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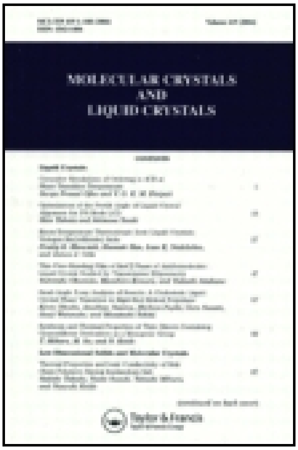

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Dynamical Behavior of Liquid Crystals Containing Dispersed Silica Particles Near Sm A - N and N - I Phase Transitions

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Dynamical Behavior of Liquid Crystals Containing Dispersed Silica Particles Near Sm A – N and N – I Phase Transitions

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The dynamical behavior of liquid crystal 8CB containing 2% of dispersed silica particles was investigated using photon correlation spectroscopy. The behavior differs from the one observed in the bulk. Near the phase transition from the nematic to the smectic A phase the orientational fluctuations of nematic director are drastically slower than in the bulk and this strongly resembles the observations in liquid crystals embedded in silica aerogel. The phase transition from the nematic to the isotropic phase is less affected by the presence of the silica particles but also in this case a few tenth of degree below the phase transition temperature a slowing down of the orientational fluctuations was observed.

Keywords: silica particles; liquid crystals; dynamic light scattering

INTRODUCTION

Recently many studies have been devoted to the effect of randomness on phase transition behavior. Liquid crystalline systems are very convenient for such studies since they are experimentally very accessible and exhibit a variety of phase transitions[1]. It is easy to induce various degrees of disorder in the structure of liquid crystals by either incorporating them into the porous materials or dispersing small particles in them. Calorimetric studies[2-5] show that in such random systems the phase transition regions become broadened and usually shifted with respect to the bulk transitions temperatures.

Dispersions of silica particles in liquid crystals are primary interesting for applications in memory devices[1]. The system is, however, interesting also for fundamental research of effects of disorder on the phase transitions. Small angle neutron scattering (SANS) showed that the silica particles form loosely connected porous aggregates[6], which impose disorder upon the orientation of the liquid crystal. The system is similar to liquid crystal - silica aerogel systems and indeed calorimetric studies showed resemblance, i.e., shifted liquid crystal phase transition temperatures and substantial changes in the shape of excess heat capacity peaks[3,5]. Also static light scattering experiments were performed to study the effect of quenched disorder on the long range order of nematic liquid crystals[7].

A powerful tool to investigate the dynamical behavior of liquid crystals is photon correlation spectroscopy[8,9]. Recently many different random liquid crystal systems have been investigated by this technique. Mostly attention has focused on the behavior of the liquid crystals embedded in various porous matrices[10-12]. Bellini *et al.*[12] have studied dynamically scattered light in the nematic and the smectic-A liquid crystal in an aerogel host. They found out very complex dynamical behavior that differs from the bulk.

In this paper we present a study of the orientational dynamics of the liquid crystal octylcyanobiphenyl (8CB) containing dispersed silica particles at the nematic-isotropic and the smectic A - nematic phase transitions using photon correlation spectroscopy.

EXPERIMENTS

In our experiments we used a standard photon correlation spectroscopy setup. The light source was a He-Ne laser with the wavelength of 632.8 nm. The intensity correlation function $g^{(2)}(\tau) = \langle I(t)I(t+\tau)\rangle / \langle I(t)\rangle\langle I(t+\tau)\rangle$ was measured using an ALV5000 correlator that enables measurements over a time range of 10^{-8} - 10^{3} seconds. We have measured the normalized intensity correlation function of light exiting the sample as a function of temperature. Following the selection rules, and to minimize the elastically scattered background, we chose orthogonal polarizations of incident and scattered light. The amount of

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statically scattered light off the sample was such that the measurements were in the heterodyne regime and the relaxation rate of the intensity correlation function is equal to the relaxation rate of the fluctuations.

The investigated sample consists of 2 wt% of hydrophobic silica particles (R 974) of diameter 12 nm dispersed in the liquid crystal octylcyanobiphenyl (8CB) that exhibit the nematic (T_{NI} = 313.6 K) and the smectic A (T_{AN} = 306.6 K) mesophases. The thickness of the sample placed between two glass plates was 10 μm. The same dispersion was studied also by small angle neutron scattering (SANS)[13].

Nematic - isotropic phase transition

In nematic phase the dispersion of silica particles in liquid crystal can be considered as polydomain system consisting of domains in which nematic director is uniform. In ordered bulk nematic liquid crystals at a given scattering vector there are two independent orientational modes that decay exponentially[9] and can be observed in the dynamic light scattering experiments as a single exponential contributions to the correlation function. On the other hand, in polydomain nematic liquid crystal due to the averaging over the different orientation of nematic director the scattered light correlation function can be described by a stretched exponential $\exp(-(t/\tau)^s)$ with stretching exponent $s \sim 0.8$[14] and average relaxation rate $1/\tau$.

The measured correlation function in the nematic phase of the composite system is indeed stretched exponential with the stretching exponent approximately 0.8 (Fig. 1). The temperature behavior of the average relaxation rate in the nematic phase is different than in the bulk. While in the bulk average relaxation rate remains almost constant in studied temperature region, in composite system it slowly decreases with temperature. The difference from the bulk becomes more pronounced a few tenths of a degree below the phase transition to the isotropic phase, where the average relaxation rate drastically decreases and the stretching exponent becomes smaller (Fig. 1). Smaller value of stretching exponent indicates the distribution of relaxation rates of the fluctuations is broader. This can be understood considering that the calorimetric studies of such systems show in this

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temperature region coexistence of phases[5]. Since the relaxation rate depends on the size of the region in the nematic phase and in the coexistence of the phases the size of nematic parts varies this can be the reason for broadened correlation function.

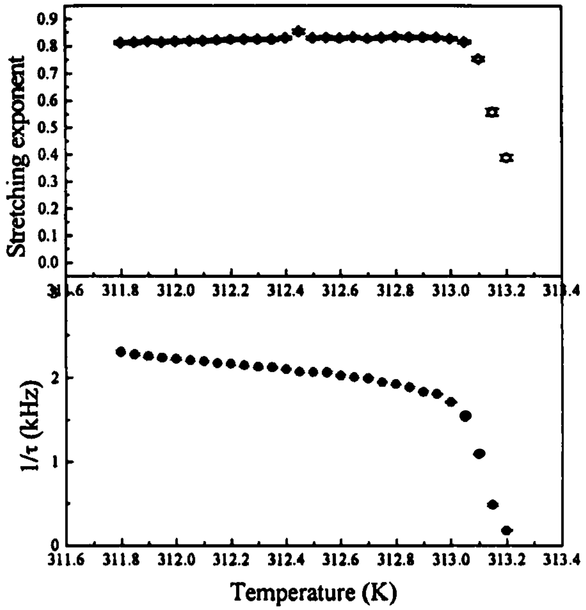

FIGURE 1: Temperature dependence of the stretching exponent and relaxation rate near the nematic - isotropic phase transition. The outer scattering angle is 30°.

Nematic - smectic A phase transition

Besides well defined nearly exponential part described in the previous subsection the measured correlation function already in the nematic phase exhibits a long logarithmic tail. When approaching the phase transition temperature to the smectic A phase the amplitude of this tail grows (Fig. 2).

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This is very similar to the behavior of liquid crystal embedded in the aerogel[12].

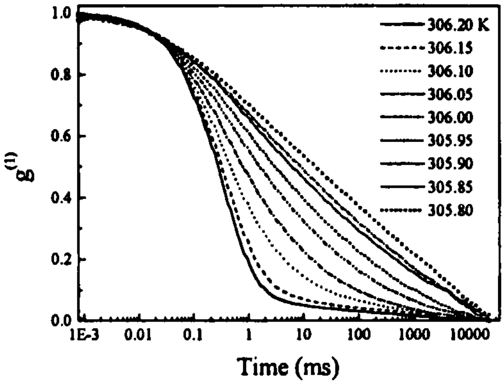

FIGURE 2: Measured scattered intensity correlation function for different temperatures at the nematic - smectic A phase transition.

We have fit the correlation function with two parts, both of stretched exponential form. The first, with fixed stretching exponent of 0.81 determined in the nematic phase, corresponds to the usual director orientational fluctuations. Their relaxation rate in the region of the transition does not show any significant temperature dependence. The situation is, however, different for the second part that describes the tail. Both the relaxation rate and the stretching exponent of the second part exhibit a discontinuity at a certain temperature that depends on whether the system is cooled or heated (Fig. 3). Below this temperature the relaxation rate exponentially decreases with temperature. It goes from a few tenths of a kHz down to 10^{-5} kHz, which is also the experimental limit of our setup. The fit $\exp(a+bT)$ to the data gives b ~ 10 K^{-1}. This behavior is not typical for conventional random-field systems which follows the Vogel-Fulcher law, i.e., relaxation rate ~ $\exp(-B/(T-T^*))$. Similar temperature dependence of the relaxation rate was also observed in the liquid crystal in the aerogel[12], but there the relaxation rate saturates at the temperature where correlation length of the smectic phase become

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comparable to the typical pore size of the aerogel. No such saturation of the relaxation rate in the liquid crystal - aerosil system suggests that no typical length as a pore size exists in this system. This is in agreement with the SANS experiments in these systems[6].

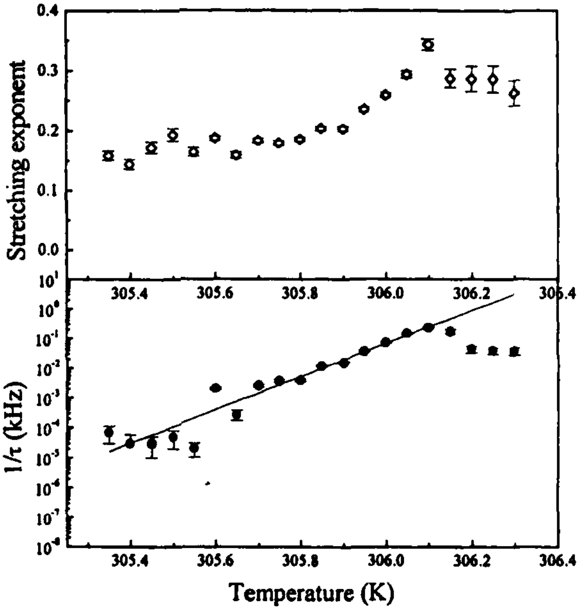

FIGURE 3: Stretching exponent and characteristic relaxation rate of the tail of the correlation function vs. temperature. Line is exponential fit exp(a+b*T*) to the data with parameter b ~ 10 K^{-1}.

CONCLUSIONS

In conclusion, we have studied the dynamical behavior of liquid crystal containing dispersed silica particles near the nematic - isotropic and the smectic A - nematic phase transitions. In the temperature region where calorimetric studies show the coexistence of the nematic and the isotropic phases the relaxation rate of the director orientational fluctuations drastically

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decreases. More interesting is dynamical behavior around and below the nematic - smectic A phase transition temperature. Similarly as in liquid crystal embedded in silica aerogel also in this system a process with a broad distribution of relaxation times is observed which amplitude significantly increases when approaching phase transition to the smectic A phase. In order to better understand the dynamical behavior close to this phase transition further experiments have to be performed, e.g. dependence on the concentration of silica particles.

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