THERMAL CHANGE OF THE REFRACTIVE INDEX OF A LINEAR ABSORBER BY AN INTENSIVE LIGHT WAVE

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The paper deals with the possibility of studying thermally induced refractive index changes, which arise when an intensive light wave passes through the index, by the method of phase-modulation of a probe light pulse. The method described here does not depend on the mechanism of inducing refractive index changes. Thermal perturbations of the refractive index of an absorbing medium, which had been induced by a Q-switch ruby laser pulse, were studied experimentally. In the course of experiments both water solutions of $CuSO_4 \times 5H_28$ and ethanol solutions of a briliant green were used.

ТЕМПЕРАТУРНОЕ ИЗМЕНЕНИЕ ПОКАЗАТЕЛЯ ПРЕЛОМЛЕНИЯ ЛИНЕЙНО ПОГЛОЩАЮЩЕГО ВЕЩЕСТВА ПРИ ПРОХОЖДЕНИИ ЧЕРЕЗ НЕГО ИНТЕНСИВНОЙ СВЕТОВОЙ ВОЛНЫ

В работе описывается возможность исследования изменений показателя преломления линейно поглощающего вещества, возникающих при переходе через это вещество интенсивной световой волны. Исследования осуществляются при помощи метода фазовой модуляции зондирующего светового импульса. Приведенный метод не зависит от механизма возникновения изменения показателя преломления. Проведены экспериментальные исследования температурных зависимостей показателя преломления водного раствора медного купороса и красителя (бриллиантовый зеленый) в этиловом спирте, которые обусловлены монохроматическим импульсом рубинового лазера.

I. INTRODUCTION

An intensive light wave changes the optical properties of the medium it passes through. The effect of this perturbation upon its space as well as time characteristics can be used to determine either the changes of the optical properties of a medium or those of a laser beam if one of them is known. This article deals with refractive index changes induced thermally in linearly absorbing isotropic liquids.

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II. THEORETICAL PART

II a. Refractive index change of a liquid

A relationship between the refractive index of an isotropic liquid and its microstructure is expressed by the known Lorentz-Lorenz relation

$$(n^2-1)/(n^2+2)=K\alpha\varrho\,, \tag{1}$$

where α is the polarizability of a molecule, ϱ the density of a medium and K is a constant proportional to the mass of the molecule. It follows from relation (1) that the index of refraction of a medium may be changed by an intensive light wave varying both the density of the medium $\Delta\varrho$ and the polarizability of the molecule

$$\Delta n=\frac{(n_0^2-1)\,(n_0^2+2)}{6n_0}\left[\frac{\Delta\varrho}{\varrho_0}+\frac{\Delta\alpha}{\alpha_0}\right], \tag{2}$$

where n_0, ϱ_0, α_0 are the initial values of the index of refraction, the density of the medium and the polarizability of its molecules, respectively.

There are two reasons which involve changes of the polarizability of molecules in the electric field of an intensive light wave. First, the molecules of the medium become anisotropic in the direction perpendicular to the optical field vector and the direction colinear with it because of the electronic Kerr mechanism [1]. The response time of this mechanism is determined by the time necessary for the electron polarization to arise, i.e. $t_e\sim10^{-15}-10^{-14}$ s [2]. In liquids, which consist of anisotropic molecules the high-frequency Kerr effect dominates as a mechanism varying the polarizability of molecules, that means such an orientation of anisotropic molecules that their induced dipole moment is maximal. The degree of orientation of molecules in the field of an intensive light wave is restricted by their thermal movement. The change of the polarizability of molecules due to the high-frequency Kerr effect, arise at the time of the order of $t_k\sim10^{-12}-10^{-11}$ s [2]. Generally the polarizability change causes a refractive index change which can be expressed formally as

$$(\Delta n)_\alpha=n_2E_0^2+n_4E_0^4+\dots\,, \tag{3}$$

where E_0 is the intensity of the electrical field of a light wave. The constant n_2, which is called the optical Kerr constant of an isotropic medium, depends both on the properties of the medium and the wavelength of the used light.

When an intensive light pases through a medium density, changes can also arise from either the heating of the medium or the electrostriction. A certain part of the energy of the light wave traversing an absorbing medium is absorbed in it. Therefore its temperature increases by $\Delta\vartheta$ and so does the pressure by the value Δp_ϑ. When the medium is polar it is deformed by the optical field through the

electrostriction pressure Δp_s, too. This pressure perturbance is positive and proportional to the time-averaged square of the intense electric field in the medium. The thermal pressure change is negative and proportional to the heat transferred to the medium by the laser beam. If we assume the perturbations of the arising pressure about its ambient value p_0 to be small, we can determine the arising density change $\Delta\varrho$ by solving the linear hydrodynamic equations [3]. These perturbations will arise at the time t_z determined by both the sound speed v_z in the medium and the radial magnitude r of the area where the change arises, i.e. $t_z\sim r/v_z$.

It follows from the mechanisms presented here that after a light wave had affected a medium, heating the medium was the dominant mechanism of changing its refraction index. The heating of the medium dominates the sooner the larger the absorption coefficient is [3]. The thermal variation in the density of a medium fades out through heat conduction. The characteristic time of heat dissipation is defined as $t_c=r^2\varrho_0c_p/\eta$ [4], where c_p is the specific heat under constant pressure, η is the thermal conductivity.

II b. Phase-modulation of a light wave

Let us suppose that a plane light wave passes through an absorbing medium in two direction perpendicular to its surface. Let n_0 be the index of refraction of the medium and h its thickness. The z-axis is identical with the direction of the beam propagation and the r axis is in the radial direction. Let the radial amplitude distribution be

$$\psi_i(r)=\psi_{0i}F(r)\,. \tag{4}$$

The traversing light beam changes the refraction index from a constant value of n_0 to

$$n(r,\,z)=n_0+\Delta nf(r,\,z)\,, \tag{5}$$

i.e. it changes the optical thickness of a medium from its initial value Δ_0 to $\Delta_0+\Delta(r)$, where

$$\Delta(r)=\Delta n\int_0^h f(r,\,z)\,\mathrm{d}z\,. \tag{6}$$

If another light wave with radial amplitude distribution traverses the medium in a time t after the refractive index change has been induced, the second light wave is phase modulated while passing through the medium. Under our conditions we can obtain from the Kirchhof-Huygens difraction formula the angular (φ) distribution of its relative energy density $E_{2R}(\varphi,\,\Delta)$ in the Fraunhofer region in the form.

$$E_{2R}(\varphi, \Delta)=\frac{E_2(\varphi, \Delta)}{E_2(0, 0)}=C\left\{\left[\int_0^a G(\varphi, r)\cos\left[\frac{2\pi}{\lambda}\Delta(r, t)\right]\mathrm{d}r\right]^2+\right.$$
$$\left.+\left[\int_0^a G(\varphi, r)\sin\left[\frac{2\pi}{\lambda}\Delta(r, t)\right]\mathrm{d}r\right]^2\right\}, \quad (7)$$

where C is a constant of proportionality and

$$G(\varrho, r)=(1+\cos\Theta)\, rF(r)\, J_0\left(\frac{2\pi}{\lambda}\varphi r\right). \quad (7a)$$

$\Theta(r)$ is the angle between the normal to the wavefront of the light emerging from an absorbing medium at the point with a radial abscisa r and the direction φ. J_0 is the Bessel integral. The upper limit of integration in (7) is determined by the radial magnitude of the area at the exit of the absorber, where the amplitude of the phase-modulated light wave differs from zero. There is advantage in using a far-field region. We can eliminate a phase-modulation of a light wave caused by the geometry of a theoretical model or the experimental arrangement. Therefore it is possible to consider the phase-modulation associated only with the refractive index change of the absorbing medium.

III. EXPERIMENTAL PART

To investigate the thermal changes of the refraction indes arrising in the linear absorber when intensive pulses of a ruby laser have been passing through it the method of the phase-modulation of a probe light pulse was used. Attention was directed to the energy density distribution of the probe beam. The distribution was analysed according to the derived relation (7).

Fig. 1 shows the arrangement of the experiment. A ruby laser 1 operated in the two-pulses mode. After a portion of the beam had been selected by the pinhole

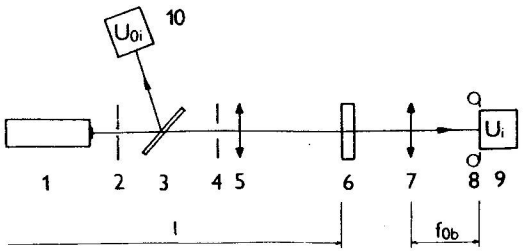

Fig. 1. Experimental arrangement for the study of the thermal changes of the refractive index. 1 — ruby laser; 2, 4 — apertures; 3 — beam splitter; 5 — lens; 6 — cell filled with an absorbing dye solution; 7 — objective; 8 — plane of film; 9, 10 — detectors.

2 and the aperture 4, it passed the lens 5 forming a space diverging light pulse. We can suppose its radial energy density distribution to be Gausian [5]. This light pulse entered a cell 6 filled with a linearly absorbing medium. In the course of experiments water solutions of $CuSO_4 \times 5\ H_2O$ with a thickness $h=0.095$ cm and absorption coefficients $k_1=13.2\ \mathrm{cm}^{-1}$ and $k_2=8.6\ \mathrm{cm}^{-1}$ were used. Ethanol solutions of a brilliant green with $h=0.097$ cm, $k_1'=14.3\ \mathrm{cm}^{-1}$, $k_2'=8.7\ \mathrm{cm}^{-1}$ were used as well. Behind the lens 5 the light flux entering a liquid cell 6 was modified by moving the cell axially (along the z direction). The Fraunhofer region was realized by observing in the local plane of the objective 7 with a focal length $f_{ob}=5.8$ cm. The angular distribution of the light energy density was recorded photographically on the film 8. Energies of the pulses were monitored by using of detectors 9 and 10 modified for this purpose [6]. The laser operated in the Q-switch mode, accomplished by a passive Q-switch (a solution of VOPc in nitrobenzene). The length of each pulse was $t_i \sim 3\times10^{-8}$ s and the time interval between them was about $t\sim10^{-5}$ s. The minimum radius of the energy distribution of light pulses was of 1.96×10^{-5} m, the wavelength $\lambda=694.3$ nm.

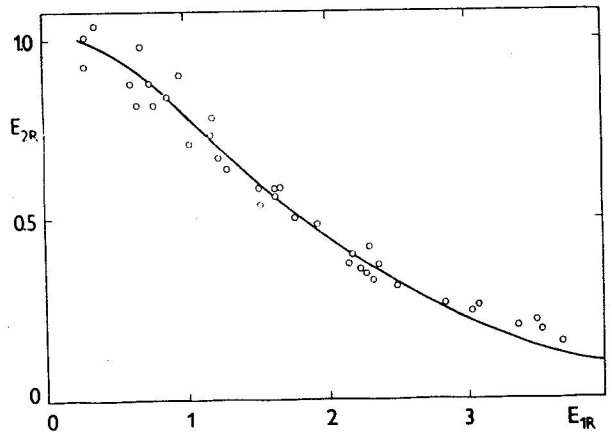

Fig. 2. The plot of the decrease of the relative energy density of a probe pulse on the beam axis vs the energy of a pulse inducing a refraction index change on a water solution of $CuSO_4 \times 5\ H_2O$, $k=8.6\ \mathrm{cm}^{-1}$, $h=0.095$ cm.

Fig. 2 illustrates the dependence of a decrease of the relative energy density $E_{2R}(0, \Delta)$ of the probe pulse in its central part after it has passed through one of the water solutions of $CuSO_4 \times 5\ H_2O$ with the energy density E_{1R} of the first pulse (for $\varphi=0$), obtained experimentally. The ratio U_{01}/S_1 (where U_{01} is the voltage of the detector 10 and S_1 is an area restricted by the radius of the Gaussian intensity distribution at the position 1 of the cell 6) was a measure of the relative unit E_{1R}. In

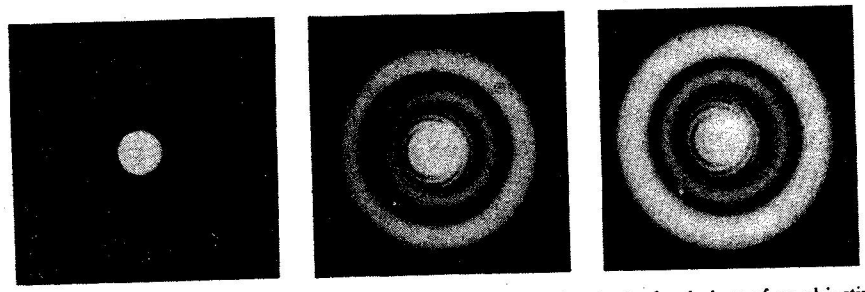

Fig. 3. Photographic records of radial energy distributions of pulses in the focal plane of an objective after they have passed an investigated medium. a — superposition of two pulses after they have passed through a solvent ethanol; b — one pulse after passing the ethanol solution of a briliant green; c — superposition of two pulses after passing the same dye solution.

this case the values of t_z and t_c were of the order of $t_z > 8 \times 10^{-8}$ s, $t_c > 5 \times 10^{-3}$ s depending on the magnitude of the beam cross section in the investigated medium.

Fig. 3 shows the records of a superposition of the radial energy density distribution of both pulses in the focal plane of the objective 7, after they have passed through one of the ethanol solutions of a brilliant green, obtained photographically. In this case $t_z > 3 \times 10^{-8}$ s and $t_c > 6 \times 10^{-4}$ s.

IV. DISCUSSION

It follows from the experimental set up that the radial amplitude distribution of both pulses entering a medium can be regarded as the Gaussian with a radius a_0. After we have introduced a dimensionless radial variable quantity $\xi = r/a_0$ the amplitude distribution function can be expressed in the form

$$F(\xi) = \exp\{-\xi^2\}. \tag{8}$$

Substituting $a = 2a_0$ (i.e. $\xi' = 2$) for the upper limit of integration in (7) the light amplitude at the distance a from the axis can be supposed to be much smaller than the amplitude on the axis ($\psi(2) \doteq 0.02\ \psi(0)$). With regard to the values of absorption coefficients of the used solutions it can be shown [3] that heating has already become important during the light pulse. Thus the refraction index change can be supposed to be proportional to a portion of the light energy absorbed and changed into heat. The change of the optical thickness of an absorbing medium

$$\Delta(\xi) = \frac{\gamma\, \mathrm{d}n/\mathrm{d}\vartheta}{\varrho_0 c_p} (1 - T)\, E_i(0) \exp\{-2\xi^2\} \tag{9}$$

depends linearly on the energy density of a light pulse. The efficiency of transformation of the absorbed light energy into heat is denoted as γ, T is the

transmittance of the medium and $\mathrm{d}n/\mathrm{d}\vartheta$ is the rate of the change of the refractive index with temperature ϑ. A Gaussian radial change distribution $\Delta(\xi)$ determined by the relation (9) starts to fade out owing to thermal conduction with a characteristic time t_c. As in our experiments the condition $a_0 \ll h$ is fulfilled at each position of the liquid cell realized it is possible to consider only the dissipation of $\Delta(\xi)$ in the radial direction. Therefore the radial distribution of the optical thickness of the medium at a time t after it has arisen is given as a solution of the heat conduction equation

$$\Delta(\xi, t) = \frac{\Delta(0)}{\tau} \exp\{-2\xi^2/\tau\} \tag{10}$$

where $\tau = 1 + t/t_c$ and the change of the optical thickness on the z axis is negativ, i.e. $\Delta(0) < 0$.

The solid curve in Fig. 2 is calculated from data obtained experimentally. We used relations (7), (8) and (10) and substituted $\tau = 1$. Analysing our experiments we assumed the conditions $t_i < t_z$, t_c and $t \ll t_c$ to be satisfied with regard to the values of t_i, t_z, t_c and t. Thus we could suppose that the selfphase-modulation of a pulse had not occurred and therefore the first pulse had induced a perturbation of the refractive index which caused the phase modulation of the second pulse (the probe one), whereby $\tau \doteq 1$. Some theoretical curves plotted in Fig. 4 illustrate the influence of parameter τ on the dependence of the relative energy density decrease of a probe pulse on the axial change of the optical thickness of the medium $\Delta(0)$.

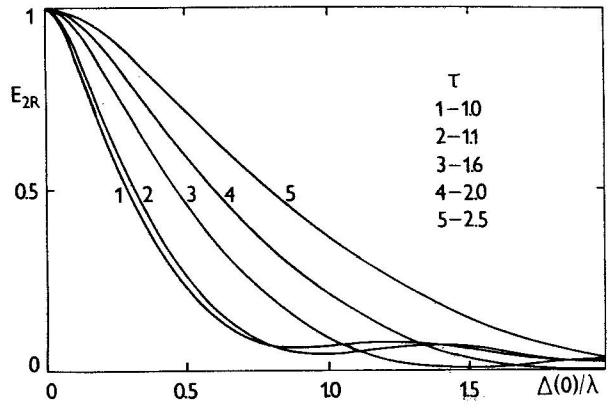

Fig. 4. The influence of parameter $\tau = 1 + t/t_c$ on the dependence of the decrease of the probe pulse energy on the beam axis after passing through an absorber on the change of its optical thickness on the beam axis (dependence obtained numerically for the Fraunhofer region).

The experimental results were analysed assuming the relation between the energy density of the incident pulse and the induced change of the optical thickness of the medium to have been linear, i.e.

$$\Delta(0) = q \,.\, E_{01} \,. \tag{11}$$

If two absorbers differ from each other by their parameters, the condition

$$q_1/q_2 = X_1/X_2 \tag{12}$$

must be fulfilled as a consequence of (9) and (10). A substitution

$$X_i = \frac{\gamma_i \, \mathrm{d}n_i/\mathrm{d}\vartheta}{\varrho_{0i} c_{pi}} (1 - T_i)$$

was introduced in order to express eq. (12). As the coefficients γ_i of the investigated solutions were not known, only the results obtained with the same type of absorbers with different transmissions were compared to verify eq. (12). The results listed in Tab. 1 confirm within the experimental errors the suppose linear

Table 1
Comparison of theory and experiment

Absorber	T	Q_T	Q_E
1	0.29 ± 0.01	1.28 ± 0.07	1.30 ± 0.04
2	0.44 ± 0.01		
3	0.25 ± 0.01	1.32 ± 0.08	1.44 ± 0.05
4	0.43 ± 0.01		

1, 2 — water solutions of $CuSO_4 \times 5\, H_2O$; 3, 4 — ethanol solutions of a brilliant green; $Q_T = X_i/X_{i+1}$; $Q_E = q_i/q_{i+1}$ where i is equal either to 1 or to 3.

dependence of the change of optical thickness of an absorber on the energy density of a light beam — eq. (11). The assumption that the dependence $\Delta(0) = f(E_{01})$ is linear is correct as it follows from the conditions of our experiment. We use linearly absorbing dye solutions and light pulses with parameters which were suitable with regard to the observation of the refractive index changes induced thermally ($t_i < t_z$, t_c and $t/t_c \ll 1$). Therefore the method showed in the present paper, i.e. the measurement of the energy density of a probe beam on its axis in the Fraunhofer region after it has passed through an absorbing solution, makes it possible to obtain some information about the thermal change of its optical thickness and information about the refractive index change through eq. (6) as well. As one can see from the relationships (6), (9) and (10), the refractive index change to be observed depend on both thermal and spectral properties of a solution and the parameters of light pulses (intervals between them, pulse width, energy density, ...). That means, that

the method presented here allows to determine either the thermal parameters of a linearly absorbing solution (if the parameters of the light pulses are known) or the parameters of pulses (if the properties of the absorbing solution are known).

When the pulsewidth t_i is comparable with a time t_z, which is necessary to develop a refraction index change, the radial distribution of the energy of the first pulse also begins to change if it passes through an absorber, too. It is illustrated in Fig. 3 a fine structure at the centre is the result of an interference from the reflections of the cell windows; its energy distribution is not symmetrical because of some deviation of the cell from the direction perpendicular to the beam axis. The assumption that a pulse has no effect on itself falls. The high intensity of the light pulse, which is nearly ten times higher than that used when the E_{1R} dependence of E_{2R} $(0, \Delta)$ has been determined, may be the reason for this fact as well. The distinct structure of the radial energy distribution of a pulse having maxima and minima indicates that a certain change of the optical thickness of an absorber has been developing during the first part of the pulse and has not been varying substantially during the next pulse-time or even during the pulsewidth of the probe beam. Therefore in this case it is necessary to consider a qualitatively new connection between the pulse intensity and the arising change of the optical thickness of the absorber. We must deal with the time development of the refractive index change, i.e. to study the time development of the phase modulation of the pulse passing through an absorbing medium, too.

V. CONCLUSION

It has been shown that the method of phase modulation of a probe beam should be useful to study the thermal changes of the refractive index arising when an intense light is passing a linearly absorbing medium. Although the method is illustrated with respect to the thermal effect, it is applicable to other nonlinear a refractive index changes induced by laser beams. It is only necessary to choose parameters of light pulses suitable with regard to the times characteristic of the developing and the dissipation of these changes, or to investigate the time development of self-phase-modulation of an intense light pulse passing the medium.

REFERENCES

[1] Brewer, R. G., Lifsitz, J. R., Garmine, E., Chiao, R. Y., Townes, C. H.: Phys. Rev. *166* (1968), 326.

[2] Volkenstein, M. V.: *Strojenie i fizičeskije svojstva molekul.* AN SSSR. Moskva—Leningrad 1955.

[3] Rajzer, Ju. P.: ŽETF X (1967), 470.

[4] Gordon, J. P., Leite, R. C. C., Moore, R. S., Porto, S. P. S., Whinnery, J. R.: Journ. of Appl. Phys. *36* (1965), 3.

[5] Vojtek, P., Senderáková, D., Štrba, A.: Univ. Comen., Physica *XIII*, (1982).

[6] Lindner, F., Štrba, A., Vojtek, P.: Čs. čas. fyz. A *31* (1981), 272.

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