

## TEMPERATURE DEPENDENCE OF TRANSIENT SELF-FOCUSING IN THE ISOTROPIC PHASE OF MBVA

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We have shown that the temperature behaviour of the transient self-focusing in the isotropic phase of the nematic liquid crystal MBVA is primarily determined by the temperature dependence of the scattering of light. The theoretical description of self-focusing based on the paraxial solution of the wave equation with the induced refractive index obtained from de Gennes' theory of liquid crystals is in good agreement with the experimental results when scattering is properly taken into account.

### ТЕМПЕРАТУРНАЯ ЗАВИСИМОСТЬ НЕСТАЦИОНАРНОЙ САМОФОКУСРОВКИ В ИЗОТРОПНОЙ ФАЗЕ МБВА

В статье показано, что температурная зависимость нестационарной самофокусировки в изотропной фазе нематического жидкого кристалла МБВА в основном определяется температурной зависимостью рассеяния света. Теоретическое описание, основанное на параксиальном приближенном решении волнового уравнения с нелинейным показателем преломления, полученным по теории жидких кристаллов де Жена, хорошо согласуется с экспериментальными результатами, если принимается во внимание влияние рассеяния света.

### 1. INTRODUCTION

Despite of its having been discovered in 1962 [1], self-focusing of light still remains one of the most interesting phenomena in nonlinear optics. It is of major importance in the design of high-power lasers because of its role in light-induced damages. The self-focusing is of fundamental interest also because of its interplay with many other nonlinear optical processes.

Recently, it has been demonstrated that nematic liquid crystals in their isotropic-liquid phase exhibit a particularly strong electrical [2] and optical [3] Kerr effect and have very low threshold powers for self-focusing [4]. One of the most remarkable features of all these effects is a strong pretransitional behaviour as the

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transition temperature from the isotropic-liquid to the nematic phase is approached. For example, it was found that the response time of the Kerr effect exhibits the characteristic critical slowing down [3]. The response times of some nematic liquid crystals are so large that we can observe transient self-focusing in these materials, when we use nanosecond pulses of a Q-switched laser. The transient self-focusing was observed in the isotropic phase of the nematic liquid crystal p-methoxybenzyliden-p-n-butylaniline (MBBA) [5, 6, 7]. At the same time it was shown that self-focusing in MBBA has a complex temperature dependence of which no satisfactory explanation was given.

In the present paper we show that the temperature behaviour of self-focusing in MBBA can be described on the basis of the Landau-de Gennes model of liquid crystals and the paraxial solution of the nonlinear wave equation when the scattering of light in the isotropic phase of the liquid crystal is taken into account. The theoretical predictions are compared with the experimentally observed temperature dependence of self-focusing in the isotropic phase of MBBA.

## II. THEORY OF SELF-FOCUSING

Nonlinear optical properties of matter result from the nonlinear dependence of polarization  $\mathbf{P}(\mathbf{r}, t)$  on the electric field strength  $\mathbf{E}(\mathbf{r}, t)$  of the light. If we do not take into account the spatial and temporal dispersion, the polarization can be represented by a power series expansion in the electric field of the form [8]

$$\mathbf{P}_a(\mathbf{r}, t) = \epsilon_0 \chi_{aa}^{(1)} E_a(\mathbf{r}, t) + \epsilon_0 \chi_{a\beta\gamma}^{(2)} E_\beta(\mathbf{r}, t) E_\gamma(\mathbf{r}, t) + \epsilon_0 \chi_{a\beta\gamma\delta}^{(3)} E_\beta(\mathbf{r}, t) E_\gamma(\mathbf{r}, t) E_\delta(\mathbf{r}, t) + \dots \quad (1)$$

where  $\epsilon_0$  is the permittivity of free space,  $\chi_{aa}^{(1)}$  is a linear susceptibility tensor,  $\chi_{a\beta\gamma}^{(2)}$  and  $\chi_{a\beta\gamma\delta}^{(3)}$  are nonlinear susceptibility tensors of the second and the third order, respectively. The second order susceptibility tensor vanishes in the isotropic media due to symmetry conditions.

Eq. (1) enables us to write the susceptibility of the nonlinear isotropic medium in the form

$$\chi(E) = \chi_0 + \delta\chi(E), \quad (2)$$

where  $\chi_0$  is the susceptibility independent of the electric field and  $\delta\chi(E)$  is the change in the susceptibility caused by the electric field of a powerful light wave. The index of refraction can be written in a similar way as

$$n(E) = n_0 + \delta n(E), \quad (3)$$

where  $n_0$  is the linear refractive index and  $\delta n(E)$  is the induced nonlinear refractive index.

Since

$$\delta n(E) = \delta\chi(E)/2n_0, \quad (4)$$

the polarization of the isotropic medium can be written as

$$\mathbf{P}(\mathbf{r}, t) = \mathbf{P}^L(\mathbf{r}, t) + 2\epsilon_0 n_0 \delta n(E) \mathbf{E}(\mathbf{r}, t), \quad (5)$$

where  $\mathbf{P}^L(\mathbf{r}, t)$  is the linear part of the polarization vector.

Self-action of a laser beam can be described as a change of the refractive index of the medium caused by the strong electric field of the light and a subsequent change in the propagation of the laser beam. The propagation of the beam is governed by the wave equation with Eq. (5) instead of the linear polarization

$$\nabla^2 \mathbf{E}(\mathbf{r}, t) - \frac{1}{v_l^2} \frac{\partial^2 \mathbf{E}(\mathbf{r}, t)}{\partial t^2} = \frac{2}{n_0 v_l^2} \frac{\partial^2 [\delta n \mathbf{E}(\mathbf{r}, t)]}{\partial t^2}, \quad (6)$$

where  $v_l = c/n_0$ .

For a monochromatic light beam propagating along the  $\hat{z}$  axis and polarized along the  $\hat{x}$  axis we can write

$$\mathbf{E}(\mathbf{r}, t) = E_0(\mathbf{r}, t) \exp [i(\omega t - kz)] \hat{x}, \quad (7)$$

where  $k = n_0 \omega / c$  and  $\omega_0$  is the angular frequency of the incident light. If  $\delta n \ll n_0$  and the amplitude  $E_0(\mathbf{r}, t)$  is a slowly varying function (these limitations on the rate of the spatial and temporal variations of the amplitude are thoroughly discussed in [10]), then Eq. (6) can be transformed into the following equation

$$\frac{1}{r} \frac{\partial}{\partial r} \left[ r \frac{\partial E_0(\mathbf{r}, z, t)}{\partial r} \right] - 2ik \left[ \frac{\partial E_0(\mathbf{r}, z, t)}{\partial z} + \frac{1}{v_l} \frac{\partial E_0(\mathbf{r}, z, t)}{\partial t} \right] + \frac{2\delta n}{n_0} k^2 E_0(\mathbf{r}, z, t) = 0, \quad (8)$$

where we have used the cylindrical coordinate system.

In order to solve this equation, we need to know the functional dependence of  $\delta n$  upon the electric field. The nonlinear refractive index depends on many physical processes, one of which is usually dominant. Each of these processes is characterized by a response time  $\tau$ , which can vary from seconds for thermal effects to femtoseconds for electronic processes. The ratio of the laser pulse length to the response time of the dominant mechanism influences decisively the behaviour of self-focusing. If the pulse length is much greater than the response time of the medium, steady-state self-focusing takes place. If, on the other hand, the pulse length is shorter than the response time, the transient character of the response of the medium dominates the self-focusing behaviour.

In the isotropic phase of nematic liquid crystals, the dominant mechanism giving the largest contribution to  $\delta n$  is molecular reorientation, i.e. the optical Kerr effect

[3]. Though the nematic liquid crystals lose the long-range orientational order at the transition from the nematic to the isotropic phase, considerable short-range orientational correlations are preserved. The molecules cannot rotate freely, but only with a "swarm" of molecules. It is obvious that orientational response times of such "swarms" must be substantially longer than those of the individual molecules. For example, the relaxation time of the optical Kerr effect in MBBA changes with rising temperature from approximately 800 to 40 nanoseconds.

### III. OPTICALLY INDUCED REFRACTIVE INDEX IN THE ISOTROPIC PHASE OF LIQUID CRYSTALS

Since the magnitude of the induced refractive index depends on the increase of the orientational order in the isotropic phase of the liquid crystal, we can use the de Gennes phenomenological model of phase transitions in liquid crystals for its description [11]. The degree of the orientational order is given in this theory by a macroscopic tensorial order parameter  $Q_{\alpha\beta}$ . Any tensorial property of the medium might be used for its definition, but for our purposes the most suitable expression of the order parameter is the one through the susceptibility tensor

$$Q_{\alpha\beta} = \frac{3}{2} \frac{1}{\Delta\chi} \left( \chi_{\alpha\beta} - \frac{1}{3} \sum_{\gamma} \chi_{\gamma\gamma} \delta_{\alpha\beta} \right), \quad (9)$$

where  $\Delta\chi$  is the anisotropy in electric susceptibility for a completely aligned liquid crystal. If this complete alignment is along the  $\hat{x}$  axis, we have

$$\Delta\chi = \chi_{xx} - \frac{1}{2} (\chi_{yy} + \chi_{zz}). \quad (10)$$

It is obvious from Eqs. (9) and (10) that  $Q_{\alpha\beta}$  is a symmetric traceless tensor with  $Q_{xx} = 1$  for complete alignment. The equilibrium value of the order parameter in the isotropic phase is zero. We can re-write Eq. (9) in the form

$$\chi_{\alpha\beta} = \frac{1}{3} \sum_{\gamma} \chi_{\gamma\gamma} \delta_{\alpha\beta} + \frac{2}{3} \Delta\chi Q_{\alpha\beta}. \quad (11)$$

In the de Gennes model the free energy per unit volume is expanded in the vicinity of the phase transition in a Landau type power series as [11]

$$F = F_0 + \frac{1}{2} A(T) Q_{\alpha\beta} Q_{\alpha\beta} + \frac{1}{3} B(T) Q_{\alpha\beta} Q_{\beta\gamma} Q_{\gamma\alpha} - \frac{1}{2} P_{\alpha} E_{\alpha}^*, \quad (12)$$

where  $F_0$  is free energy independent of  $Q_{\alpha\beta}$ ,  $P_{\alpha}$  is the polarization of the medium,  $A(T)$  and  $B(T)$  are temperature dependent coefficients. We have neglected in the above equation terms of higher powers of  $Q_{\alpha\beta}$  and terms describing the spatial variations of  $Q_{\alpha\beta}$ . For a phase transition to occur it is necessary that the coefficient

$A(T)$  go through zero. The simplest form of  $A(T)$  as obtained from the mean field theory is given by

$$A(T) = a(T - T^*), \quad (13)$$

where  $a$  is a constant and  $T^*$  is the transition temperature of a fictitious second-order phase transition. In MBBA  $T^*$  is 0.7 K lower than the phase transition temperature  $T_c$  from the nematic to the isotropic phase [12]. The cubic term in Eq. (12) reflects the symmetry conditions of the phase transition. Due to these conditions, the phase transition is of the first order but with a very small latent heat. Since the expansion (12) is applicable for small  $Q_{\alpha\beta}$  only, and also because of the "weakness" of the transition, we may drop the cubic term in Eq. (12).

Substituting Eq. (1) for polarization into Eq. (12) and neglecting the terms coming from the nonlinear part of the polarization (these terms are many orders of magnitude smaller than the linear term) we obtain

$$F = F_0 + \frac{1}{2} a(T - T^*) Q_{\alpha\beta} Q_{\alpha\beta} - \frac{1}{3} \epsilon_0 \Delta\chi E_{\alpha}^* E_{\beta} Q_{\alpha\beta}. \quad (14)$$

In writing Eq. (14) we have used Eq. (11) for  $\chi_{\alpha\beta}$  and we have included all the terms independent of  $Q_{\alpha\beta}$  into  $F_0$ . The steady state value of  $Q_{\alpha\beta}$  induced by the optical field can then be obtained by the minimization of  $F$  with respect to  $Q_{\alpha\beta}$  and is given by

$$\langle Q_{\alpha\beta} \rangle = \frac{\epsilon_0 \Delta\chi}{3a(T - T^*)} \left( E_{\alpha}^* E_{\beta} - \frac{1}{3} |E|^2 \delta_{\alpha\beta} \right). \quad (15)$$

However, if a short optical pulse is used, the order parameter is not able to respond instantaneously. In this case the transient response of the order parameter is governed by the equation [11]

$$\nu \frac{\partial Q_{\alpha\beta}(t)}{\partial t} = -a(T - T^*) Q_{\alpha\beta}(t) + \frac{1}{3} \epsilon_0 \Delta\chi (E_{\alpha}^*(t) E_{\beta}(t) - \frac{1}{3} |E(t)|^2 \delta_{\alpha\beta}), \quad (16)$$

where  $\nu$  is a viscosity coefficient. The solution of the above equation is

$$Q_{\alpha\beta}(t) = \frac{\epsilon_0 \Delta\chi}{3\nu} \int_{-\infty}^t (E_{\alpha}^*(t') E_{\beta}(t') - \frac{1}{3} |E(t')|^2 \delta_{\alpha\beta}) \exp \left[ -\frac{t' - t}{\tau} \right] dt', \quad (17)$$

where  $\tau$  is the relaxation time given by

$$\tau = \frac{\nu}{a(T - T^*)}. \quad (18)$$

The induced index of refraction for polarized light,  $n = kE$ , can be written with the help of Eqs. (4) and (11) in the form

$$\delta n = \frac{\Delta\chi}{3n_0}(Q_x - Q_y). \quad (19)$$

From Eqs. (19) and (17) we finally obtain for the nonlinear induced refractive index the following equation

$$\delta n(t) = \frac{\epsilon_0(\Delta\chi)^2}{9v n_0} \int_{-\infty}^t |E(t')|^2 \exp\left[\frac{t'-t}{\tau}\right] dt', \quad (20)$$

which gives the contribution of the optical Kerr effect to the induced refractive index. It has been demonstrated that Eqs. (20 and (18) are in very good agreement with the experimental results on the optical Kerr effect in MBBA [4] if the temperature dependence of  $v$  in the isotropic phase is given by [13]

$$v = v_0 \exp [2800/T]. \quad (21)$$

#### IV. SCATTERING OF LIGHT

It is well known that nematic liquid crystals in the isotropic phase display a considerable light scattering caused by thermal fluctuations of the order parameter [14].

The intensity of light that has travelled a distance  $l$  in a scattering medium can be written as

$$I(l) = I_0 \exp [-\alpha l], \quad (22)$$

where  $\alpha$  is the scattering coefficient and  $I_0$  is the intensity of the incident light. We can write the scattering coefficient as

$$\alpha = \int_{\Omega} R \, d\Omega, \quad (23)$$

where  $\Omega$  is the solid angle and  $R$  is the Rayleigh ratio given by [14]

$$R = \frac{1}{I_0} \frac{dI_s}{d\Omega} = \left(\frac{\omega_0}{c}\right)^4 \frac{V}{(4\pi)^2} \langle \partial\chi_{\hat{r}}^2(\mathbf{q}) \rangle, \quad (24)$$

where  $dI_s/d\Omega$  is the intensity of light scattered per unit solid angle,  $c$  is the speed of light in vacuum,  $V$  is the scattering volume and  $\partial\chi_{\hat{r}}(\mathbf{q})$  is the Fourier component of susceptibility fluctuations that couples incident light polarized along  $\hat{r}$  to scattered light polarized along  $\hat{r}$ .

The change in free energy associated with the fluctuations of the order parameter in volume  $V$  is obtained from Eq. (14) as [14]

$$\delta\Phi_0 = \frac{1}{2} \int_V \delta F \, dV = \frac{1}{2} \int_V \left[ a(T - T^*) Q_{\delta\phi} Q_{\delta\phi} - \frac{1}{3} \epsilon_0 \Delta\chi E_{\delta\phi}^* E_{\delta\phi} Q_{\delta\phi} \right] dV, \quad (25)$$

The equilibrium values of all  $Q_{\delta\phi}$  in the isotropic phase are zero and the field dependent term in Eq. (25) vanishes if the orientational action of electric field is neglected. The order parameter fluctuations may be decomposed into statistically independent Fourier components, which gives

$$\delta\Phi_0 = \frac{V}{2} \sum_{\mathbf{q}} \left\{ a(T - T^*) \left[ \frac{3}{2} Q_x^2(\mathbf{q}) + \frac{1}{2} (Q_y(\mathbf{q}) - Q_z(\mathbf{q}))^2 + 2(Q_{xy}^2(\mathbf{q}) + Q_{xz}^2(\mathbf{q}) + Q_{yz}^2(\mathbf{q})) \right] \right\}, \quad (26)$$

where we made use of the tracelessness of  $Q_{\delta\phi}$ .

Applying the equipartition theorem to various terms in Eq. (26) we obtain the fluctuations in the order parameter. We find, for example

$$\langle Q_x^2(\mathbf{q}) \rangle = \frac{2k_B T}{3Va(T - T^*)}, \quad (27)$$

where  $k_B$  is the Boltzmann constant.

The scattering vector  $\mathbf{q}$  is absent in Eq. (27) because we have not taken into account the spatial variations of  $Q_{\delta\phi}$ . Experiments confirmed a negligible influence of these variations on the scattering of light in the isotropic phase of liquid crystals [14].

From the fluctuations of the order parameter we obtain with the help of Eqs. (24, (23) and (11) the scattering coefficient for a linearly polarized light

$$\alpha = \frac{1}{4\pi} \left( \frac{\omega_0}{c} \right)^4 (\Delta\chi)^2 \frac{8k_B T}{27a(T - T^*)}. \quad (28)$$

It is obvious that the scattering of light rapidly increases as the temperature approaches the fictitious second-order transition temperature  $T^*$ . The scattering depletes the power carried by the light beam and thus the electric field strength acting on the molecules is smaller. In all equations describing the molecular reorientation effective field strengths corrected for scattering must therefore be used.

#### V. APPROXIMATE SOLUTION OF THE WAVE EQUATION

The Eq. (8) for the transient case can be solved numerically as in [15] or approximately analytically using the paraxial approximation [9]. Since numerical calculations for very large relaxation times, as is the case of MBBA, are not available, we use the paraxial approximation in order to solve Eq. (8) with the induced refractive index given by Eq. (20). We assume the beam to have a Gaussian transverse profile and look for the solution of the form

$$E_0(r, z, \xi) = \frac{A(z=0, \xi)}{f(z, \xi)} \exp \left[ -\frac{r^2}{2f^2(z, \xi)r_0^2 z=0, \xi)} \right] \exp [-iks(r, z, \xi)], \quad (29)$$

where  $f(z, \xi) = r_0(z, \xi)/r_0(z=0, \xi)$  is the reduced dimensionless radius,  $s(r, z, \xi)$  is the eikonal of the wave and  $\xi = t - z/v_r$  is the local time. The paraxial approximation gives for the reduced radius equation [8, 16]

$$\frac{1}{f} \frac{\partial^2 f(\xi)}{\partial z^2} = \frac{1}{k^2 r_0^4 f^4(\xi)} - \frac{e_0(\Delta\chi)^2}{9n_0^2 r_0^2 v} \int_{-\infty}^{\xi} \frac{A^2(r')}{f^4(r')} \exp \left[ \frac{t' - \xi}{\tau} \right] dr'. \quad (30)$$

The first term in Eq. (30) comes from linear diffraction, while the second term is due to self-focusing. The integral in Eq. (30) builds up gradually with time in transient self-focusing. Therefore the leading part of the pulse suffers diffraction and the beam radius starts shrinking in later parts of the pulse when the integral becomes larger. After a certain propagation distance the reduced radius of the lagging part of the pulse reaches a minimum value and the pulse then travels on without appreciable change over the diffraction length of the leading part of the pulse [18]. If  $f$  varies only slightly with  $\xi$ , we are able to solve Eq. (30) and obtain [16]

$$f^2(z, \xi) = z^2 \left[ \frac{1}{k^2 r_0^4} - \frac{e_0(\Delta\chi)^2 \exp[-az]}{9n_0^2 r_0^2 v} \int_{-\infty}^{\xi} A^2(r') \exp \left[ \frac{t' - \xi}{\tau} \right] dr' \right] + 1, \quad (31)$$

where by  $\exp[-az]$  we have taken into account the scattering losses. We may regard the right-hand side of Eq. (31) as the first two terms of the exponential function expansion and transform Eq. (31) into the form resembling the solution for stable propagation in [15]

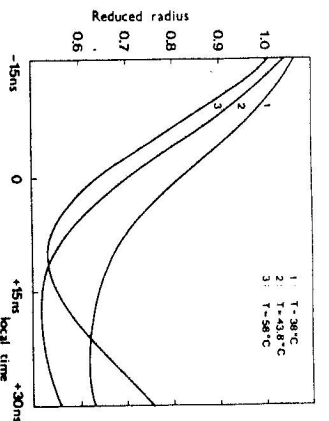
$$f(z, \xi) = \exp \left\{ \frac{z^2}{2} \left[ \frac{1}{k^2 r_0^4} - \frac{e_0(\Delta\chi)^2 \exp[-az]}{9n_0^2 r_0^2 v} \int_{-\infty}^{\xi} A^2(r') \exp \left[ \frac{t' - \xi}{\tau} \right] dr' \right] \right\}. \quad (32)$$

The temporal variation of the reduced radius for a Gaussian input pulse with a pulse-width  $2b$

$$A^2(r) = A_0^2 \exp[-r^2/b^2] \quad (33)$$

as calculated from Eq. (32) is illustrated in Fig. 1. In the calculations we have used  $z = 10$  cm,  $r_0 = 0.12$  mm and the temperature  $T^* = 35.4^\circ\text{C}$ . The material constants have been taken from [3b]. We see that for temperatures in the vicinity of the transition temperature the pulse has a horn shape with the radius of the leading part much larger than the radius of the trailing part. As the temperature is increased, the relaxation time is decreasing and the radius starts to diverge at the

Fig. 1. The reduced radius of the laser beam with input power  $P = 1500$  W after travelling a 10 cm distance in MBBA as a function of local time. The three curves are calculated from Eq. (32) for three different temperatures: 1.  $T = 38^\circ\text{C}$ ; 2.  $T = 43.8^\circ\text{C}$ ; 3.  $T = 58^\circ\text{C}$ .



end of the pulse. Such behaviour is in good agreement with the results of numerical calculations [15] and with experimental observations [5, 18].

The threshold power for self-focusing can be inferred from Eq. (32) in a very straightforward way. The threshold power is defined as that power of the incident beam at which diffraction is compensated by self-focusing, i.e. the radius of the beam does not change. Hence, we have

$$P_n = 9cn_0\lambda^3 v / 4\pi(\Delta\chi)^2 \exp[-az] \int_{-\infty}^{\xi} \exp \left[ -\frac{t'^2}{b^2} + \frac{t' - \xi}{\tau} \right] dt', \quad (34)$$

where  $P_n = \pi r_0^2 n_0 (E_0/\mu_0)^{1/2} A_0^2$ .

Eq. (34) gives extremely low threshold powers of hundredths of watts. The threshold power is decreasing with increasing pulse lengths at constant temperature and the critical power for self-focusing (critical power is the threshold power for pulse lengths much larger than the relaxation time of the medium) is in the region of 50 Watts.

## VI. EXPERIMENTAL ARRANGEMENT

The experimental arrangement is shown in Fig. 2. Our experiments were performed with a ruby laser passively Q-switched by phthalocyanine in a nitrobenzene solution. The pulse-width was approximately 30 nsec. To improve the transverse mode structure a pinhole of a 2 mm diameter was placed inside the cavity of the laser. The power incident on the sample was varied by neutral density filters. In front of the sample-cell there was placed a further pinhole of a 0.24 mm diameter to ensure maximal spatial homogeneity of the incident beam. With this pinhole self-focusing into a single filament was regularly observed. The maximum energy of the beam behind the second pinhole was approximately 0.16 mJ.

The sample used was a nematic liquid crystal *p*-methoxybenzylidene-*p*-n-butylaniline obtained from REACHIM (USSR). The transition temperature from the nematic to the isotropic phase was  $36.1^\circ\text{C}$ . The transition temperature dropped

not this value from 42 °C after a period of heating. This is a well-known effect of "aging" of MBBA, which is known to be a rather unstable compound. A small fraction of MBBA molecules decompose and the impurities thus created cause the transition temperature to fall. As was shown in [17] the optical Kerr effect is not influenced by impurities.

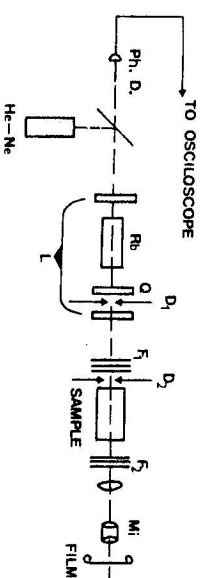


Fig. 2. Experimental arrangement for observing the self-focusing. D1, D2 pinholes with a 2 mm and a 0.24 mm diameter, respectively, F1, F2 Schott neutral filters, L, O, switched ruby laser, Mi, microscope objective, He-Ne helium-neon laser used for optical alignment.

The liquid crystal was placed in a 10 cm long stainless steel cell that was thermally stabilized better than to 0.3 °C.

The beam cross-section was monitored at the exit window of the cell with a microscopic objective and the magnified image of the filament was photographed. The reduced radius was determined at half-maximum from microdensitometer traces that were divided by the radius of the pinhole placed in front of the cell.

## VII. RESULTS AND DISCUSSION

The reduced radius as a function of input power of the laser beam is shown in Fig. 3. When the input power increases, the reduced radius decreases almost exponentially until it reaches a limiting value. The solid curve in Fig. 3 is the theoretical curve calculated from Eq. (32) at 0.0 nsec local time. We see that our description is valid for input powers larger than the threshold power which is approximately 600 W. At 3 kW some nonlinear process probably sets in to limit the beam diameter. Since simulated Brillouin and stimulated Raman scattering appear at appreciably higher input powers [5] and the electric field strength in the filament is not sufficiently large to cause the dielectric breakdown or complete alignment of molecules [18], the two-photon absorption seems to be the most probable limiting mechanism.

We have measured the reduced radius at half-maximum, whereas Eq. (32) gives the reduced radius at 1/e-th of the maximum. Therefore we had to divide all calculated values by a factor 1.2. We had to calculate the reduced radius in the centre of the pulse, i.e. at zero local time or better to say in the vicinity of the zero

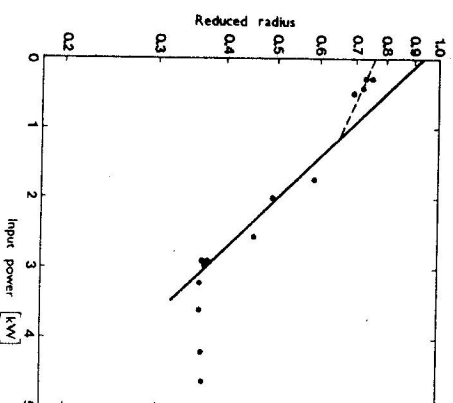
local time. The reason is that photography is an integrating process, thus the largest contribution in the image of the filament comes from regions with the highest intensity, i.e. from the centre of the pulse (for Gaussian input pulses). The centre of the pulse is not always at zero local time, but may, due to self-focusing, shift to positive local times, as illustrated in Fig. 1. This is also the reason why the determination of the threshold power for transient self-focusing by means of photography is not very reliable.

The threshold power is usually determined from photographic measurements as the power of the incident beam at which the radius of the beam after travelling a certain distance in the nonlinear medium starts shrinking. Since in the case of transient self-focusing the threshold power is different for different parts of the pulse, the trailing parts of the pulse start to shrink at substantially lower input powers than the leading parts. The local time at which the reduced radius becomes smaller than 1 depends also on the temperature. These reasons together with the change of the pulse shape cause great difficulties in interpreting the results of the photographic (i.e. time integrating) determination of the threshold powers.

We therefore decided to study the temperature dependence of self-focusing by studying the temperature dependence of the reduced radius at a constant input power rather than by studying the temperature dependence of the threshold power.

In Figs. 4 and 5 we show the reduced radius as a function of temperature for two different input powers. We have chosen the input powers high enough not to fall into the region where our approximation breaks. On the other hand, the input powers had to be low enough not to set into action the limiting mechanism. The experimental data are compared with theoretical curves calculated from Eq. (32) with scattering (full line) and without scattering (broken line). The material constants used were taken from [3b], the length of the sample was 10 cm and the

Fig. 3. The reduced radius as a function of the input power at constant temperature  $T = 43.8$  °C. The solid curve is obtained from Eq. (32) for zero local time.



radius of the incident beam was taken to be 0.12 mm (the radius of the pinhole placed in front of the sample cell). The fictitious second-order transition temperature used in our calculations was 0.7 °C below the transition temperature, i.e. 35.4 °C. All the numerical results had to be divided by a factor 1.201 to transform the radius at  $1/e$ -th of maximum obtained by calculation to the radius at half-maximum obtained from experiments.

We can see from Figs. 4 and 5 that the agreement between experimental points and the theoretical curves with scattering losses taken into account is very good. The theoretical curves shown were calculated for different local times, all in the vicinity of the zero local time. At low temperatures the best fit to the experimental points is given by the curves for positive local times (the lagging part of the pulse). With rising temperature the local time giving the best fit is shifting towards the centre of the pulse (zero local time). This behaviour results from the change of the pulse shape with time, i.e. from the shifting in the local time of the maximum intensity of the pulse. From the pictures of the filaments we find in fact the radius of that part of the pulse which has the largest intensity, especially when the radius is measured at half maximum of the intensity. The shifting of the maximum intensity in local time was found also in time resolved experiments [5, 18]. It is interesting to notice that from our essentially time-integrating experiments we can obtain also some information about the changes of the pulse in time.

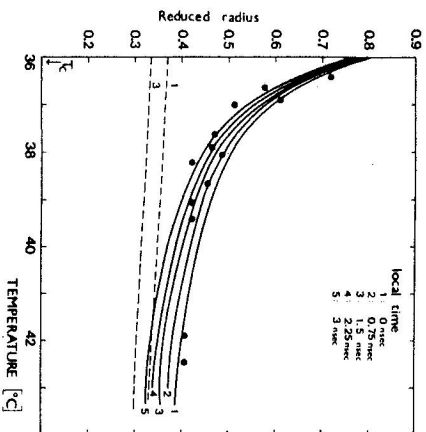


Fig. 4. The reduced radius as a function of temperature at constant input power  $P = 2900$  W. The solid curves are obtained from Eq. (32) with scattering given by Eq. (28) taken into account. The broken curves are obtained from Eq. (32) without scattering. The curves were calculated for different local times.

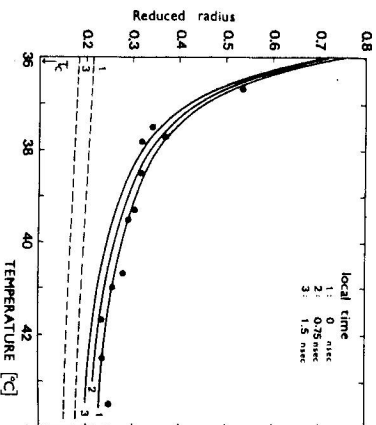


Fig. 5. The reduced radius as a function of temperature at constant input power  $P = 4600$  W. The solid curves are obtained from Eq. (32) with scattering given by Eq. (28). The broken curves are obtained from Eq. (32) without scattering. The curves were calculated for different local times as shown in the figure.

The reduced radius calculated from Eq. (32) without scattering shows very weak dependence on temperature in contradiction with experimental results. At higher temperatures when the scattering coefficient is small the omission of scattering causes only a minor disagreement with the experiment. As the temperature approaches the transition temperature from the isotropic to the nematic phase, the scattering coefficient increases dramatically (see Eq. (28)) and thus the effective field acting on the molecules is weaker. This results in a weaker self-focusing effect at a constant input power. We see that the temperature behaviour of self-focusing in the isotropic phase of MBBA is primarily determined by the temperature dependence of the scattering of light and only moderately influenced by the temperature dependence of the molecular reorientations (the optical Kerr effect).

## VIII. CONCLUSION

In conclusion, we have shown that the temperature behaviour of self-focusing in the isotropic phase of the nematic liquid crystal MBBA is primarily determined by the temperature dependence of the scattering of light. The theoretical description of the transient self-focusing in this compound based on the paraxial approximate solution of the nonlinear wave equation with the induced refractive index obtained from the de Gennes theory is in good agreement with the experimental results. Only for input powers in the vicinity of the threshold power the paraxial approximation breaks down because of the greater role played by the off-axis parts of the beam.

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Received January 3<sup>rd</sup>, 1980