Neutron Scattering Studies on Reentrant Nematic Phase

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NEUTRON SCATTERING STUDIES ON REENTRANT NEMATIC PHASE

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__Abstract__   The binary mixtures of 6OCB and 8OCB exhibit reentrant nematic behavior over a narrow range of compositions. We have studied a mixture having 27.12% of 6OCB by weight by neutron scattering technique. The experimental intensity values I(Q) are fitted with a Lorentzian function. From the fitted parameters we have calculated the short-range correlation length (ξ) perpendicular to the director ($\hat{n}$) and the intermolecular distance (D) throughout the mesomorphic range. The results are discussed.

INTRODUCTION

The reentrant nematic phase has been discovered by P.E.Cladis[1] in 1975. This phase has been observed in mixture[1,2] as well as in single component at both high pressure[3] and atmospheric pressure.[4] Soon after its discovery, this reentrant phase has been studied extensively. There have been questions as whether the reentrant nematic and the normal nematic phases are identical. The nature of the nematic to smectic A (NA) and smectic A to reentrant nematic (AN$'$) transitions have also been investigated.[1,2] X-ray measurements suggested that the two nematic phases are similar.[5] Lushington et al.[6] have confirmed from their calorimetric studies on some systems that NA and AN$'$ transitions are not of first order type. Here we have studied the binary mixtures of hexyloxy cyanobiphenyl (6OCB) and octyloxy cyanobiphenyl (8OCB), one of the most common systems showing the reentrant nematic phase, by neutron scattering technique. The aim of this work is to find out how the short-range ordering (SRO) changes as we go from the high temperature nematic to reentrant nematic phase. The birefringence in the mixtures of 6OCB/8OCB has been studied by Chen et al.[7] near the NA and AN$'$ transitions. The variation of the orientational order parameter $\langle P_2 \rangle$ of this binary system with composition from X-ray diffraction and optical birefringence techniques have been studied by Das and Paul.[8]

EXPERIMENTAL

The chemicals 6OCB and 8OCB were obtained from E. Merck, U.K. (formerly BDH, U.K.) and F. Hoffmann-La-Roche and Co., Basel Switzerland respectively. The samples were used without further purification. After mixing the weighed amount of components, each mixture was heated to a temperature slightly above the nematic-isotropic transition temperature of 8OCB and kept at that temperature for more than 48 hours to ensure the formation of a homogeneous mixture. Transition temperatures of the pure compounds and these mixtures were determined using Mettler FP52 Thermosystem and Leitz Polarizing Microscope. The phase diagram we obtained agrees with that reported by others.[7,8,9] In this study we have taken a mixture having 27.12% of 6OCB. The transition temperatures of the phase sequence of this mixture are given below

$$\text{Solid} \ (24^\circ C) \ N_r \ (32.8^\circ C) \ \text{Sm A} \ (41.7^\circ C) \ N \ (77.9^\circ C) \ \text{Iso.}$$

For neutron scattering experiment the sample was put in a thin walled cylindrical container made of aluminium (size : 15 mm length and 7 mm diameter). The sample container is fitted with side wings of aluminium sheet which was wound by non-magnetic heating coil. The sample was electrically heated, the temperature being controlled by a temperature controller (Indotherm 401) with a temperature uncertainty of $\pm 0.5^\circ C$. The sample was first heated to its isotropic state and then cooled down very slowly to the desired temperature in the presence of a magnetic field of 2.5 KOe of a permanent magnet so as to align the sample in the vertical direction. The whole heating and cooling cycle was repeated several times to make sure of its alignment. The neutron scattering experiment was done at Dhruva Reactor of Bhabha Atomic Research Centre, Bombay. The wavelength of the monochromatic incident neutrons was 1.094 Å. The scattered neutrons were recorded by using the Profile Analysis Spectrometer (PAS) at TT-1015 at Dhruva,[10] which can horizontally scan an angular range of 30° at a time. The resolution of the spectrometer ($\Delta d/d$) is ~1%. From the horizontal linear scan of the scattered neutrons recorded by the PAS we obtained diffuse equatorial maximum which is related to the positional short-range order (SRO) perpendicular to the nematic director ($\tilde{n}$).

RESULTS AND DISCUSSION

We have assumed the Lorentzian functional form for the scattered intensity profile with linear background in the vicinity of the peak as[11]

$$I(Q) = I_b(Q) + c/[k^2 + (Q - Q_o)^2] \tag{1}$$

with $I_b(Q) = a + bQ$, a background term. Q_o is the magnitude of the wave vector (Q) at the peak position. Here a, b, c, k, and Q_o are treated as fitting parameters. From the fitted values of k and Q_o we have calculated the positional correlation length (ξ) perpendicular to the nematic director ($\tilde{n}$) and the average intermolecular length (D) using the relations,[12] $\xi = 2\pi/k$ and $D = (2\pi/Q_o)$ respectively. Figures 1,2 and 3 are given to show
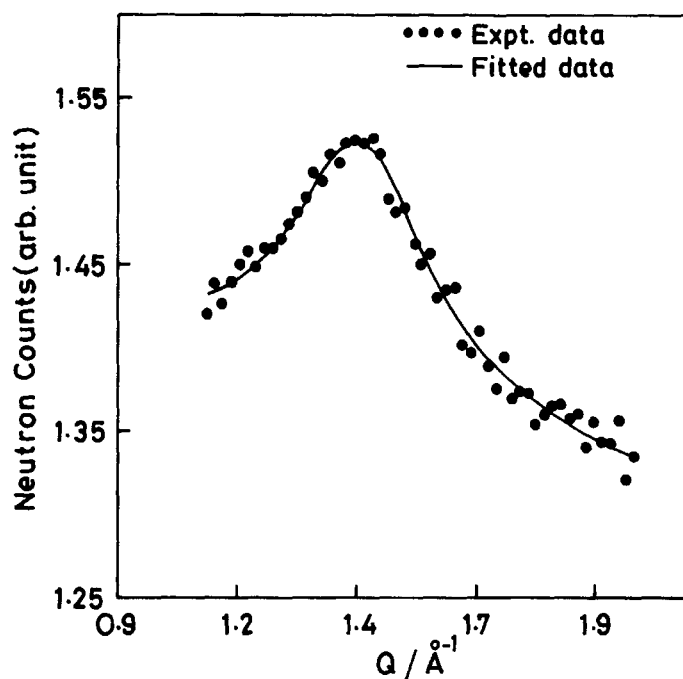
FIGURE 1 Profile of neutron scattering at 28°C (Reentrant nematic phase).
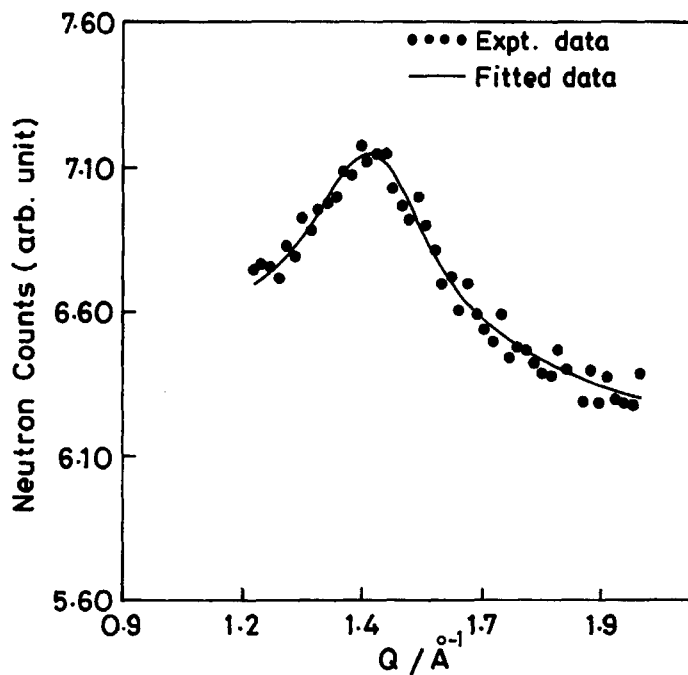

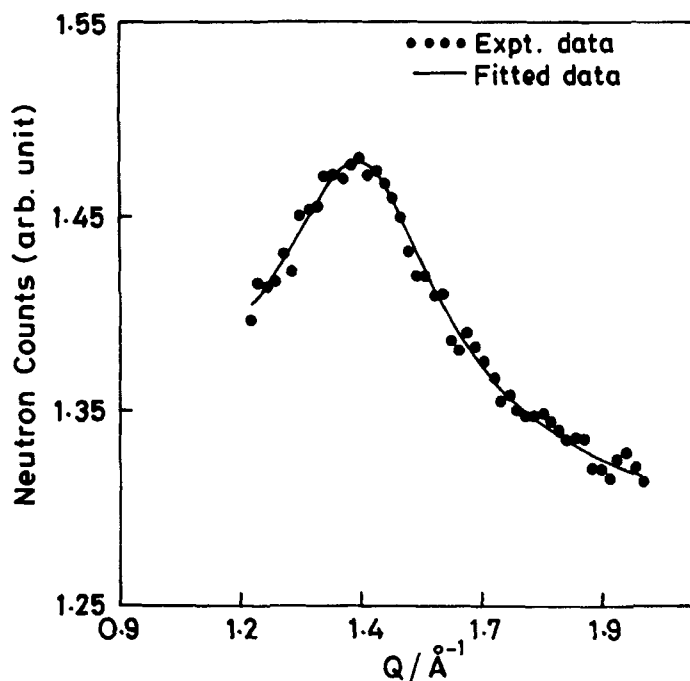FIGURE 2 Profile of neutron scattering at 40°C (Smectic A phase).

FIGURE 3 Experimental and fitted profile of neutron scattering at 60°C (Normal nematic phase).

the experimental and fitted profiles of the neutrons scattering at 28^{C}C (Reentrant nematic), 40°C (Smectic A) and 60°C (normal nematic) respectively for the mixture under study. Table I contains the values of short-range correlation length (ξ) perpendicular to the nematic director ($\tilde{n}$), the intermolecular length (D) and the ratio of ξ/D in different phases. From this Table it can be seen that the D values decreases with temperature in the reentrant nematic phase and it finally tends to the value in the smectic-A phase. In the smectic-A phase the intermolecular distance is almost independent of temperature. But in the high temperature nematic phase, the intermolecular distance is quite sensitive with temperature and it increases with temperature. On the other hand, if we look into the ratio of ξ/D in the reentrant nematic phase we see that this ratio slightly increases as we go towards the smectic-A phase by increasing the temperature. There seems to be no discontinuity in ξ/D values at the AN' transition. This may imply coupling between the reentrant and smectic order parameters as suggested by Cladis.[13] In the smectic-A phase the correlation length is slightly higher than in the nematic phase. In the normal nematic phase, the values of correlation length lies between 7 to 6 times the intermolecular distance (D), which means that in the perpendicular direction of the nematic director ($\tilde{n}$) the molecules becomes uncorrelated after 6 to 7 intermolecular distance. Our observation agrees quite well with the reported values for other nematic compounds.[14,15]

TABLE I

Values of correlation length (ξ), intermolecular length (D) and the ration (ξ/D) at different temperatures

Temp.($^{\circ}$C)	Phase	Value of ξ in Å	Value of D in Å	ξ/D
28.0	N_r	31.71	4.51	6.8
30.0	N_r	31.75	4.46	7.1
35.0	S_A	32.06	4.44	7.2
40.0	S_A	33.56	4.43	7.6
48.0	N	29.9	4.46	6.7
60.0	N	28.89	4.50	6.4
70.0	N	27.89	4.56	6.1
74.5	N	27.19	4.61	5.9

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