Self-Focusing of TE_{01} and TM_{01} Light Beams: Influence of Longitudinal Field Components

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The self-focusing (SF) of TE_{01} and TM_{01} ruby-laser pulses in CS_2 was studied experimentally and theoretically. The TE_{01} and TM_{01} modes are characterized by strictly transverse circularly electric and magnetic fields, respectively. Rotational symmetry and, in the case of TE_{01}, absence of longitudinal field components allow computational predictions on the self-focusing which are valid even close to a "catastrophic" focus where scalar approximations become problematic. Comparison between the critical powers of TE_{01} and TM_{01} yields information on the induced birefringence selectively from the focal zone. The difference in critical powers depends on the convergence of the light beam incident on the nonlinear medium and disappears for parallel incidence. These theoretical predictions are supported by our experimental findings: giant pulses (~ 70 nsec, ~ 300-kW peak) and mode-locked pulse trains (~ 1 nsec, ≥ 1-MW peak) in the TE_{01} mode were obtained from a Q-switched ruby laser by an appropriate mode selection. These pulses could be easily converted into TM_{01} by rotating the electric field by 90°. Little difference in critical power of the two modes was seen with single-mode pulses; but at 1-nsec duration a distinct difference in threshold developed with increasing convergence. This finding is clear evidence for the influence of longitudinal field components (of TM_{01}) on nonlinear light propagation—a phenomenon which, to our knowledge, has never been observed before. Interestingly, the experimental results are closest to the theoretical values for electrostriction and not to the ones attributed to the optical Kerr effect. Although a restraint in conclusions is necessary in view of the well-known complications of the SF process, it may tentatively be concluded that electrostriction plays an important role in the focal zone even at pulse durations as short as 1 nsec—which in turn provides an estimate for the size of the focus.

TE_{01} and TM_{01} laser modes[1,2] are similar to the waveguide modes with the same designation and the lowest-order Gaussian light beams except for TEM_{00}. Their intensity distributions are rotationally symmetric, ring shaped and identical in isotropic linear media. The electric fields of TE_{01} and TM_{01} point in azimuthal and radial directions, respectively. TE modes are distinguished from all other configurations by the strict transversality of their electric fields.

The following facts motivated our interest in these modes: (i) The electric field of TM_{01}, like most laser modes, consists of both transverse and longitudinal components. Although negligible in collimated beams, it turns out that the latter significantly influence the SF of strongly convergent light beams. Comparison between TE_{01} and TM_{01} allows the investigation of this influence in a simple manner. (ii) The effect of longitudinal field components depends on the nature of the nonlinearity which produces self-focusing (SF), i.e., the induced birefringence (similar to the SF of circularly polarized light[3]). (iii) Most interesting, the difference in SF of the two modes depends mainly on the nonlinearity in the focal zone, the longitudinal components being significant at small diameters only.

Let us begin with a theoretical discussion which will provide the basis for our experimental investigation. The wave equation for the TE_{01} light beam is characterized by the absence of radial and longitudinal electric field components and by rotational symmetry. After splitting the rapidly varying part $e^{i\omega t-ikz}$, with $k=n\omega/c$ (ω, t, k have the usual meaning, and z points in the direction of propagation), the wave equation can be written in the following dimensionless form:

$$i\frac{\partial E_\varphi^*}{\partial z^*}=\frac{\partial^2 E_\varphi^*}{\partial r^{*2}}+\frac{1}{r^*}\frac{\partial E_\varphi^*}{\partial r^*}-\frac{E_\varphi^*}{r^{*2}}+P_\varphi^{\mathrm{NL}*}, \qquad (1)$$

with[4] $r^*=r/w_0$, $z^*=z/4p$, $E_\varphi^*=(\epsilon_2/\epsilon_0)^{1/2}(2/\theta)E_\varphi$, $P_\varphi^{\mathrm{NL}*}=E_\varphi^*|E_\varphi^*|^2$. The spot size w_0, the beam parameter p, and the angle of convergence θ of the corresponding fundamental mode are simply related by $\theta=w_0/p$ and $p=kw_0^2/2$. E_φ^* is the normalized amplitude of the electric field, and ϵ_0, ϵ_2 are the linear and nonlinear dielectric constants, respectively. The usual $|E|^2$ nonlinearity is assumed,[3,4] i.e., saturation is not yet taken into account. The field distribution of TE_{01} in a linear medium is $E_\varphi^*=E_0^*\, r^*\, e^{-r^{*2}}$ at beam waist. Equation (1) is correct in all respects except for the neglected $\partial^2E_\varphi^*/\partial z^{*2}$ term.

The independence of (1) from parameters should be noted. Light entering the nonlinear medium with different convergence self-focuses according to simple coordinate transformations. The minimum power flux of an initially converging light beam which is necessary for the formation of a "catastrophic focus" (= critical power $P_{cr,TE}$), in particular, is a constant in a given material. The value $28.8\lambda^2 c/16\pi^3 n_2$ found by numerical integration is approximately four times larger than P_{cr} of TEM_{00}.[2]

The propagation of a TM_{01} wave is influenced by two components of the nonlinear polarization (P_r^{NL*}, P_z^{NL*}) which are related tensorially to the electric field[3] ($i = r$, $j = z$, and vice versa) by

$$P_i^{NL*} = E_i^*(|E_i^*|^2 + \beta |E_j^*|^2)\,. \qquad (2)$$

The induced birefringence $\beta = \delta n_\perp/\delta n_\parallel$ depends on the nature of the nonlinear processes, and varies between 1 (electrostriction), 0.3 (nonlinear electronic polarizability), and -0.5 (optical Kerr effect, molecular alignment). Note that P_z^{NL*} is eventually larger than E_z^* since $|E_r^*|$ can reach values of up to 10, while $E_z^* \simeq 2.5$ (see below). The TM wave equation is written

$$i\frac{\partial E_r^*}{\partial z^*} = \left(\frac{2i}{\theta} - \frac{\theta}{4}\frac{\partial}{\partial z^*}\right)\frac{\partial E_z^*}{\partial r^*} + P_r^{NL*}\,, \qquad (3a)$$

$$\frac{1}{r^*}\frac{\partial}{\partial r^*}\left[r^*\left(\frac{2i}{\theta} - \frac{\theta}{4}\frac{\partial}{\partial z^*}\right)E_r^* + r^*\frac{\partial E_z^*}{\partial r^*}\right] + (4/\theta^2)E_z^* + P_z^{NL*} = 0. \qquad (3b)$$

Unlike Eq. (1), Eqs. (3) depend on θ and, through Eq. (2), also on β. The two parameters introduce differences in the SF of TE_{01} and TM_{01} which disappear only at $\theta = 0$. In reality the situation is even more involved: There will be gradual variations both in magnitude and anisotropy of the nonlinear index which can be further complicated by transient phenomena and make numerical calculations difficult.

More readily accessible to computation is the steady-state filament, i.e., the solutions of (1) and (3) which satisfy $\partial|E^*_{\varphi,r,z}|/\partial z^* = 0$. It has turned out in investigations on less involved models[5] that the power propagating in such a filament is just equal to the critical power mentioned above. Assuming that the equality holds at least approximately in our situation, we studied the trapped solutions of Eqs. (1) and (3). The results for $\beta = \delta n_\perp/\delta n_\parallel = -0.50$, 0.15 (see below), 1.00, normalized to the constant $P_{cr,TE}$, are plotted versus θ in Fig. 1 (solid curves).[6] The obtained growth of critical power is partially due to the energy stored by the longitudinal components of the electric field. An additional increase is produced by the decrease of the longitudinal polarizability connected with molecular alignment ($\delta n_\perp/\delta n_\parallel = -0.50$). Electrostriction (1.00), on the other hand, favors trapping since it increases the polarizability homogeneously.

In extending the calculation up to $\theta = 0.20$ rad in Fig. 2 we actually considered light beams with a linear spot size w_0 as small as two wavelengths. At these dimensions, saturation should not be neglected which will shift both $P_{cr,TE}$ and $P_{cr,TM}$ to higher values. To account for saturation the previously assumed $|E^2|$ nonlinearity was replaced by an $|E|^2/(1 + |E|^2/E^2_{SAT})$ expression. E_{SAT} was chosen in such a way that the maximum index change was 0.58 (CS_2).[7] There is, however, little known on the saturation in self-focusing light beams (see, example, Ref. 7). Therefore we shaded the areas between the former and the new results assuming that the true ratio of critical powers would be found somewhere in these regions or their close vicinities. From the figure obtained we can still predict an increasing ratio of critical powers if molecular alignment contributes significantly to the SF. A weaker increase followed by a decrease is expected if electrostriction is important in the focal region.

The theoretical results discussed so far were examined experimentally. The light source, a Q-switched ruby laser with an appropriate selection mechanism favoring the TE_{01} mode, was operated either single mode (peak power 300 kW, full half-width $\simeq 70$ nsec) or mode locked (peak power $\simeq 1$ MW, full half-width $\simeq 1$ nsec).[7a] Conversion into the TM_{01} mode was obtained by means of a zero-degree quartz plate which rotates the direction of polarization by 90° but preserves completely the intensity distribution of the incident light [Fig. 2(a)]. The beam passed an optical delay line of up to 35-m length in order to separate the laser pulse and the stimulated Brillouin radiation from the probe.[8] Lenses or complete objectives made the light strongly convergent inside the probe. Convergences of up to $\theta = 0.20$ were obtained without severe distortion of the

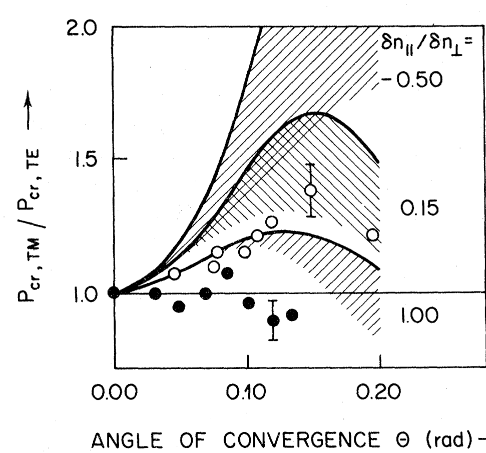

FIG. 1. Theoretical and experimental ratio of critical powers for TM_{01} and TE_{01} vs convergence. Curves: calculated ratio for various values of the induced birefringence. Saturation shifts the theoretical results down into the shaded regions. Experimental results: open circles, 1-nsec pulses; full circles, 70-nsec pulses.

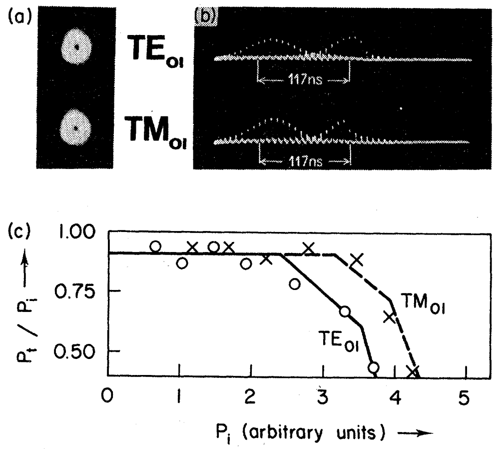

FIG. 2. (a) Far field pattern of the two modes; distance from the laser approximately 4 m. (b) Oscilloscope displays of incident (left) and transmitted (right) light pulses. Note the sharp break in transmission for the TM_{01} pulse. (c) Transmission vs peak power of the spikes displayed in (b). Solid and dashed lines: best fits to five TE_{01} and TM_{01} pulse trains obtained under the same conditions as in (b).

light beam. The probe material, mostly CS_2, was contained in cells of 3- to 10-cm length. The laser pulse entered the liquid in front of the negative confocal position ($z^* < -0.25$, for large θ even $z^* \ll -0.25$) and left it behind the positive one ($z^* > 0.25$ or $\gg 0.25$). We believe, therefore, that the onset of "catastrophic" focusing in this arrangement yields P_{cr}, the magnitude which is approximately equal to the (theoretical) power flux in a filament. SF was inferred from the onset of stimulated Brillouin scattering (long pulses), or the change of divergence at the exit window (short pulses). At small θ the light had to pass a stop behind the cuvette which almost completely blocked the linearly propagated light; a transmitted signal was then direct evidence for SF. At larger θ a measurement of the transmission through a small aperture was more convenient. This method provided useful results since stimulated scattering (Brillouin and Raman) became negligibly small at $\theta > 0.05$ (1-nsec pulses). For still larger convergence, $\theta > 0.10$, SF was usually accompanied or followed by plasma formation. The incident laser pulse and the transmitted light were simultaneously recorded on a Tektronix 519 oscilloscope but delayed with respect to each other. Two typical oscilloscope traces are reproduced in Fig. 2 (b). The series of spikes on the left-hand side represent two incident mode-locked giant pulses; those on the right-hand side are the same pulses after passage through a cuvette with $\theta = 0.15$ and an appropriately chosen aperture. A pronounced decrease in transmission is readily deduced from these oscilloscope traces. Note the different heights of the first spikes which produce SF (on top of the left white markings). Toward the ends of the pulse trains the light paths are almost completely blocked by plasma formation. In Fig. 2(c) the transmission of the two pulse trains is plotted. The circles represent the spikes of the TE_{01} wave and the crosses, those of TM_{01}. The change from linear to nonlinear propagation is clearly seen. The solid and the dashed lines represent the best fits to 5 TE_{01} and 5 TM_{01} pulse trains which all provided data with similar fluctuations as the ones in the graph. The steep lines towards the right-hand side indicate plasma formation. Similar effects were observed in cyclohexane and nitrobenzene. The simplicity of the experimental scheme, in particular of the mode converter, should be emphasized. It allows rapid change from TE_{01} to TM_{01} and vice versa, while all the other experimental conditions are kept constant.

CS_2 was chosen not only because of its large nonlinearity but also because both electrostriction and optical Kerr effect may contribute in this material.[3,9] Molecular alignment (responsible for the Kerr effect) is achieved within a few picoseconds. It will always adjust to its steady-state value in our experiment. Electrostriction, however, associated with the migration of matter, is a slower process: Transient times are of the order of beam diameter/velocity of sound, e.g., 10 nsec and 10μ. In the focal zone, however, the diameter of the beam comes down to a few microns and the material might be able to respond to the electrostrictive force even within 1 nsec. It is an interesting question, therefore, whether electrostriction does in fact contribute and whether we should be able to see this effect in our experiment.

The experimental threshold for SF and TE_{01} light (single mode) was 50 ± 20 kW fairly independent of convergence. This value is approximately four times larger than the one of TEM_{00}, in agreement with theory.[2] The same result was obtained with collimated TM_{01} light ($\theta < 0.02$). The experimental ratios $P_{cr,TM}/P_{cr,TE}$ which can be measured more accurately than absolute values have also been introduced into Fig. 2. Each circle represents the average of 10 to 20 TE_{01} and TM_{01} laser pulses.

As expected, the ratio of critical powers is 1 at $\theta \simeq 0$ for both single-mode and mode-locked pulses. Little effect of the convergence is seen in the investigation with single-mode pulses (solid circles). We should have to introduce an extremely small saturation index change into the previous calculation in order to cover the experimental points by one of the shaded regions. It is interesting to note that the observed diameter of TEM_{00}-type filaments in CS_2 similarly requires the assumption of a rapid saturation.[7] However, in working with strongly convergent light, it may well be that the onset of stimulated Brillouin scattering no longer occurs very close to P_{cr},[9] and the measured deviations from 1 may reflect the influence of the longitudinal field on stimulated scattering rather than on SF.

In order to see the true critical power, we resorted to mode-locked laser operation (open circles). At 1-nsec pulse duration stimulated scattering was weak and disappeared completely at large θ. An obvious increase of $P_{cr,TM}/P_{cr,TE}$ was found this time which depends on θ in the same way as the calculated curves. Most interesting, however, the experimental values are in between the areas ascribed to electrostriction (1.00) and its superposition with the optical Kerr effect (0.15). They are clearly away from the region of the optical Kerr effect alone (−0.50).

Hence, one might tentatively conclude that electrostriction does in fact contribute significantly to the SF in our investigation even at pulse durations as small as 1 nsec. This result would be in good agreement with the focal-zone transient times estimated above. But it should be kept in mind that various details of the SF process have been omitted in our investigation, and more work has to be done in order to clear up completely the role of the mechanisms involved. The mere fact, however, that there is a difference in the SF of TE_{01} and TM_{01} light is clear evidence for the influence of longitudinal field components on the nonlinear propagation of light; a phenomenon which, to our knowledge, has never been observed before.

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Temperature Dependence of the Helium Metastability-Exchange Cross Section in the Range 15–115 °K*

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An optical-pumping experiment has been used to measure the cross sections for the exchange of metastability between He^3 atoms in the $1\,^1S_0$ ground state and the $2\,^3S_1$ metastable state in the temperature range from 15 to 115 °K. A density-matrix treatment of the He^3 system yields a new relationship between observed linewidth and cross section.

I. INTRODUCTION

An optical-pumping technique was used by Colegrove, Schearer, and Walters[1] to measure the temperature dependence of the cross section for exchange of metastability in collisions between He^3 atoms in the $1\,^1S_0$ ground state and the $2\,^3S_1$ metastable state. They found that the exchange rate was a strong function of the temperature and that it could be understood in terms of a long-range repulsive interaction between metastable and ground-state helium atoms in the $^3\Sigma_u^+$ or $^3\Sigma_g^+$ state of the He-He* molecule.

This behavior was first predicted theoretically for the $^3\Sigma_u^+$ state by Buckingham and Dalgarno.[2] Their calculations indicated a binding minimum for an internuclear separation R of about $2a_0$ and a repulsive maximum at $R=4a_0$, where a_0 is the Bohr radius,

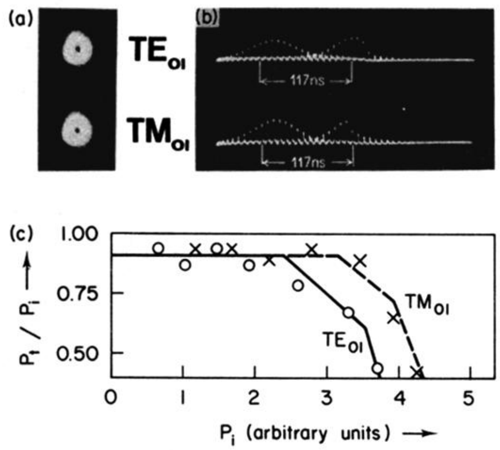

FIG. 2. (a) Far field pattern of the two modes; distance from the laser approximately 4 m. (b) Oscilloscope displays of incident (left) and transmitted (right) light pulses. Note the sharp break in transmission for the TM_{01} pulse. (c) Transmission vs peak power of the spikes displayed in (b). Solid and dashed lines: best fits to five TE_{01} and TM_{01} pulse trains obtained under the same conditions as in (b).