Pretransitional behavior of self-focusing in nematic liquid crystals*

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We have studied the self-focusing of ruby laser light in the isotropic phase of the nematic liquid crystals *p*-azoxyanisole (PAA), *p*-(*p*-ethoxybenzylidene) amino-benzonitrile (EBAB), and *p*-methoxybenzylidene *p*-*n*-butylaniline (MBBA) as a function of temperature. The observed threshold power for self-focusing follows a $T - T^*$ relationship with the critical exponent unity for PAA and EBAB, in agreement with de Gennes's theory. We could also explain our results for MBBA, observed with three laser pulse widths, taking into account the transient response of the order parameter. Calculated values of the nonlinear index from the observed thresholds for self-focusing are found to be large for the three materials compared to that for CS_2.

I. INTRODUCTION

Self-focusing of an intense light beam in a fluid is a consequence of the converging lens action of an intensity-dependent index of refraction of the form $n = n_0 + n_2 E^2$, where n_0 is the field-independent index of refraction and E is the electric field. The dominant contribution to the nonlinear coefficient n_2 in liquids is due to the optical Kerr effect. Freiser and Joenk[1] considered the enhancement of self-focusing in liquid crystals by cooperative phenomena and obtained a $(T - T_K)^{-1}$ behavior of n_2 with temperature instead of the usual T^{-1} behavior in normal liquids, where T_K is indicative of the ordering temperature of the liquid crystal. Hanus[2] studied in more detail the effect of molecular interaction between anisotropic molecules in liquids on the nonlinear index and showed that the temperature dependence of the nonlinearity is modified to $(T - T_i)^{-1}$, where T_i is the limiting temperature at which the isotropic phase can exist, and that the electric field required to achieve a certain nonlinear index can be very much smaller than that required when only molecular reorientation is considered. de Gennes[3] has given a phenomenological model for describing short-range-order effects in the isotropic phase of nematics and cholesterics. Wong and Shen,[4] and Prost and Lalanne[5] observed, in accordance with de Gennes's predictions, that a relatively weak laser field induced appreciable ordering in the isotropic phase of a nematic. We reported the first direct observation[6] of self-focusing in the isotropic phase of the nematic liquid crystal, *p*-methoxybenzylidene *p*-*n*-butylaniline (MBBA), using *Q*-switched ruby laser light.

This article contains interesting results of the temperature variation of threshold power P_{th} for self-focusing in the isotropic phase of the nematic liquid crystals *p*-azoxyanisole (PAA), *p*-(*p*-ethoxybenzylidene) amino-benzonitrile (EBAB), and MBBA. The motivation for this work is the possibility of studying steady-state as well as transient self-focusing in view of the wide range of relaxation times of the order parameter in these materials. Since P_{th} is inversely proportional to the nonlinear coefficient n_2, which itself is proportional to the order parameter Q in liquid crystals, we interpret the observed behavior in terms of the phenomenological model of Landau[7] as applied by de Gennes[3] for liquid crystals.

Following de Gennes,[3] we may write the Gibbs free energy density for the isotropic phase as

$$F = F_0 + \tfrac{1}{2} A Q^2 - \tfrac{1}{3} \Delta\chi E^2 Q , \quad (1)$$

where the coefficient $A = a(T - T^*)$, T^* is the second-order phase transition temperature somewhat lower than the clearing temperature T_c, a is a constant, E is the electric field, and $\Delta\chi Q$ is the anisotropy in the susceptibility induced by the ordering. The order parameter $Q = \frac{3}{2}\langle \cos^2\theta - \frac{1}{3} \rangle$, where θ is the angle between the long axis of the molecule and the local optic axis, and corresponds to that defined by Maier and Saupe.[8] For a completely aligned nematic phase $Q = 1$. The steady-state value of Q induced by the optical field is obtained by minimization of F with respect to Q and is given by $Q = \Delta\chi E^2/3A$. Q is thus proportional to $1/A$ and hence to $1/(T - T^*)$. Since P_{th} varies as $1/n_2$ and hence as $1/Q$, the self-focusing threshold power for the steady-state case should go as $(T - T^*)^1$. This indeed was found to be the case for PAA and EBAB.

II. EXPERIMENTAL RESULTS: STEADY STATE

The samples of PAA and EBAB were obtained from Eastman Kodak. The clearing temperatures T_c were 134 and 128.5 °C, respectively. The experimental arrangement, illustrated in Fig. 1, consists of a single-mode ruby laser of pulse width 30 nsec passively Q switched with crytpo-

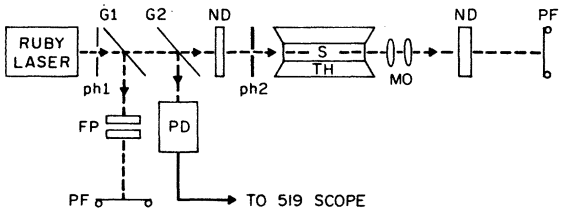

FIG. 1. Experimental setup for self-focusing. G_1, G_2—beam splitters; ph1, ph2—apertures of diameter 3 and 0.775 mm; FP—Fabry-Perot interferometer; PD—photodiode; ND—neutral-density filters; MO—microscope objective; PF—polaroid film; S—liquid-crystal sample cell surrounded by thermostat (TH).

cyanine 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 of length 7 cm 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 105 B detector with S_1 photosurface and displayed on a Tektronix 519 oscilloscope, and the Fabry-Perot interferogram, are shown in Figs. 2(a) and 2(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 patterns obtained for PAA are illustrated in Figs. 2(c)–2(f). The threshold power for self-focusing was determined by studying the beam diameter at

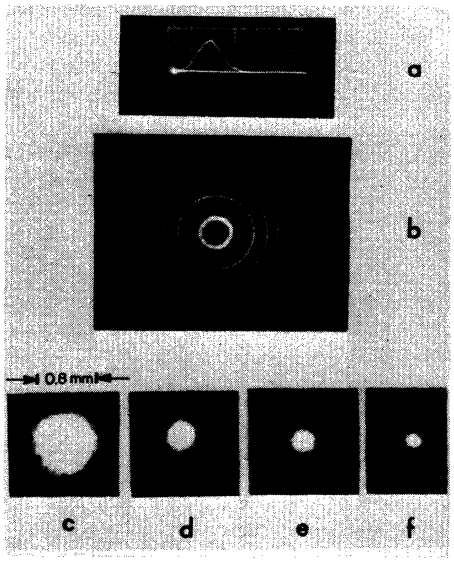

FIG. 2. (a) Typical oscilloscope trace of input laser pulse; horizontal scale 20 nsec/div. (b) Fabry-Perot interferometer pattern of the laser pulse; plate spacing 1.5 cm. (c)–(f) Image of the laser beam at the exit window for PAA sample length 7 cm at increasing laser powers of 0.15, 1.0, 3.0, and 8 kW, respectively.

the exit end as a function of incident power. The beam diameter was plotted against incident power, and the threshold power for the onset of self-focusing was determined where the beam diameter starts shrinking. This gives essentially the critical power in the case of steady-state self-focusing which we believe is the case for PAA and EBAB. In Fig. 3, we present our measured values of P_{th}, corrected for scattering loss of sample, for PAA

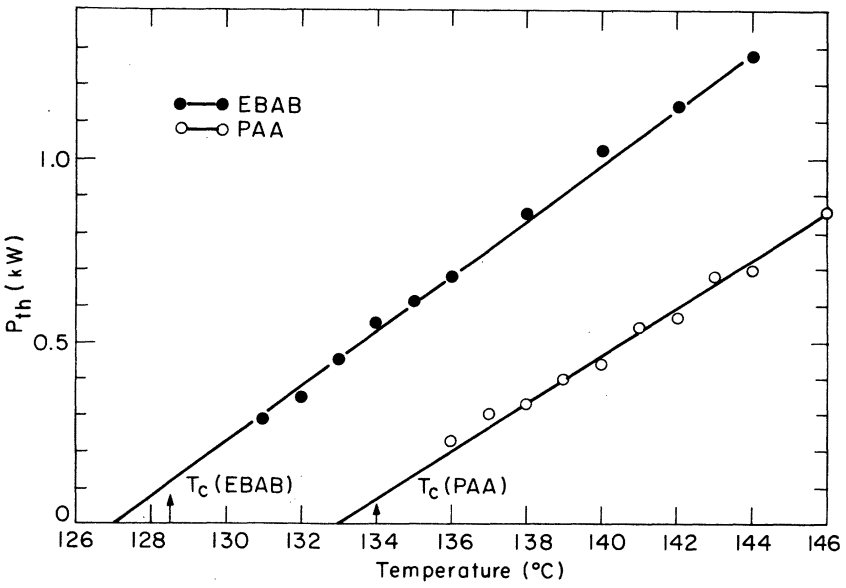

FIG. 3. Self-focusing threshold power P_{th} as a function of temperature for PAA and EBAB; sample length 7 cm.

and EBAB as a function of temperature. Both PAA and EBAB follow $(T-T^*)^1$ dependence, agreeing quantitatively with de Gennes's theory[3] of phase transition in nematic liquid crystals. Values of T^* come out as 133 and 127 °C for PAA and EBAB, about 1 °C lower than the respective clearing temperatures. The relaxation time of EBAB reported[9] from experiments on dc Kerr response is 13 nsec at 1.2 °C above the clearing temperature as compared to 3 μsec for MBBA under similar conditions at the corresponding temperature from the same experiments. The molecular dimensions[10] as well as the viscosity of PAA are smaller than those of EBAB. So it is expected that the relaxation time of PAA would also be much smaller than that of MBBA. We are thus led to believe that the relaxation times of the ordering induced by the optical field for PAA and EBAB are small compared to our laser pulse width; the situation corresponds to steady-state self-focusing and hence the observed temperature dependence with exponent unity. Computed values of n_2 from the observed critical powers for PAA and EBAB at 3 °C above T_c are 0.71 and 0.56×10^{-9} esu, which are large compared to the value 2×10^{-11} esu for CS_2.

III. EXPERIMENTAL RESULTS: TRANSIENT CASE

In the case of MBBA, the relaxation time of the order parameter obtained directly from experiments at optical frequencies varies from 30 nsec to several hundred nanoseconds[4,5,11] as the clearing temperature is approached. Hence the ordering induced by the short optical pulse must be of transient nature. We have here a case of transient self-focusing.[12] The transient response of the order parameter is governed by the equation

$$\nu\frac{\partial Q}{\partial t}+AQ=f(t), \tag{2}$$

with $f(t)=C\Delta\chi I(t)$, where $I(t)$ is the laser pulse intensity, C is a constant, and ν is the viscosity coefficient. The solution to the above equation may be written as

$$Q(t)=\frac{1}{\nu}\int_{-\infty}^{t} f(t')e^{(t'-t)/\tau}\,dt', \tag{3}$$

where $\tau=\nu/A=\nu/a(T-T^*)$ is the relaxation time, and $\nu=\nu_0 e^{(2800/T)}$.[11] Since it is difficult to get an analytic solution for the above integral equation particularly in the case of transient self-focusing, which may occur some time after the peak of the pulse,[13] we use a simple rectangular approximation for the pulse envelope, $f(t)=f_0$, $0\leq t\leq t_p$, where t_p is $1/e$ width of the laser pulse. Then the solution for Q is given by

$$Q=(f_0/A)(1-e^{-t_p/\tau}). \tag{4}$$

Hence P_{th} varies as $(T-T^*)/(1-e^{-t_p/\tau})$. For the steady-state case, $t_p\gg\tau$, P_{th} is proportional to $T-T^*$, thus yielding the same result as before. From the known[11] viscosity and the constant a, we could calculate the temperature dependence of P_{th} for a given t_p. We measured P_{th} for MBBA, sample length 30 cm, as a function of temperature for three different pulse widths 15, 40, and 80 nsec ($1/e$ width). We used fresh samples of MBBA (obtained from Eastman Kodak, $T_c=46.8$ °C) of the same batch for each experiment, since the clearing temperature was found to decrease on second melting, probably owing to molecular dissociation. Figure 4 shows the results along with the theoretical curves calculated using Eq. (4) with $T^*=46$ °C. When $t_p=15$ nsec, $t_p/\tau<1$; so P_{th} varies approximately as $e^{2800/T}$. Our observations do indicate that P_{th} is increasing towards the clearing temperature. This is in conformity with the measurements of Wong and Shen,[4] who also observed in MBBA with a laser pulse 10 nsec long that the nonlinear index decreased towards the clearing temperature. For $t_p=40$ nsec, P_{th} is almost flat and slightly decreasing towards the clearing temperature (these are the results from our previous preliminary report[6] for a pulse width at half-height 20 nsec which corresponds to $1/e$ width ~ 40 nsec). For $t_p=80$ nsec, P_{th} is decreasing as $T\to T_c$. Similar behavior was observed by Prost and Lalanne[5] on the optical Kerr effect induced by laser pulse of width 30 nsec; the optical Kerr constant of MBBA increased towards the clearing temperature. Extrapolating the pulse-width dependence, we expect to observe $(T-T^*)^1$ dependence in the case of MBBA also for $t_p\gg\tau$. The transient na-

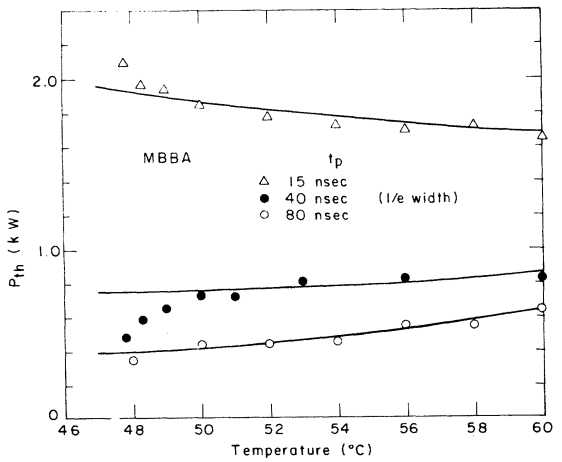

FIG. 4. Self-focusing threshold power P_{th} as a function of temperature for MBBA, sample length 30 cm, for three different laser pulse widths, 15, 40, and 80 nsec. The continuous lines represent theoretically calculated values.

ture of the ordering induced by the optical pulse is responsible for the observed sublinear dependence. Even in this case, our observations are in qualitative agreement with de Gennes's theory. (We find that correction of the results for scattering loss in our sample MBBA, does not affect the conclusions.)

IV. CONCLUSIONS

In conclusion, we have observed a linear $T - T^*$ dependence of the self-focusing threshold in the isotropic phase of the liquid crystals PAA and EBAB. In the case of MBBA, because of the sluggish response of the order parameter, we observed transient self-focusing with a sublinear dependence. It is interesting to point out that it may be possible to observe steady-state self-focusing in MBBA with continuous wave laser beams. From our results on MBBA, we could write a relation for the threshold for self-focusing as $P_{th}(t_p) = P_{cr}(t_p \gg \tau)/(1 - e^{-t_p/\tau})$. For $t_p = 15$ nsec and $\tau = 105$ nsec at 52 °C, we got P_{th} as ~ 1.8 kW. Using these parameters, P_{cr} for MBBA at 52 °C comes out as ~ 240 W. The critical power for steady-state self-focusing goes as $(T - T^*)^1$, and for 3 °C above T^*, $P_{cr} \simeq 120$ W, which corresponds to $n_2 \simeq 1.4 \times 10^{-9}$ esu. Continuous-wave operation of argon lasers at 500 W output power was recently reported[14] in the literature. One can alternatively use the second harmonic of a Nd : YAG laser. Thus the self-focusing phenomenon in Kerr liquids may be brought from high power levels of Q-switched pulsed lasers to the relatively low power levels of continuous-wave lasers.[15]

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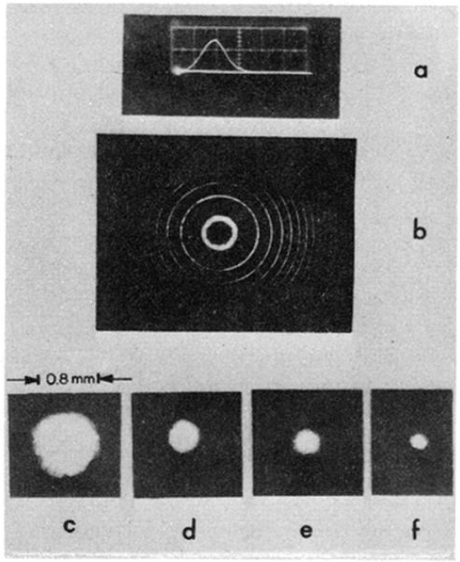

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