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Self-focusing of laser light in the isotropic phase of a nematic liquid crystal*

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Self-focusing of ruby laser light is studied as a function of sample length and temperature in the isotropic phase of nematic liquid-crystal MBBA. The critical power for self-focusing, 0.36 kW, observed near the phase transition temperature is a factor of 20 times less than that for CS_2. The corresponding nonlinear index 4.4×10^{-10} esu is the largest value known so far for any material.

In a previous paper we reported[1] the observation of intense spikes of Stokes Raman radiation in the backward stimulated scattering of nematic liquid-crystal *N*-(*p*-methoxy benzylidene)-*p*-butylaniline (MBBA) in the isotropic phase, probably initiated by self-focusing. In this letter we report the first direct observations of self-focusing and its variation as a function of sample length and temperature in the isotropic phase of MBBA. The interest in liquid crystals for these experiments is due to the large molecular anisotropy and cooperative phenomena. Wong and Shen[2] observed, in agreement with theoretical predictions,[3] that a relatively weak laser field induced appreciable ordering in the isotropic phase of MBBA. The resultant field-induced refractive index should increase strongly as the temperature approaches the isotropic-mesomorphic phase transition. As the threshold for self-focusing varies inversely as the nonlinear index n_2, we expect to observe self-focusing at low laser powers, particularly in the vicinity of the phase transition temperature. This indeed is found to be the case.

The liquid-crystal sample MBBA was obtained from Eastman Kodak. The nematic-to-isotropic phase transition temperature was found to be 46.8 °C. The experimental arrangement consists of a single-mode ruby laser (pulse width, 20 nsec) passively *Q*-switched with cryptocyanine in methanol solution. A pinhole of diameter 0.775 mm was introduced in front of the sample cell to ensure maximum spatial homogeneity of the laser beam. The sample was separated by a distance of over 4 m from the laser to eliminate multiple pulses of amplified Brillouin light. A typical input pulse, detected using a TRG model 105B detector with a S_1 photosurface and displayed on a Tektronix 519 oscilloscope and the Fabry Perot interferogram, are shown in Figs. 1(a) and 1(b). The beam cross section at the exit window was monitored through a microscope objective. The magnified image of the filaments was photographed on Polaroid film. We usually observed a single filament. Occasionally two or three filaments showed up in the picture, particularly at the higher laser powers. The threshold power P_{th} for self-focusing was determined by studying the beam diameter at the exit end as a function of incident power. The patterns obtained are illustrated in Figs. 1(c)–1(f).

The threshold power for self-focusing was studied as a function of temperature for a sample length 30 cm.

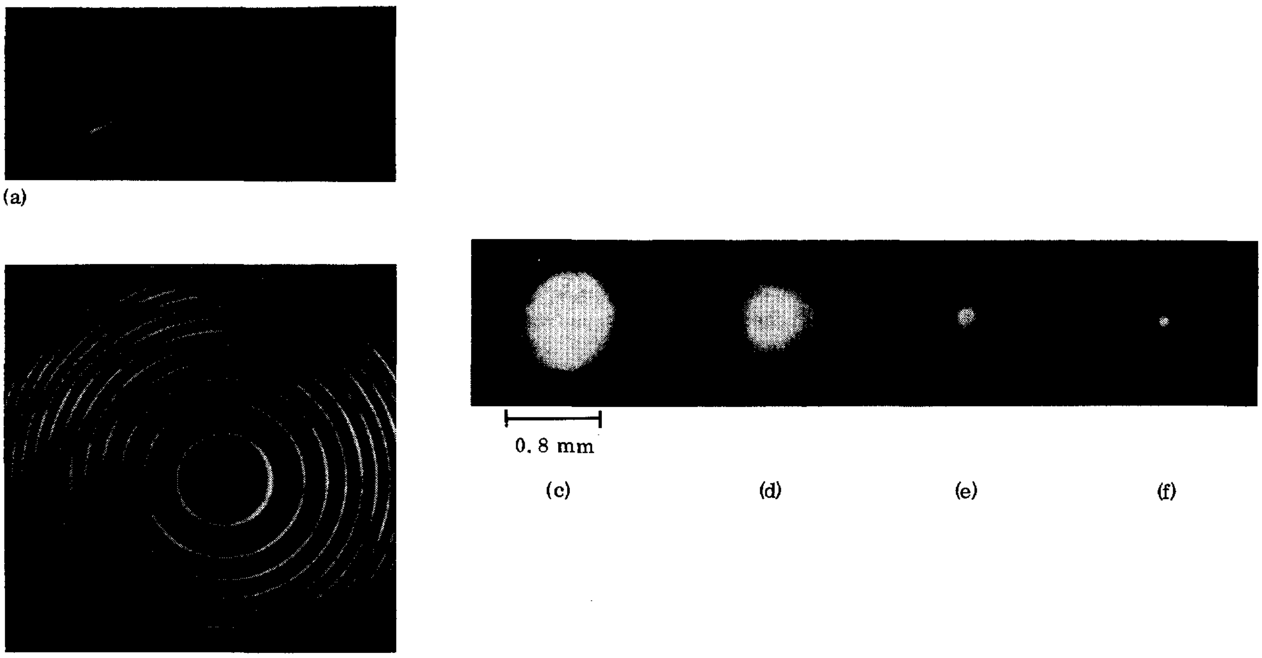

FIG. 1. (a) Typical oscilloscope trace of an input laser pulse; horizontal scale, 20 ns/div. (b) Fabry Perot interferometer pattern of the laser pulse; plate spacing, 1.5 cm. (c)–(f) Image of the laser beam at the exit window for sample length $L = 30$ cm at increasing laser powers of 0.2, 1.0, 8.0, and 16.0 kW, respectively.

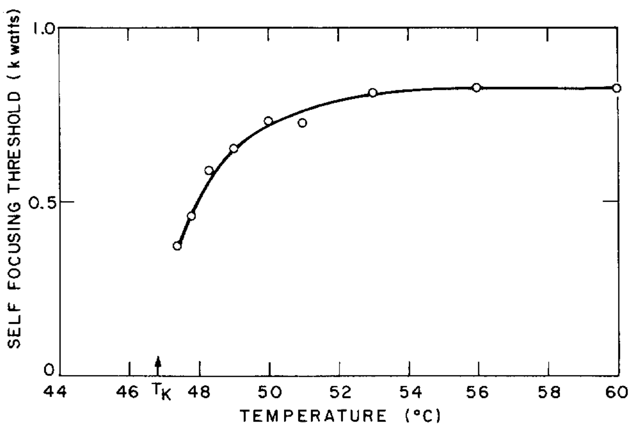

FIG. 2. Self-focusing threshold power as a function of temperature for MBBA sample length 30 cm.

The results are presented in Fig. 2. Our results indicate P_{th} is proportional to $(T-T^*)^\beta$, with the exponent[4] $\beta = 0.16$ and $T^* = 46\,^\circ\mathrm{C}$. Deviations from this relation are, however, observed close to the phase transition, as in the case of other similar experiments[5] which led to the conclusion that the mean field approximation used in theoretical treatment does not hold in the critical region.[6] Since P_{th} is inversely proportional to the nonlinear index, our results are in conformity with Wong and Shen's[2] measurements of the nonlinear index at 53 °C.

We have also carried out measurements of P_{th} as a function of sample length L for two temperatures, 53 and 47.8 °C. Figure 3 illustrates plots of $(P_{th})^{1/2}$ vs $1/L$ which are linear as expected from the theoretical equation[7] for self-focusing $(P_{th})^{1/2} = (P_{cr})^{1/2} + A/L$, with $P_{cr} = (5.763\lambda^2\epsilon_0 c)/4\pi^2 n_2$, where λ is the wavelength of the light beam, ϵ_0 is the vacuum permittivity, and c is the speed of light in vacuum. $A = a(\pi\epsilon_0 c n^2/8n_2)^{1/2}$, where a is the beam radius at which the intensity drops to $e^{-2} = 0.135$ of its on-axis value and n is the field-independent index of the sample. The nonlinear index was calculated using P_{cr} obtained from the plots of Fig. 3. It is significant to observe that $P_{cr} = 0.36$ kW at 47.8 °C, which is a factor of 20 times less than the corresponding value for a strongly self-focusing liquid like CS_2. The value of n_2 at 53 °C is 2.3×10^{-10} esu and at 47.8 °C (close to the phase transition) it is 4.4×10^{-10} esu which is about 20 times the value for CS_2. This is the highest nonlinear index reported for any material until now. It is quite possible that other liquid crystals may have even higher values. The unusually large nonlinear index makes these materials attractive candidates for potential applications in nonlinear optics, e.g., generation of intense short pulses[1] of coherent Raman radiation with a high conversion efficiency.

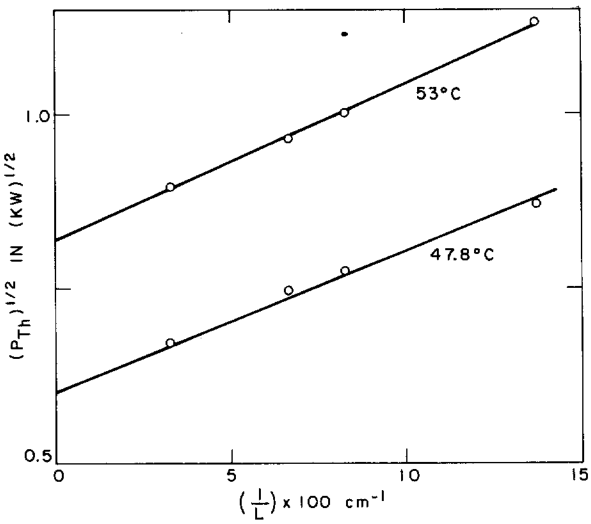

FIG. 3. Plot of $(P_{th})^{1/2}$ vs $1/L$ for self-focusing in MBBA for 47.8 and 53°C.

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[4]The exponent β should be 1 for the steady-state case. However, if we include the transient response of the ordering induced by the short optical pulse, our results are in agreement with de Gennes' theory (Ref. 3; see also Wong and Shen, Ref. 2), except for a small region close to the phase transition temperature. Detailed analysis of the behavior of self-focusing as a function of laser pulse width will be published elsewhere.
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