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The Effect of Magnetic Field and Convective Flow on Nematic Director Fluctuations

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The slow orientational fluctuations of the director in the nematic state of d-PAA have been studied by real-time neutron scattering. By analysis of the measured time series we have derived the Hurst exponent H as a function of the applied magnetic field and of the applied vertical temperature difference of the sample. For zero values of both parameters we found $H \geqslant 0.9$, indicating a high degree of temporal correlation between orientational fluctuations. At increasing field or temperature gradient H approaches a value of 0.5, corresponding to zero temporal correlations.

KEY WORDS: Neutron scattering; liquid crystal; director fluctuations; Hurst exponent.

1. INTRODUCTION

Some of the applications of nematic liquid crystals in display devices make use of their strong scattering power for light. De Gennes[1] showed that the scattering is caused by orientational fluctuations of the director. The wavelength scale of the orientational fluctuations extends over several orders of magnitude, from the dimension of a molecule to the size of the sample container. The long-wavelength part is due to fluctuations of the director rather than those of the order parameter.

In a Fourier representation the thermal average of fluctuations of the transverse components of the director is given by[1]

$$\langle |n_\perp(q)|^2 \rangle \sim \frac{k_B T}{(Kq^2 + \chi_a h^2)} \tag{1}$$

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K is a suitable combination of the three elastic constants, χ_a is the anisotropic diamagnetic susceptibility, and h is the applied magnetic field. Introducing the magnetic correlation length

$$\xi = (K/\chi_a)^{1/2}\,(1/h) \tag{2}$$

in the denominator of (1), we obtain the correlation function

$$\langle n_\perp(0)\, n_\perp(r)\rangle \sim \frac{1}{r}\, e^{-r/\xi} \tag{3}$$

In zero field ξ is infinite, and the correlations have a slow, power-law falloff as $1/r$.

For $h > 0$ the intensity of the fluctuations is reduced according to (1) and the correlation length becomes finite.

The correlations are subject to a viscous type of relaxation[2] with a rate Ω, for $h = 0$, given by

$$\Omega = D_0 q^2 \tag{4}$$

The diffusion constant for orientation is $D_0 = k/\eta$, η being an averaged viscosity coefficient. As $q \to 0$, Ω^{-1} becomes infinite. Together with Eq. (3), this implies that the long-wavelength director fluctuations, in zero field, have the character of critical fluctuations throughout the entire nematic phase.

We have so far tacitly assumed that there is no convective flow in the nematic sample. In the presence of flow, the velocity gradient exerts an aligning torque on the nematic director, in the same way as a magnetic field. Hence an applied thermal gradient is also expected to reduce the intensity of the director fluctuations and to make ξ become finite.

The dynamics of correlated temporal fluctuations has been termed fractional Brownian motion by Mandelbrot,[3] as distinct from ordinary Brownian motion or white noise. Fractional Brownian motion is characterized by a $1/\omega^r$ power spectrum with $r > 0$, a spectral form found in the analysis of many natural phenomena. For white noise $r = 0$. According to Bak *et al.*,[4] nonequilibrium fractal systems self-organize to a critical state with a $1/\omega$ spectrum.

In a bulk sample coherent neutron scattering is an ideal tool for studying director fluctuations. Formula (4) gives $\tau \sim 10^3$ sec for $q^{-1} \sim 0.1$ cm, for d-PAA. Hence these fluctuations are sufficiently slow that they may be studied by a real-time technique, i.e., by recording a time series of the neutron scattering intensity. If the incident beam is limited by an entrance slit of dimension a, only fluctuations of $q < 1/a$ are sampled, whereas shorter-wavelength fluctuations are averaged out in real-time observations.

Some of the results of our experiments have been reported earlier.[5-7] In this paper we study more closely the effect on the director fluctuations of field alignment and of flow alignment.

2. EXPERIMENTAL METHOD AND RESULTS

2.1. Determination of the Threshold Field and Threshold Temperature Gradient for Director Alignment

The sample used is fully deuterated *para*-azoxy-anisole (d-PAA). It melts to the nematic phase at 119°C and further to the isotropic liquid phase at 135°C. The sample dimensions are $3 \times 3 \times 0.3\ \mathrm{cm}^3$, as defined by a vertical slab aluminum vessel. The neutron spectrometer is set to record scattering from the first diffraction peak at $1.8\ \text{Å}^{-1}$. For a given temperature and field, the intensity of this peak depends only on the orientation of the director relative to the scattering vector.[8] Orientational fluctuations, therefore, manifest themselves as temporal variation of the intensity $I(t)$ of scattered neutrons:

$$\overline{(I(t)-\bar{I})^2} \sim \langle |n_\perp(q)|^2 \rangle \tag{5}$$

where averages on the left are time averages. The relevant dimension of the q-vector is given by the size of the irradiated sample.

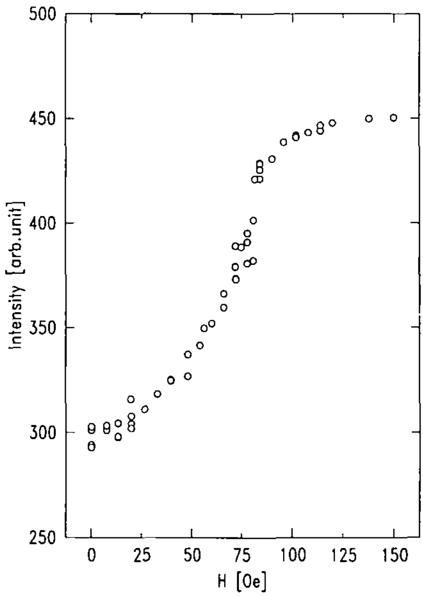

Fig. 1. The time-averaged neutron intensity from nematic d-PAA versus applied vertical field, in the absence of a temperature gradient.

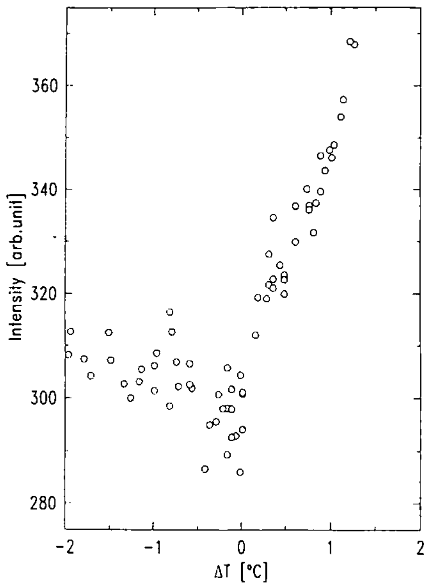

Fig. 2. The time-averaged neutron intensity versus applied vertical temperature difference, in zero external field. Positive values mean heating from below.

In most of the experiments we collected an intensity–time series of 2000 points, with 20 sec counting for each point. A magnetic field could be applied in the vertical direction, along a long dimension of the sample and normal to the scattering vector. Figure 1 gives the time-averaged scattered neutron intensity as a function of the applied magnetic field. It appears from these observations that it takes only ~ 75 Oe to introduce a common director alignment in the sample, i.e., to break the orientational symmetry. No precautions were taken to prepare the anchoring conditions of the molecules at the walls of the sample cell. For a properly prepared cell one expects a well-defined threshold field h_C, the socalled Freedericksz field, to align the director. The field h_C is defined by (2) when setting $\xi = d/\pi$, corresponding to a distortion of half-wavelength d, a characteristic dimension of the sample. Taking the smallest dimension 0.3 cm for d, one calculates $h_C \sim 25$ Oe for d-PAA, which agrees roughly with Fig. 1.

In the present experiments we took great care to determine the threshold gradient for convective flow. The sample container is provided with electrical heating coils at the bottom and top lids, and thermistors and thermocouples monitor the temperature there and on the side. Throughout our experiments the temperature at the bottom plate was kept at 119°C.

Figure 2 give the time-averaged scattered neutron intensity as a function of the applied gradient. The aligning effect of the flow is due to the coupling between orientation and flow, and the curve shows that the threshold value ΔT_C for convective flow is $0.25 \pm 0.20°C$. The zero of the ΔT scale was chosen at the point where an equal amount of electrical power was fed to the top and bottom heating elements.[2]

2.2. Data Processing

A recorded time series is given in Fig. 3a. We have tried different methods for the processing of such data. The most effective use of the data is to base the analysis on the accumulated fluctuations, i.e., on the increment function

$$X(t, \tau) = \sum_{u=1}^{t} [I(u) - \bar{I}_\tau] \tag{6}$$

where τ is the length of the time series. Figure 3b gives $\chi(t, \tau)$ derived from Fig. 3a.

Following Hurst,[9] wa can now define the range R and standard deviation S over the time span τ by

$$R = \max X(t, \tau) - \min X(t, \tau) \tag{7}$$

$$S = \left\{ \frac{1}{\tau} \sum_{t=1}^{\tau} [I(t) - \bar{I}_\tau]^2 \right\} \tag{8}$$

Quite generally

$$\frac{R}{S} \sim \left(\frac{\tau}{2} \right)^H \tag{9}$$

For independent, random fluctuations the Hurst exponent is $H = 1/2$, but for correlated fluctuations $H \neq 1/2$.[3]

The power spectrum $P_X(\omega)$ of $X(t)$ is then given by

$$P_X(\omega) \sim \omega^{-\beta} \tag{10}$$

[2] The zero temperature used here is 0.9°C lower than used in ref. 6.

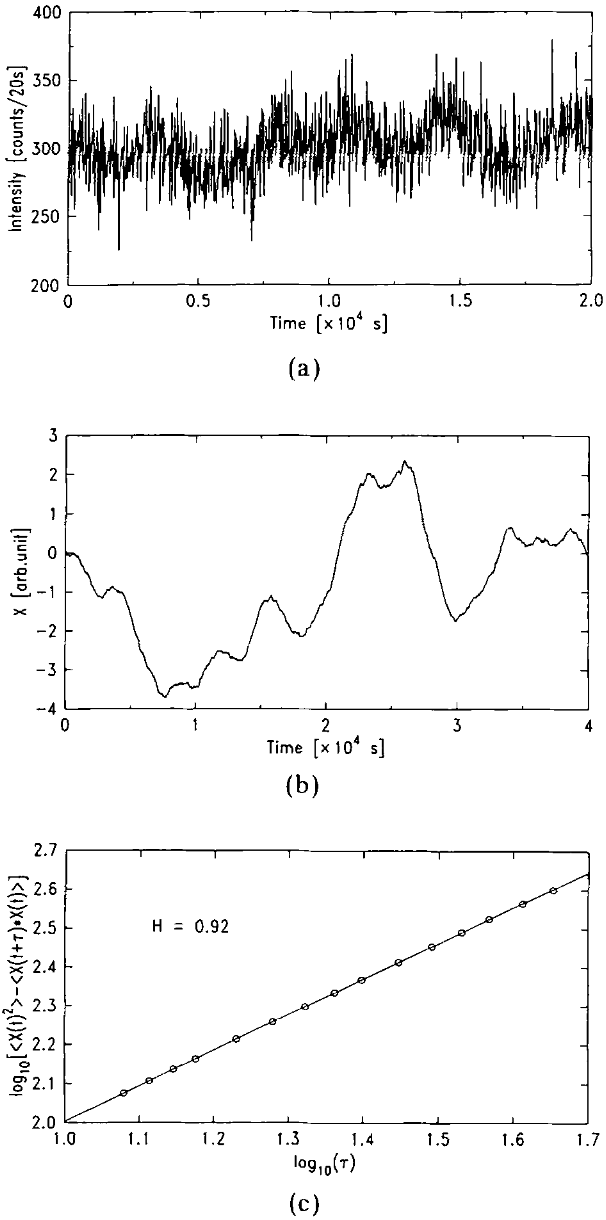

Fig. 3. An example of raw and processed data at $\Delta T = 0$ and $h = 20$ Oe. (a) Portion of a time series. (b) Increment function X calculated from (a) by means of Eq. (6). (c) A curve calculated by a correlation analysis of (b), giving the Hurst exponent H.

where

$$\beta = 2H + 1 \tag{11}$$

As indicated above, the power spectrum of $I(t)$ is $\sim \omega^{-r}$. The β and r are related by $r = \beta - 2$.

In our earlier analysis we used formula (9) in deriving H. In the present work we have derived H from the autocorrelation function $C(\tau)$ of $X(t)$ defined by

$$C(\tau) = \overline{X(t+\tau)\, X(t)} \tag{12}$$

with τ here denoting the delay time, and using the relation[10]

$$1 - C(\tau) \sim \tau^{2H} \tag{13}$$

The latter way of deriving H was found to give more consistent values than (9), and could also be used when there was some periodicity in the data.[11] Figure 3c shows a correlation analysis of Fig. 3b.

2.3. Field and Flow Dependence of the Hurst Exponent

In Fig. 4 we show the field dependence of H in the absence of flow. Except for a crossover region around $h = 75$ Oe, the data points reside in a high or low plateau. In the crossover region there is more scatter in the data, but the width of the region corresponds quite well to the sloping part of Fig. 1. Formula (1) predicts a more gradual reduction of the fluctuations by the field that what is seen in Fig. 4, but formula (1) bears no direct

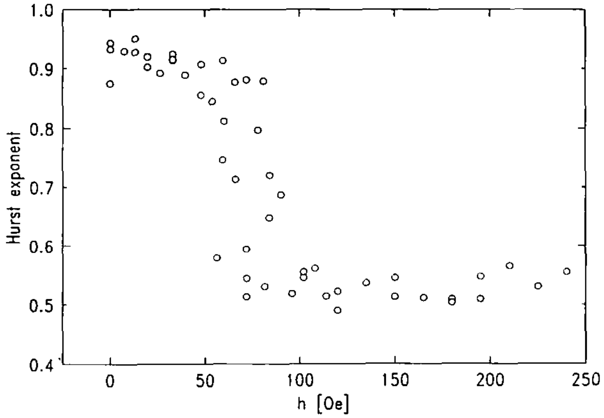

Fig. 4. Plot of the Hurst exponent, derived as in Fig. 3c, as a function of the applied magnetic field, in the absence of a temperature gradient.

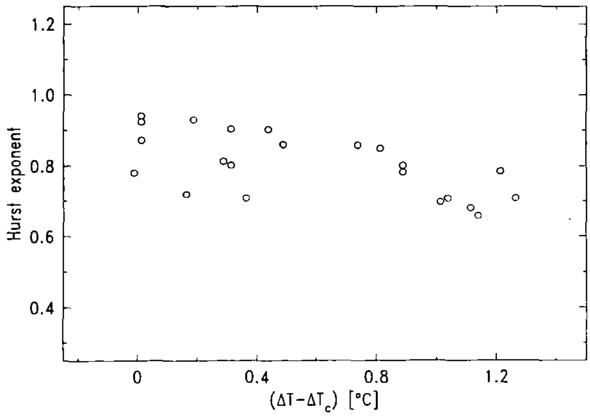

Fig. 5. Plot of the Hurst exponent as a function of the applied vertical temperature difference, and with zero external field.

relation to H. The data in the last figure were taken when the vertical temperature difference ΔT was adjusted to zero.

Next we held $h = 0$ and studied the dependence on ΔT, as shown in Fig. 5. It is clearly seen that H decreases more slowly with ΔT than with h, as can be expected from the following qualitative argument: According to formula (1), a reduction in the fluctuations can be caused by an aligning magnetic torque $\chi_a h^2$. A reduction can also be obtained by a viscous torque $\alpha(\nabla u)$, where α is an appropriate combination of the nematic viscosity coefficients and ∇u is the velocity gradient.[1] For laminar,

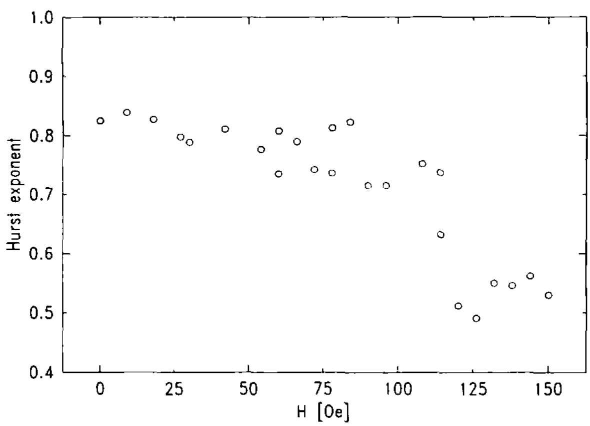

Fig. 6. At a fixed $\Delta T = 0.85°$C, plot of the Hurst exponent versus the applied magnetic field.

convective flow we can further write $\nabla u \sim u \sim (\Delta T - \Delta T_C)^{1/2}$, which for ΔT_C small would give $\Delta T^{1/2}$ instead of h^2 in the denominator of (1).

Figure 6 shows the combined effect of an applied temperature difference, held constant, and a magnetic field. Comparing with Fig. 4, we see that the crossover field is now higher, due to the competition between the aligning actions of the magnetic and viscous torques.

3. Discussion

In Figs. 4 and 6, the crossover from $H \simeq 1$ at low magnetic fields to $H \simeq 0.5$ at high fields implies a crossover from statistical dependence to independence of the director fluctuations as a symmetry-breaking Freedericksz field is applied. When only a vertical temperature gradient is applied, the change in the value of H as a function of the gradient (Fig. 5) is more gradual. It should be pointed out however, that in all three figures, the data giving Hurst exponents in the range $1/2 < H < 1$ were always of the same quality as those giving $H \simeq 1$ and $H \simeq 0.5$, i.e., they defined a unique exponent over the whole time range, and not just in an asymptotic time range. This seems to be in conflict with the theoretical simulations presented in ref. 7, which gave either 0.5 or 1, depending on the time range. This raises the question whether scale-invariant processes other than fractional Brownian motion should also be considered,[3] such as some kind of Lévy process.[(12)]

In any case, a high value of H is connected with scale-variance and power-law correlations. As already pointed out by de Gennes,[(1)] the nematic phase in this respect belongs to a wider class of physical systems in which (a) the ordered state is characterized by a priveleged axis ($\mathbf{u}_0$), but the direction of $\mathbf{u}_0$ is arbitrary, and (b) the interactions are short range.

A Heisenberg ferromagnet in zero field fulfills these criteria. Zhang *et al.*[(13)] have recently analyzed the temporal magnetization-direction fluctuations in a three-dimensional Heisenberg ferromagnet by Monte Carlo simulations and found a field-induced crossover from $H \sim 1$ to $H \sim 0.5$, just as for a liquid crystal. In a monodomain ferromagnetic sample this phenomenon might be studied with the real-time neutron scattering technique.

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[3] We are indebted to Prof. J. Klafter for making this suggestion.

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