MAX-PLANCK-GESELLSCHAFT
ZUR FÖRDERUNG DER WISSENSCHAFTEN E.V.
PROJEKTGRUPPE FÜR LASERFORSCHUNG
D-8046 GARCHING bei München/Germany

# Wave Front Distortions due to Combined Photolysis and Gasdynamic Effects in a Light Amplifier Operated with $C_3F_7I$ as the Active Medium

# Photolytisch-gasdynamisch ausgelöste Wellenfrontdeformationen in einem mit $C_3F_7I$ als dem aktiven Medium betriebenen Lichtverstärker

K.J. Witte

PLF 2 September 1977

*Diese Arbeit wurde gefördert durch das Bundesministerium für Forschung und Technologie und durch Euratom.*

PLF 2 K.J. Witte

# Wave Front Distortions due to Combined Photolysis and Gasdynamic Effects in a Light Amplifier Operated with $C_3F_7I$ as the Active Medium

September 1977 (in English)


## Abstract

The smooth variation of the refractive index occurring in the central part of an amplifier operated with $C_3F_7I$ as the active medium is theoretically and experimentally investigated. It is shown that the perturbation of the refractive index arises from spatially inhomogeneous pumping and can be explained as a combined photolysis and gasdynamic effect. The wave front distortion of a laser beam resulting therefrom is calculated. Its influence on the focussing properties of the beam is also discussed.

Für einen mit $C_3F_7J$ als dem aktiven Medium betriebenen Lichtverstärker wird die in dem zentralen Bereich auftretende stetige Änderung des Brechungsindex theoretisch und experimentell untersucht. Es konnte nachgewiesen werden, daß sie durch räumlich inhomogenes Pumpen verursacht wird und als ein kombinierter photolytisch-gasdynamischer Effekt angesehen werden kann. Die daraus resultierende Wellenfrontdeformation eines Laserstrahles wird berechnet. Ferner wird ihr Einfluß auf die Fokussierungseigenschaften des Strahles diskutiert.

Contents

## 1. Introduction

It is well known /1/ - /5/ that flash photolysis can produce marked changes of the refractive index in alkyl iodides as $CF_3I$ or $C_3F_7I$. This fact may be important for the pulse propagation in high power iodine laser systems which employ alkyl iodides as the active medium in the amplifiers. Two phenomena have to be distinguished in this context. Firstly there is a shock wave generated at the ignition of the flash and propagating from the walls towards the center with a speed of 1.5 - 2 greater than the speed of sound. It is accompanied by a sharp perturbation of the refractive index. There is no doubt that any interaction of this shock wave with the pulse to be amplified has to be avoided; otherwise a severe distortion of the wave front will result /6/. Secondly, there is a smooth variation of the refractive index typical for the central part of the amplifier tube which has not yet been reached by the shock wave. Up to now this effect has been generally ascribed to the photolysis process /1/, /4/, /5/.
In this paper the nature of this phenomenon will be investigated in more detail. It will be shown that the prerequisite of its occurrence is a spatially inhomogeneous photolysis rate (number of dissociations per unit volume and unit time) leading to a transverse temperature gradient which would not be possible in the case of a spatially homogeneous photolysis rate. If such a temperature gradient and thereby also a pressure gradient exists the gas will be set into motion what then changes the gas density and thus the refractive index. This may result in an intensity independent distortion of the pulse wave front (besides the intensity dependent ones /6/ which will not be discussed here) which in principle could be corrected by a phase correction device (although practically it might not be quite easy to do). The theory given below allows to make an estimate of how much the wave front can be distorted in amplifiers of high power photolysis laser systems.

## 2. Theory

According to the well known Lorenz-Lorentz formula the index of refraction $n_{IR}$ of a gas mixture consisting of $C_3F_7I$ and a buffer gas can be

expressed as follows

$$\frac{n_{IR}^2 - 1}{n_{IR}^2 + 2} = \frac{1}{3\varepsilon_o}\left\{\alpha_{BG} N_{BG} + \alpha_{C_3F_7I} N_{C_3F_7I}\right\} \qquad (1)$$

where $\varepsilon_o$ is the dielectric constant, $\alpha_{BG}$ and $\alpha_{C_3F_7I}$ the wave length dependent polarizabilities of the buffer gas and the $C_3F_7I$-molecules and $N_{BG}$ and $N_{C_3F_7I}$ the corresponding number densities. Because $n_{IR}$ is only slightly different from 1 eq. (1) can be simplified to

$$n_{IR} - 1 \simeq \frac{1}{2\varepsilon_o}\left\{\alpha_{BG} N_{BG} + \alpha_{C_3F_7I} N_{C_3F_7I}\right\}. \qquad (2)$$

For compounds with covalent bondings like $C_3F_7I$ the polarizability is additive, i.e. it can be expressed as a sum of <u>atomic</u> polarizabilities so that eq. (2) becomes

$$n_{IR} - 1 = \frac{1}{2\varepsilon_o}\left\{\alpha_{BG} N_{BG} + \alpha_C N_C + \alpha_F N_F + \alpha_I N_I\right\} \qquad (3)$$

where $\alpha_C$, $\alpha_F$ and $\alpha_I$ are the polarizabilities of the Carbon-, Fluor- and Iodine atoms and $N_C$, $N_F$ and $N_I$ the corresponding number densities given by

$$N_C = 3 N_{C_3F_7I}\ ,\quad N_F = 7 N_{C_3F_7I}\ ,\quad N_I = N_{C_3F_7I}\ .$$

According to eq. (3) the refractive index $n_{IR}$ can vary if either the polarizabilities or the number densities or both quantities are changed. The first case can happen by photolysis, induced emission or chemical reactions among which only the fast recombination ($C_3F_7\cdot + I \longrightarrow C_3F_7I$), the slow recombination ($C_3F_7\cdot + I^{*} \longrightarrow C_3F_7I$) and the dimerization ($C_3F_7\cdot + C_3F_7 \longrightarrow C_6F_{14}$) will be considered; quenching of excited iodine atoms and the formation of molecular iodine can be excluded on the time scale of interest here ($\lesssim$ 100 µsec). The polarizability of the iodine atoms bound to the carbon atoms in the $C_3F_7I$-molecules and those of the free iodine atoms in the excited and in the ground state are not very different from each other. According to our estimates and the measurements of /7/ the difference is less than 3 %. Quantitatively similar results hold for the polarizability

of the carbon- and fluor-atoms in the radical $C_3F_7\cdot$, in the $C_3F_7I$- and in the $C_6F_{14}$ molecules. These differences are small enough to be neglected in the context of the following considerations. This conclusion will be further supported by some experimental data presented in section 3.

The refractive index can then only vary if the various atomic number densities present in the mixture are changed. This is not the case when the pump flux distribution - with the absorption in the laser medium taken into account - is homogeneous over the laser cell cross section although the temperature of the mixture is raised as a consequence of the fact that at each dissociation process an energy of 1.5 eV is set free.[1] The situation is, however, different when a temperature gradient is present because of spatially inhomogeneous pumping.[2] In this case a pressure gradient then also occurs which is made up by two contributions. The first is due to the temperature variation and the second due to a number density variation of the different molecular species of the mixture over the aperture of the cell. A rough estimate shows that the second contribution is at least smaller by a factor of 5 compared with the first one in a mixture without any buffer gas; with buffer gas the second contribution is even less influential depending on the amount of buffer gas so that it will be neglected in the following considerations.

Once a pressure gradient has been established the gas originally at rest begins to move what changes the gas density and also the atomic number density of the various species and thereby the index of refraction $n_{IR}$. In this sense the smooth variation of the refractive index $n_{IR}$ is a combined photolysis and gasdynamic effect.

Under these conditions the quantity $(n_{IR} - 1)$ can be assumed to be proportional to the gas density $\rho$ or equivalently

$$n_{IR} = 1 + (n_{IR,o} - 1)\left(\frac{\rho}{\rho_o}\right) \qquad (4)$$

---

[1] The excitation energy of the $C_3F_7I$-molecules is 4.7 eV. The energy needed to crack the C-I bonding is 2.3 eV and the laser transition energy 0.9 eV. The remaining energy of 1.5 eV is firstly partially stored in the various degrees of freedom of the $C_3F_7$-radical but then quickly thermalized so that 1.5 eV are fully active in heating the gaseous medium.

[2] Diffusion can be neglected because it is too slow in the time scale considered.

where the subscript "0" denotes the state of the gas prior to the photolysis.

A measure of the wave front distortion is the path difference as taken between the center and the edge of the wave front. In units of the wavelength $\lambda$ one obtains for an amplifier of length L

$$\frac{\Delta s}{\lambda} \simeq \frac{L}{\lambda}\,(n_{IR,0}-1)\left\{\left(\frac{\rho}{\rho_0}\right)_{edge} - \left(\frac{\rho}{\rho_0}\right)_{center}\right\}. \qquad (5)$$

Simularly, the additional beam spreading $\Delta\varphi$ caused by the gradient of the refractive index (in the lateral direction) is given by

$$\Delta\varphi = 2L\,(n_{IR,0}-1)\,\mathrm{grad}\left(\frac{\rho}{\rho_0}\right) \qquad (6)$$

where the ray equation of geometrical optics has been used.

We now turn to the question how to determine the space-time behaviour of the gas density. For this purpose we adopt the acoustic approximation /8/ which leads to the following system of coupled diff. eqs. for the gas density $\tilde{\rho}/\rho_0$ , the flow Mach number $\tilde{M}$ and the pressure $\tilde{p}/p_0$ (the superscript "$\sim$" denotes perturbation quantities, for example $\rho = \tilde{\rho} + \rho_0$ with $\tilde{\rho} << \rho_0$ ) in cylindrical coordinates with the assumption of azimuthal symmetry and no variations in axial direction:

Conservation of mass: $$\frac{\partial(\tilde{\rho}/\rho_0)}{\partial\tau} + \frac{1}{\xi}\frac{\partial(\tilde{M}\xi)}{\partial\xi} = 0\,,$$

Conservation of momentum: $$\frac{\partial(\tilde{p}/p_0)}{\partial\xi} + \frac{\gamma}{\xi}\frac{\partial(\tilde{M}\xi)}{\partial\tau} = 0, \qquad (7a,b,c)$$

Conservation of energy: $$\frac{\partial(\tilde{p}/p_0)}{\partial\tau} - \gamma\frac{\partial(\tilde{\rho}/\rho_0)}{\partial\tau} = (\gamma-1)\phi\,\frac{t_c}{p_0}$$ [3]

---

[3] Heat conduction can be neglected in the time scale considered here.

$\xi$ is the dimensionless, independent space coordinate in the radial direction and defined by

$$\xi = r/r_T \tag{8}$$

where $r_T$ is the laser tube radius. $\tau$ is the dimensionless, independent time coordinate and given by

$$\tau = t/t_c \tag{9}$$

where $t_c$ is the time needed by the sound wave to travel the distance from the edge to the center

$$t_c = r_T/a_o. \tag{10}$$

$a_0$ is the speed of sound in the gas at rest and can be calculated using the relation

$$a_o = \sqrt{\gamma\, T_o\, \mathcal{R}/\mathcal{M}} \tag{11}$$

$\gamma$ is the ratio of the specific heats at constant pressure and constant volume of the mixture, $\mathcal{M}$ its average molecular weight, $\mathcal{R}$ the universal gas constant and $T_0$ the gas temperature at rest. With $a_0$ the definition of the Mach number $\tilde{M}$ is

$$\tilde{M} = u/a_o \tag{12}$$

with u being the gas velocity.

Finally, $\emptyset$ is the heat production per unit volume and unit time. $\emptyset$ is dependent on the kind of operation of the laser cell. Two limiting cases will be discussed. The first one refers to the oscillator mode of operation. Here it is assumed that the threshold energy is small compared with the output energy. The excited iodine atoms provided by the photolysis go over to the ground state by induced emission where they quickly recombine with the free radicals to $C_3F_7I$. Because of the low threshold

energy these three processes are almost in equilibrium so that the pump rate P (number of dissociations per unit volume and unit time) is balanced by the recombination rate. Other processes like the dimerization or the recombination of excited iodine atoms with the free radicals to $C_3F_7I$ can be ruled out because their time scales are too slow /9/. As already mentioned an energy $e_P$ = 1.5 eV is released in each dissociation process whereas in each recombination process the C-I bonding energy $e_{C-I}$ = 2.3 eV is gained back so that the heat production $\phi$ is given by

$$\phi_{osc} = P(e_p + e_{C-I}) \qquad 4) \qquad (13)$$

The time dependence of the pump rate P is the same as that of the flash light. Its spatial dependence can be estimated from small signal amplification measurements leading to the expression

$$P = P_0 \tau e^{-a^2\tau^2/2}(1 + b\xi^3). \qquad (14)$$

Here a is the ratio of 2 time scales, namely

$$a = t_c/t_M \qquad \text{(usually} > 1) \qquad (15)$$

where $t_M$ is the time needed by the pump light to reach its maximum value. The parameter b reflects the spatially inhomogeneous pumping. $b = 0$ corresponds to an uniform flux over the aperture of the laser cell what can be realized at very low alkyl iodide pressures. In amplifiers employed in high power laser systems where the stored energy must be as large as possible (the limitation is usually due to internal parasatic oscillations) higher pressures are necessary what then leads to b values of about 1.

The constant $P_0$ occurring in eq. (14) can be related to the energy $E_{osc}$ measured in the oscillator mode of operation because the total number of dissociation processes multiplied by the laser transition energy

---

4) For $CF_3I$ the situation is more complicated and will not be dealt with here.

$e_L$ = 0.9 eV must be equal to $E_{osc}$:[5]

$$E_{osc} = P_o \, e_L \, 2\pi L \int_0^{\infty} \tau \, e^{-a^2\tau^2/2} \, dt \int_0^{r_T} (1+b\xi^3) \, r \, dr \qquad (16)$$

yielding

$$P_o \simeq 0.75 \, \frac{E_{osc}}{e_L} \, \frac{a^2}{V t_c} \qquad (17)$$

where $V = \pi \, r_T^2 \, L$ is the laser cell volume.

We now investigate the second limiting case which refers to the amplifier mode of operation where it will be assumed that the threshold is so high that no lasing occurs. All the energy is then stored in the excited iodine atoms. Their deexcitation by the parent molecules or by the buffer gas will be neglected in the time scale of interest here ( $\lesssim$ 100 µsec) what limits the $C_3F_7I$-pressure to values $\lesssim$ 100 Torr. Under these conditions the following processes have to be considered: photolysis, slow recombination ($C_3F_7 + I^* \longrightarrow C_3F_7I$) and the dimerization ($2C_3F_7 \rightarrow C_6F_{14}$). In the slow recombination process the C-I bonding energy $e_{C-I}$ and the laser transition energy $e_L$ is gained back, whereas in the dimerization process the C-C bonding energy $e_{C-C}$ = 4.1 eV is set free. The heat production then reads

$$\phi_{Amp} = P e_p + (e_{C-I} + e_L) \, k_R^* \, N_R \, N_I^* + e_{C-C} \, k_D \, N_R^2 \qquad (18)$$

where $N_R$ is the number density of the free $C_3F_7\cdot$ radicals, $N_I^*$ the number density of the excited iodine atoms, $k_R^*$ and $k_D$ the rate constants of the slow recombination- and the dimerization process. $N_I^*$ and $N_R$ obey the diff. eqs.

---

[5] This assumption requires a minimization of the influence of the shock wave; otherwise it might happen that the laser radiation is scattered by the shock wave in such a way that it misses the calorimeter. This can be avoided by using as little buffer gas as possible (just enough not to get pyrolysis) and a resonator where the partially transmitting mirror has a low reflectivity ( $\sim$ 4 %).

$$\frac{\partial N_{I^*}}{\partial t} = P - k_R^* N_{I^*} N_R \,, \qquad \frac{\partial N_R}{\partial t} = P - k_R^* N_{I^*} N_R - k_D N_R^2 \tag{19a,b}$$

which - unfortunately - cannot be solved analytically. But in view of the fact that neither $k_R^*$ nor $k_D$ are known very accurately - the uncertainty is at least a factor of 3 according to the values given in the literature /10/ - we linearize the eqs. (19a,b) by assigning a constant value to the characteristic time scale $t_{I^*} = 1/\hat{N}_R\, k_R^*$ of the recombination process as well as to the characteristic time scale $t_D = 1/\hat{N}_{J^*}\, k_D$ of the dimerization process. $\hat{N}_{J^*}$ and $\hat{N}_R$ are average values assumed to be one half of the value attained by $N_{J^*}$ and $N_R$ if only the pumping process would be present:

$$\hat{N}_{J^*} = \hat{N}_R = \int_0^\infty \int_0^1 P\, dt\, \xi\, d\xi = \frac{1}{2} P_o t_c / a^2 = \frac{3}{8} E_{osc} / V e_L \tag{20}$$

The diff. eqs. (19a,b) can now be written

$$\frac{\partial N_{I^*}}{\partial t} \simeq P - \frac{N_{I^*}}{t_{I^*}} \,, \qquad \frac{\partial N_R}{\partial t} \simeq P - \frac{N_R}{t_{I^*}} - \frac{N_R}{t_D} = P - \frac{N_R}{t_D^*}$$

yielding the solution

$$N_{I^*} = \frac{P_o t_c}{a^2} \left(1 + b\xi^3\right) G\left(\tau, a, g_{I^*} = t_c / t_{I^*}\right), \qquad N_R = \frac{P_o t_c}{a^2} \left(1 + b\xi^3\right) G\left(\tau, a, g_D = t_c / t_D^*\right) \tag{21a,b}$$

with

$$G(\tau, a, g) = e^{-\tau g - a^2\tau^2/2} - e^{-\tau g + g^2/2a^2} + \frac{g}{a}\sqrt{\frac{\pi}{2}}\, e^{-\tau g + g^2/2a^2} \left\{\phi_W\left(\frac{g}{a}\right) - \phi_W\left(\frac{g}{a} - a\tau\right)\right\} \tag{22}$$

where $\phi_W$ is the probability integral. Using the same reasoning as above we can approximate the heat production $\phi_{Amp}$ according to eq. (18) by the

expression

$$\phi_{Amp} \simeq P \ell_p + (\ell_{c-I} + \ell_L) \frac{N_{I^*}}{t_{I^*}} + \ell_{c-c} \frac{N_R}{t_D}$$

or

$$\phi_{Amp} \simeq P_o (1 + b\xi^3) \left\{ \ell_p \ell^{-a^2\tau^2/2} \cdot \tau + \frac{\ell_{c-I} + \ell_L}{a^2} \frac{t_c}{t_{I^*}} G(\tau, a, g_{I^*}) + \frac{\ell_{c-c}}{a^2} \frac{t_c}{t_D} G(\tau, a, g_D) \right\} . \qquad (23)$$

We now continue with the integration of the diff. eqs. system (7a,b,c). This will be done by using the method of characteristics (the eqs. 7a,b,c are of the hyperbolic type). Three types of characteristics $C_1$,$C_2$ and $C_3$ with the three different slopes $\mu_{1,2,3} = \frac{d\xi}{d\tau} = \pm 1,0$ (see Fig. 1) can be defined along which the following relations hold:

along $C_1$ ($\mu_1 = +1$):
$$d(\tilde{p}/p_o) + \frac{\gamma}{\xi} d(\tilde{M}\xi) = (\gamma - 1) \frac{t_c}{p_o} \phi \, d\tau ,$$

along $C_2$ ($\mu_2 = -1$):
$$d(\tilde{p}/p_o) - \frac{\gamma}{\xi} d(\tilde{M}\xi) = (\gamma - 1) \frac{t_c}{p_o} \phi \, d\tau , \qquad (24a,b,c)$$

along $C_3$ ($\mu_3 = 0$):
$$\gamma \left(\frac{\tilde{\rho}}{\rho_o}\right) = \left(\frac{\tilde{p}}{p_o}\right) - (\gamma - 1) \int_0^\tau \frac{t_c}{p_o} \phi \, d\varepsilon .$$

The initial conditions are

$$\left.\begin{array}{l} \tau = 0; \\ 0 \le \xi \le 1 \end{array}\right\} \quad \frac{\tilde{\rho}}{\rho_o} = \frac{\tilde{p}}{p_o} = \tilde{M} = 0,$$

and the boundary conditions are

$$\tilde{M}(\tau, \xi = 0) = \tilde{M}(\tau, \xi = 1) = 0 .$$

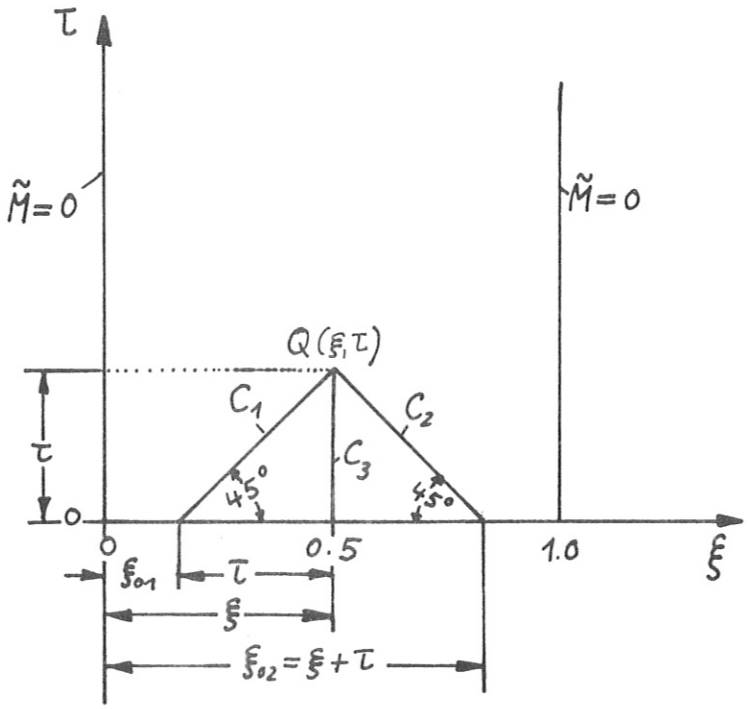



Fig.1: Characteristics, initial and boundary conditions in the $\xi, \tau$-plane

We are mainly interested in the state of the gas mixture at the end of the pumping process when the light pulse to be amplified propagates through the amplifier or the laser emission is completed. This situation usually corresponds to a $\tau$ value of about or less than two or three times $t_M/t_c = 1/a$ which is usually much smaller than unity. This simplifies the integration procedure considerably because the boundary conditions at $\xi = 1$ have not to be taken into account.

An analytical integration of the diff. eqs. (24a,b) is not possible without any simplifying assumption. Along $C_1$ ($\tau = \xi - \xi_{01}$) one obtains

$$\frac{\tilde{p}}{p_0} + \gamma \int_{t_1} \frac{d(\tilde{M}\xi)}{\xi} = (\gamma-1)\,\frac{\phi_{0,osc}}{p_0/t_c} \int_0^{\tau} \varepsilon\, e^{-a^2\varepsilon^2/2}\,(1+b\xi^3)\,d\varepsilon$$

Because it is not known how $\tilde{M}$ varies with $\xi$ along $C_1$ we approximately solve the integral involving $\tilde{M}$ by assigning to $\xi$ an average value in the integration interval which is chosen to be $(\xi - \tau/2)$. A similar consideration can be applied to the other integral.

The solution of the diff. eqs. (24a,b,c) is then

$$\underline{\text{along } C_1}: \quad \frac{\tilde{p}}{p_0} + \gamma \tilde{M} \frac{\xi}{\xi - \tau/2} = \frac{\gamma-1}{a^2} \frac{P_0 \ell_P}{p_0/t_c} \left(1 + b\left[\xi - \tau/2\right]^3\right) H(\tau),$$

$$\underline{\text{along } C_2}: \quad \frac{\tilde{p}}{p_0} - \gamma \tilde{M} \frac{\xi}{\xi + \tau/2} = \frac{\gamma-1}{a^2} \frac{P_0 \ell_P}{p_0/t_c} \left(1 + b\left[\xi + \tau/2\right]^3\right) H(\tau), \quad (25a,b,c)$$

$$\underline{\text{along } C_3}: \quad \gamma \frac{\tilde{\rho}}{\rho_0} = \frac{\tilde{p}}{p_0} - \frac{\gamma-1}{a^2} \frac{P_0 \ell_P}{p_0/t_c} \left(1 + b\xi^3\right) H(\tau).$$

The function $H(\tau)$ is dependent on the mode of operation. It reads

$$H(\tau) = \begin{cases} (1 - e^{-a^2\tau^2/2})(1 + \ell_{c-I}/\ell_P) & \text{for the oscillator mode of operation} \\ (1 - e^{-a^2\tau^2/2}) + \dfrac{\ell_{c-I} + \ell_L}{\ell_P} \dfrac{t_c}{t_{I^*}} \displaystyle\int_0^\tau G(\varepsilon, a, g_{I^*})\, d\varepsilon & \text{for the amplifier mode of operation} \\ \quad + \dfrac{\ell_{c-c}}{\ell_P} \dfrac{t_c}{t_D} \displaystyle\int_0^\tau G(\varepsilon, a, g_D)\, d\varepsilon \end{cases} \quad (26a,b)$$

For $g_{I^*}, g_D \ll a$ eq. (26b) can be simpliefied to

$$H(\tau) = 1 - e^{-a^2\tau^2/2} + 2.13\left\{1 - e^{-\tau g_{I^*}} - \frac{g_{I^*}}{a}\phi_W(a\tau)\right\} + 2.73\frac{t_D^*}{t_D}\left\{1 - e^{-\tau g_D} - \frac{g_D}{a}\phi_W(a\tau)\right\}$$

It can be clearly seen how the heat production due to dissociation is enhanced by either the fast recombination or by the slow recombination and the dimerization. For the latter case the values of the two rate constants $k_R^*$ and $k_D$ are essential.

With these formulas the density $\tilde{\rho}/\rho_0$ can be calculated at any point $\xi, \tau$ as long as the condition

$$\tau \leq \xi \leq (1 - \tau) \quad (27)$$

is fulfilled what means that points with $0 < \xi < \tau$ are excluded.

For points satisfying relation (27) we first combine eqs. (25a,b) to calculate the pressure rise $\tilde{p}/p_0$ and then by inserting this result in eq. (25c) we obtain for the density perturbation:

$$\left(\frac{\tilde{\rho}}{\rho_0}\right)(\xi,\tau) = 1.9\, b\, \frac{\gamma-1}{\gamma}\, \frac{E_{osc}}{V p_0}\, \xi\tau^2 \left\{1+\frac{1}{6}\left(\frac{\tau}{2\xi}\right)^2\right\} H(\tau). \qquad (28a,b)$$

For points on the axis only eqs. (25b,c) are needed. But the approximation employed for the integral $\int d(\tilde{M}\xi)/\xi$ in eq. (25b) would yield zero here what is too far off the real value. A better way of doing the integration in this case is the following one:

$$\int_{\xi_{02}}^{0} \frac{d(\tilde{M}\xi)}{\xi} = \int_{\tilde{M}(\xi=\xi_{02})}^{\tilde{M}(\xi=0)} d\tilde{M} + \int_{\xi_{02}}^{0} \frac{\tilde{M}}{\xi}\, d\xi = \int_{\xi_{02}}^{0} \frac{\tilde{M}}{\xi}\, d\xi \simeq -\left(\frac{\tilde{M}}{\xi}\right)_{av} \tau$$

Use has been made of the boundary conditions $\tilde{M}(\xi = 0) = \tilde{M}(\tau = 0) = 0$ and of the requirement $\xi_{02} = \tau$. The average value of the quantity $\tilde{M}/\xi$ will be set equal to its value at $\xi = \xi_{02}/2 = \tau/2$ which can be calculated combining eqs. (25a,b). The result is

$$\int_{\xi_{02}}^{0} \frac{d(\tilde{M}\xi)}{\xi} \simeq -\left(\frac{\tilde{M}}{\xi}\right)_{av} \tau = \frac{9}{32}\, \frac{b}{a^2}\, \frac{\gamma-1}{\gamma}\, \frac{p_0 \varrho_p}{p_0/t_c}\, \tau^3 H(\tau/2).$$

The density perturbation on this axis then reads

$$\left(\frac{\tilde{\rho}}{\rho_0}\right)(\xi=0,\tau) = 0.16\, b\, \frac{\gamma-1}{\gamma}\, \frac{E_{osc}}{V p_0}\, \tau^3 \left\{H(\tau) + 2.44\, H(\tau/2)\right\}. \qquad (29a,b)$$

A comparison of the eqs. (29) and (28) shows that the density perturbation on the axis is smaller than that in the outer parts of the cell what is to be expected because the pressure gradient gets weaker in the neighbourhood of the axis.

Another question concerns the difference of the density perturbation in the oscillator and amplifier mode of operation under otherwise equal conditions. It can perhaps be used to estimate the order of magnitude of the two rate constants $k_R^*$ and $k_D$.

## 3. Comparison with Experimental Results

Strong experimental evidence can be given for the explanation of the smooth variation of the refractive index as outlined above. Figs. 2, 3 and 4 show the interference pattern of a laser cell equipped with 6 flashlamps which are directly placed in the medium to be irridiated. The laser cell was put in one arm of a Mach Zehnder interferometer which was illuminated by an Argon ion laser ($\lambda$ = 515 nm[6]). The interference pattern was recorded in time by a streak camera both in the framing and the streaking mode. In Fig. 2 the cell is only filled with Argon.

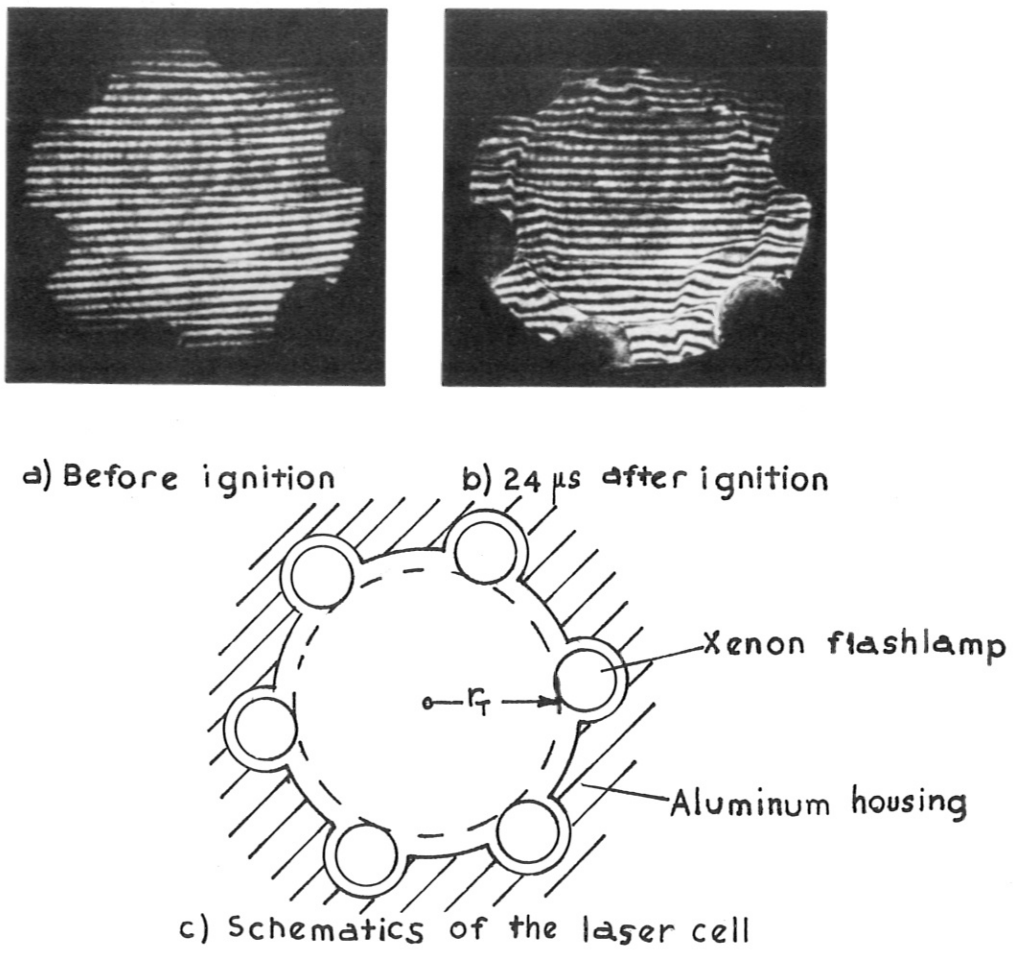



Fig.2: Interference pattern in pure Argon (760 Torr)

---

[6] The fact that the probing laser light falls inside a strong absorption band of $I_2$ is not harmful to the measurements because - as already stated - the formation of molecular iodine is much too slow a process to be of any importance here. Only if the same mixture is flashed several times care has to be taken.

The picture on the left (a) is taken prior to the photolysis, the one on the right (b) 24 µsec after the start of the flash. Except for the area already covered by the shock wave the interference pattern remains unchanged as it is to be expected because there is no light absorption in pure Argon.

The situation is, however, different when $C_3F_7I$ is present as it is demonstrated in Fig. 3. Picture a) is again taken before the ignition of the

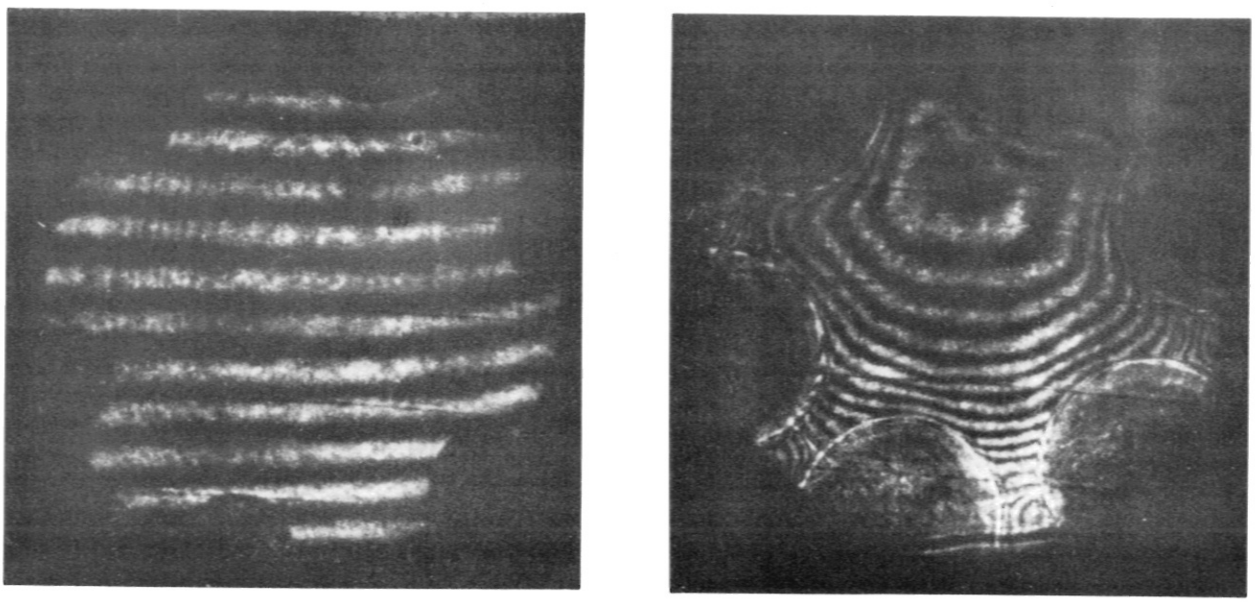



Fig.3: Interference pattern in a mixture of 30 T $C_3F_7I$ + 720 T Argon (Amplifier mode of operation)
Data of the laser cell: $r_T$ = 2.5 cm; L = 40 cm; $t_M$ = 3.7 µs
b = 1; $E_{osc}$ = 10 J (within $r_T$; see Fig. 2c)

flash, pictures b) 24 µs after the start of the flash. Now the fringes are curved because of the inhomogeneous heating of the gaseous medium. The interference fringes are shifted upwards what corresponds to an increase in the gas density. This can be concluded from Fig. 2b where across the shock wave which is accompanied by a positive density jump the interference fringes are also moved upwards. This behaviour is predicted by the theory (see eqs. (28) and (29)).

Fig. 4 is a streak photograph obtained by scanning the slit at a streak velocity of 10 µs/cm. In this case pure $C_3F_7I$ at a pressure of 40 T was used.

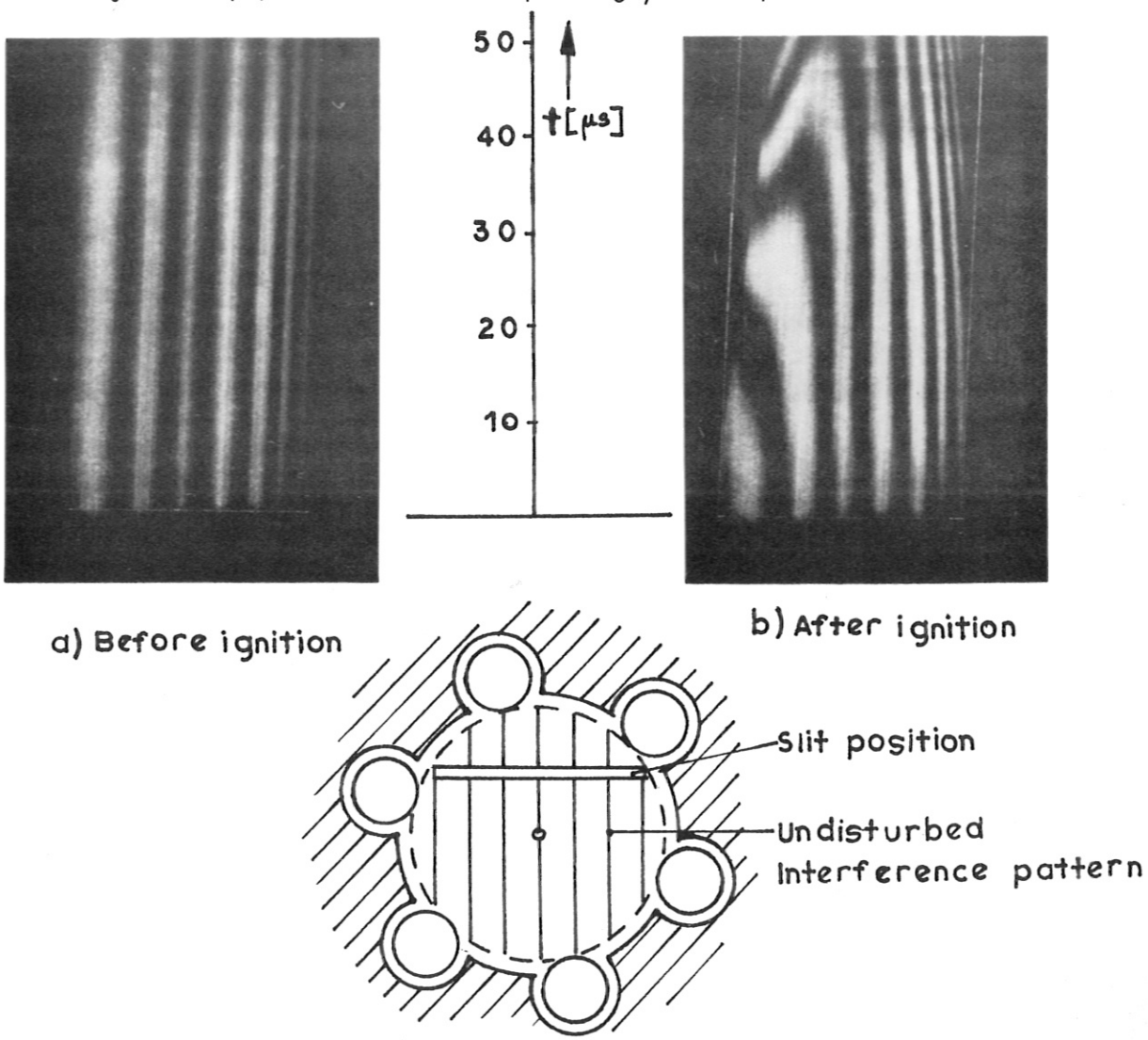



Fig.4: Interference pattern in pure $C_3F_7I$ at 40 T
(Amplifier mode of operation, $E_{osc}$ = 12 J within $r_T$ ; b = 1)

In both cases (Fig. 3 and 4) the laser cell was operated as an amplifier. Whereas in the case of Fig. 3 the threshold was high enough to avoid any lasing at all in the case of Fig. 4 lasing was observed after 8 µs so that thereafter the situation is somewhere between the two limiting cases discussed so far, but still such that the displacement of the fringes should follow closely a time square dependance as predicted by eq. (28a) for $\tau > 2/a$ (lasing period)

$$\frac{s}{\lambda} = \frac{L}{\lambda}\left(n_{IR,o} - 1\right)\left(\frac{\tilde{P}}{P_o}\right) \sim \xi\tau^2 \quad \left(\tau \leq \xi \leq 1-\tau\right) \qquad (30)$$

what is confirmed by the experiment (Fig.4b).

In order to check the accuracy of the theory we calculate the displacement of the fringes and compare it with the experiment. For this purpose we choose in Fig.3b the fringe going through the center. For t = 24 µs and $\xi$ = 0.6 we arrive - according to the eqs. (5), (28b) and (29b) - at a value of $\Delta s/\lambda = 1.0$ which is in reasonable agreement with the experiment. ($t_c$ = 93 µs; $\gamma$ = 1.49; $a_o$ = 268 m/s; a = 25; $\tau$ = 0.26; $k_R^*$ = 1 · $10^{-13}$ $cm^3/s$; $k_D$ = 1.5 · $10^{-13}$ $cm^3/s$; $(n_{IR,o} - 1) = 3.7 \cdot 10^{-4}$). With regard to Fig.4b we choose the 6th fringe from the left corresponding to $\xi$ = 0.7 and calculate the displacement of this fringe at t = 50 µs both for the oscillator- and amplifier mode of operation. According to the eqs. (28a,b) and (30) one obtains $(s/\lambda)_{osc}$ = 2.8 and $(s/\lambda)_{Amp}$ = 2.3 ( $\gamma$ = 1.065, $a_o$ = 94 m/s; $t_c$ = 266 µs, a = 72; $\tau$ = 0.19; $k_R^*$ = 1 · $10^{-13}$ $cm^3/s$; $k_D$ = 1.5 x $10^{-13}$ $cm^3/s$; $(n_{IR,o} - 1) = 1.72 \cdot 10^{-4}$). The experimental value is 2.5 and lies between the two theoretical values. Thus, in both cases considered we observe good agreement between theory and experiment. Although this is a nice result one should not think too highly of it because of the approximations made in the theory (azimuthal symmetry, integration procedure) and the uncertainty regarding the value of some parameters which have to be determined experimentally like b, $E_{osc}$ and the rate constants $k_R^*$ and $k_D$. But nevertheless, an accuracy within a factor of 1.5 - 2 seems very likely.

At the beginning of the second paragraph (theory) the assumption was made that changes of the polarizability of the various atoms caused by photolysis, induced emission or chemical reactions can be neglected. Now experimental

evidence can be given for this hypothesis. In the example of Fig.4 lasing started after 8 µs. The light was emitted in a series of spikes (checked by a diode) the first one being by far the strongest and shortest one ($\sim$ 100 ns). This dump spike almost instaneously produced a considerable concentration of iodine atoms in the ground state. If it would have been followed by a drastic change of the polarizability of the iodine atoms involved a discontinuity in the course of the interference fringes should have resulted after 8 µs. But no such thing could be detected in Fig. 4b. Furthermore, immediately after the induced emission the fast recombination process sets in ($t_R = 1/n_R\, k_{Rec} \lesssim 1$ µs; $k_R = 1 \cdot 10^{-11}$ cm$^3$/s) what alters the polarizability of the free radicals and the ground state iodine atoms which are now bound. But again, this change is so small that no corresponding bend of the fringes could be observed. These considerations give therefore strong support to our explanation of the fringe shift according to which it is basically a combined photolysis and gasdynamic effect.

We will conclude this paragraph by comparing the theory presented with some experimental data of reference /4/ where a similar set up was used as the one described here with the laser cell operated as an oscillator so that the eqs. (28a) and (29a) must be employed. Fig.5 gives the displacement of the interference fringes at the center of the cell ($\xi = 0$) plotted versus time.

Because there is no information in /4/ regarding the oscillator energy $E_{osc}$ the points of maximum displacement in the curves a) and b) were chosen to determine $E_{osc}$. In case a) one arrives at a value of 41 J which would correspond to an overall efficiency of 0.5 % which is not unreasonable. To fit the data of case b) $E_{osc}$ = 22 J had to be assumed anything else left unchanged. This decrease is larger than the corresponding one of the electrical input energy from 8 to 6 kJ. A reason for this discrepancy could be a change in the lamp characteristic: perhaps a slight enhancement of the time $t_M$ after which the pump light reaches its maximum. Curve c) has been calculated with $E_{osc}$ as in case b). It can be seen that the time behaviour of the experimental data is quite well reproduced by the theory presented which - considering the uncertainty of the fitting procedure - should be accurate within a factor of 2.

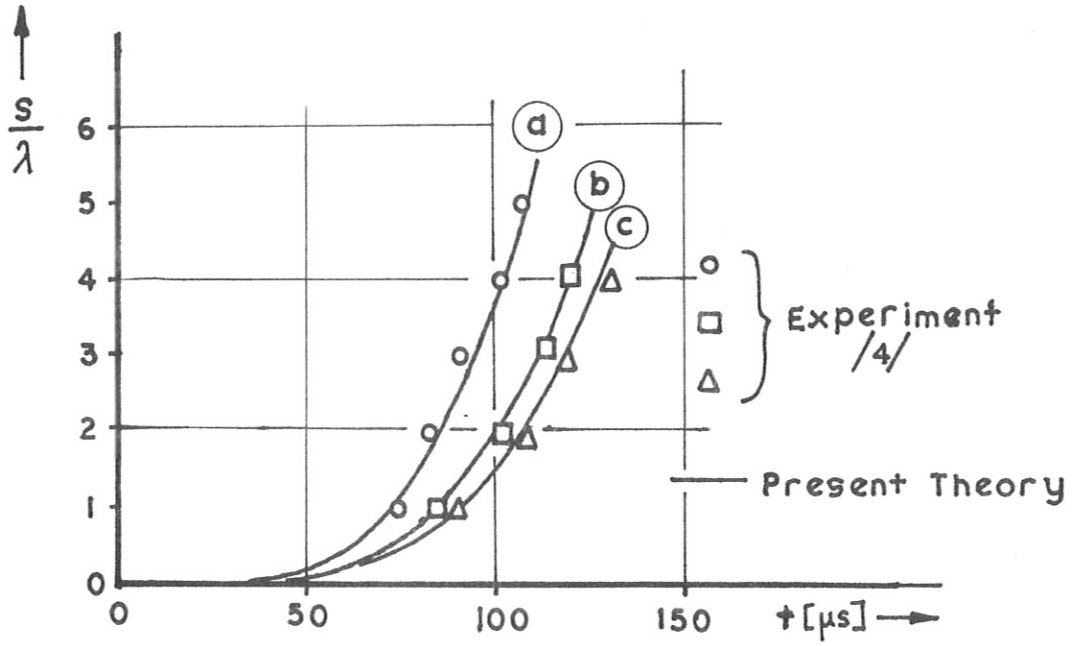
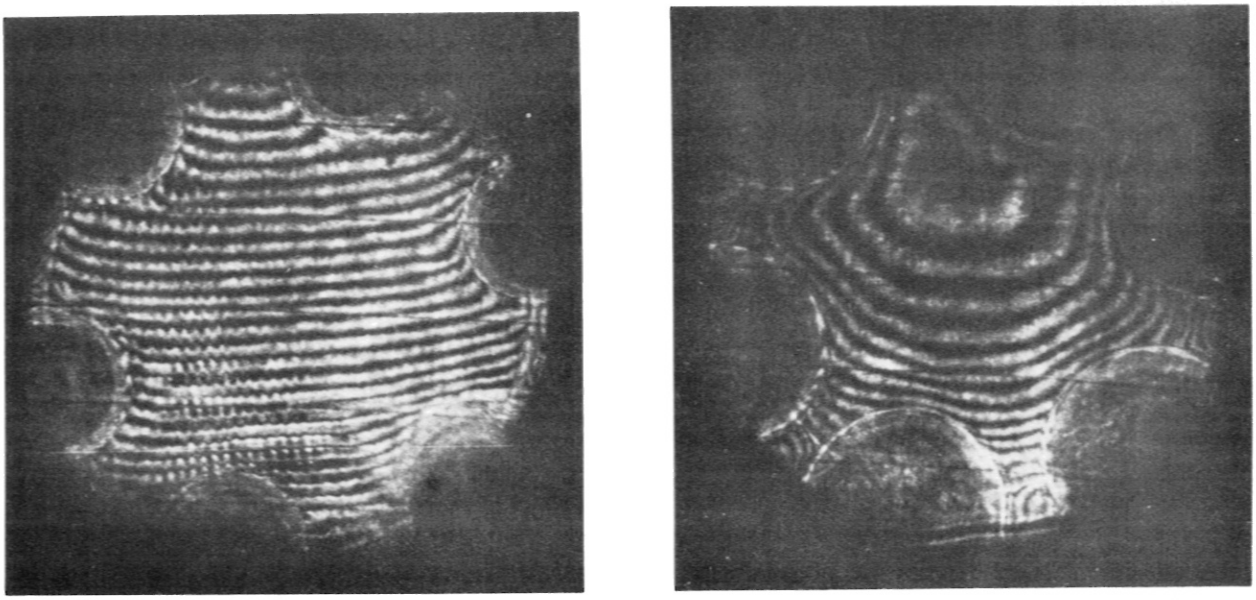



Fig.5: Displacement of the central interference fringe ( $\xi = 0$) according to ref. /4/
(L = 28 cm; $r_T$ = 2.25 cm; $t_M$ = 60 μs; b = 1; $\lambda = 0.633 \cdot 10^{-4}$ cm)
a) 30 T $C_3F_7I$ + 400 T $SF_6$: $\gamma$ = 1.09; $a_o$ = 130 $\frac{m}{s}$; $t_c$ = 173 μs; a = 2.9; $(n_{IR,o} - 1) = 5.3 \cdot 10^{-4}$; $E_{el}$ = 8 kJ
b) Data as in a), but $E_{el}$ = 6 kJ
c) Pure $C_3F_7I$ at 30 Torr: $\gamma$ = 1.060; $a_o$ = 94 m/s; $t_c$ = 239 μs; a = 4; $(n_{IR,o} - 1) = 1.3 \cdot 10^{-4}$; $E_{el}$ = 6 kJ

Another important result - equally well confirmed by the theory presented - is the fact that the addition of $SF_6$ does not help to damp the perturbation of the refractive index. Two effects counteract each other. By addition of a buffer gas the temperature gradient is reduced on the one hand, but the index of refraction is increased on the other hand. In case of $SF_6$ the net effect is almost zero. This conclusion does not hold for Argon because of its very low heat capacity compared to that of $SF_6$: $c_{v,A} / c_{v,SF_6} = 1/8$, but $(n_{IR,o} - 1)_{Argon} / (n_{IR,o} - 1)_{SF_6} = 1/3$. Thus with Argon added to the alkyliodide the perturbations of the refractive index are even more pronounced as can be seen from Fig.6 which shows the interference pattern in pure $C_3F_7I$ at 30 T (picture 6a) and with 720 T Argon added (picture 6b ; it is the same as in Fig.3b) 24 μs after the start of the flash light. If we again choose the fringe going through the center and take $\xi = 0.6$ we then obtain from the theory for the pure $C_3F_7I$ case $\Delta s/\lambda = 0.2$ and for the other case $\Delta s/\lambda = 1.0$, as already given. These values are confirmed by the experiment.



a) 30 Torr $C_3F_7I$ b) 30 Torr $C_3F_7I$ + 720 Torr Argon

Fig.6: Interference pattern in pure $C_3F_7I$ at 30 T and in a mixture of 30 T $C_3F_7I$ + 720 T Argon 24 µsec after ignition

## 4. Concluding Remarks

The smooth variation of the refractive index occurring in the central part of an amplifier operated with $C_3F_7I$ as the active medium was theoretically and experimentally investigated. It could be shown that the perturbation of the refractive index is due to spatially inhomogeneous pumping and can be explained as a combined photolysis and gasdynamic effect. When the pump light is not homogeneously absorbed a temperature gradient and thereby also a pressure gradient arises which sets the gas into motion what then changes the gas density and simultaneously the index of refraction. Other effects like changes of the polarizability of the various atomic species present in the mixture due to photolysis, induced emission or chemical reactions are smaller by at least one order of magnitude. It could be further shown that the

theory presented can reproduce the available experimental data in a satisfactory manner so that it can be used with confidence to predict the wave front distortion in high power photodissociation laser systems.

As an example we investigated the beam aberration in the Terawatt laser system ASTERIX III /11/ which consists of one oscillator and four amplifiers. At the exit of the last amplifier we arrived at a phase variation of 1 up to 1.5 waves across the aperture for typical operational conditions. The distortion is assumed to be circularly symmetric which is reasonable in view of the many flashlamps displaced against each other. Its effect is similar to what is known from glass lasers as "whole beam self-focussing" except that there the phase is retarded on the axis with regard to the edge - vice versa to our situation - and the distortion is intensity dependent, whereas here it is intensity- or time independent and can therefore more easily be corrected. The properties of interest are the intensity profile and the phase of the beam in transverse planes located in the region between the focal plane of the lens and planes several hundred Rayleigh lengths on either side of the focus. The phase distortion moves the best focal plane away from the lens and increases the depth of the focus as well as the beam size of the distorted beam relative to the undistorted beam. An excact quantitative analysis of these effects is beyond the scope of this paper. But what one has to expect can be approximately estimated from the results of /12/ where the focussing properties of an aberrated laser beam from a solid state laser were investigated having a nonlinear whole beam phase distortion $B = 0.75\lambda$. Applied to our situation a phase distortion of one wave would shift the best focal plane away from the lens a distance given by $\sim 20\,\lambda\,(F\#)^2$ where $F\# = f/D$ is the F-number of the focussing lens. The best focal plane is defined as the plane in which the circle containing 90 % of the total energy has the smallest radius. The radius of this circle is for the aberrated beam twice as big as for the diffraction limited beam. More pronounced are the differences of the transverse intensity profiles in planes far removed from the best focus (about  hundred Rayleigh lengths). Alltogether one can say that the focussing properties are not changed very drastically by a phase distortion of one wave and can still be tolerated.[7)]

---

[7)] In case of ASTERIX III they could be even reduced by switching to $SF_6$ as the buffer gas instead of Argon.

For large sized amplifiers using pumping times of 100 µs ore more (in ASTERIX III the pumping time is $\lesssim$ 15 µs) the phase distortion cannot be kept on the one wave level. In this case the focussing properties will be seriously affected and may render the beam useless for high power applications unless a phase correction devise is introduced.

Acknowledgement

The author wishes to thank Dr. Brederlow, Dr. Eidmann and Dr. Volk for the many useful discussions.

## 5. Literature

/ 1/ I.M. Belousova, O.B. Danilov, I.A. Sinibina, V.V. Spiridonov
Sov. Phys. JETP Vol. 31, No. 5, p. 791 (Nov. 1970)

/ 2/ K. Hohla, K.L. Kompa
Zeitschrift f. Naturforschung, Bd. 27a, Heft 6, S. 938 (1972)

/ 3/ L.E. Gobulev, V.S. Zuev, V.A. Katulin, V.Yu. Nosach, O.Yu. Nosach
Kvant. Elekr. 6 (18), p. 23 (1973)

/ 4/ O.B. Danilov, N.A. Novoselov, V.V. Spiridonov
Opt. Spectrosc. Vol. 39, No. 4, p. 382 (Oct. 1975)

/ 5/ O.B. Danilov, N.A. Novoselov, V.V. Spiridonov
Opt. Spectrosc. Vol. 41, No. 1, p. 68 (July 1976)

/ 6/ K. Witte, G. Brederlow, K. Eidmann, E. Fill, R. Volk
ASTERIX III, A Terawatt Iodine Laser
Conference on "High Power Lasers and Applications",
Munich, June 20 - 22, 1977

/ 7/ T.D. Padrick, R.E. Palmer
J. Chem. Phys. Vol. 62, No. 8, p. 3350 (April 1975)

/ 8/ E. Becker
Gasdynamik
B.G. Teubner Verlagsgesellschaft, Stuttgart, 1966

/ 9/ K. Witte
Eine Abschätzung der Ausbleichverluste am Jodlaser
IPP-Report IV/88, Juni 1976

/10/ G.A. Fisk
The Effect of Chemical Kinetics and Starting Material Regeneration
on the Efficiency of an Iodine Laser Amplifier
SAND 77 - 0880, May 1977

/11/ G. Brederlow, K.J. Witte, E. Fill, K. Hohla, R. Volk
The ASTERIX III Pulsed High Power Iodine Laser
IEEE J. Quant. Electr. Vol. QE-12, No. 2, p. 152 (Febr. 1976)

/12/ J.T. Hunt, P.A. Renard, R.E. Nelson
Appl. Optics, Vol. 15, No. 6, p. 1458 (June 1976)