed by a decay, probably an exponential decay, that completes the relaxation. To date, there is no theoretical description of the nematic phase dynamical data presented in this paper.

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