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EXPLOITATION OF THE GAUSSIAN BEAM FOR THE INVESTIGATION OF THE NONLINEAR ABSORPTION OF LIGHT

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In the present paper the transition of the Gaussian light beam through a nonlinearly absorbing medium is investigated. The transmission of the beams with a homogeneous and a Gaussian transverse intensity distribution, respectively, in the absorber with two excited singlet energy levels is theoretically compared. The nonlinear absorption of the Gaussian beam in the solution of two organic dyes is experimentally investigated. The method enables the determination of all parameters which characterize the nonlinear absorber and demonstrates the convenience of the Gaussian beam for the investigation of the nonlinear absorption of light.

ИСПОЛЬЗОВАНИЕ ГАУССОВА ПУЧКА ДЛЯ ИЗУЧЕНИЯ НЕЛИНЕЙНОГО ПОГЛОЩЕНИЯ СВЕТА

В работе изучается распространение светового пучка гауссова типа в среде с нелинейным поглощением. Проводится теоретическое сравнение прохождения пучка в поглотителе с двумя одиночными энергетическими уровнями для случаев однородного и гауссова распределения поперечной интенсивности пучка соответственно. Экспериментально исследовано нелинейное поглощение гауссова пучка в растворе двух органических красителей. Данный метод позволяет определить все параметры, которые характеризуют нелинейный поглотитель и доказывает преимущества гауссова пучка при исследовании нелинейного поглощения света.

I. INTRODUCTION

The decrease of the light intensity $\mathrm{d}u$ at a distance $\mathrm{d}z$ in an absorbing medium in the region of the low intensities is a linear function of intensity u

$$\mathrm{d}u = -k\, u\, \mathrm{d}z, \tag{1}$$

where k is the absorption coefficient. We can determine the light intensity u_2 of the

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beam at the distance z by integrating equation (1), which gives the well-known Bouguer law

$$u_2 = u_1 \, e^{-kz}, \tag{2}$$

where u_1 is the input intensity of light. The absorption coefficient k is a function of the light intensity for intensive laser beams. If the decrease of intensity du at the distance dz is not a linear function of the intensity u, then we speak about a nonlinear absorption of light. Then the Bouguer law in the form (2) is not valid and by integrating equation (1) we get the relation

$$\int_{u_1}^{u_2} \frac{du}{k(u)u} = -z \tag{3}$$

that enables us to determine the intensity u_2. A medium in which nonlinear absorption can be observed will be called a nonlinearly absorbing medium. The dependence of the transmission on the intensity of light is a fundamental characteristic of that medium. The transmission is defined as a ratio of the radiant power Φ_2 at the exit from to that at the entrance to the absorbing medium Φ_1. The quotient of powers Φ_2/Φ_1 is equal to the quotient of intensities $T = u_2/u_1$ for the light beam with the homogeneous distribution of intensity in the cross section. We shall determine the intensity u_2 from eq. (3). We labelled the quotient Φ_2/Φ_1 as T_G for the beam with the Gaussian distribution of intensity in the cross section. We shall show that the dependence of the transmission T_G on the intensity of the light u can render the same information about an absorbing medium as the dependence $T(u)$. Furthermore, measuring the radiant powers Φ_2 and Φ_1, respectively, enables us to avoid the unfavourable influence of the phase modulation of the beam that occurs particularly at high intensities [1].

II. THE NONLINEAR ABSORBING PROPERTIES OF THE MEDIUM

The quantum theory describes an absorbing medium as a set of energy levels corresponding to the allowed states of the molecules of the medium. The absorption coefficient is then a function of the transition probabilities between the levels. In the model with two excited singlet levels the absorption coefficient has the form [2]

$$k = k_0 \frac{1 + Au}{1 + \alpha u}, \tag{4}$$

where k_0 is the absorption koefficient for the low intensities of light, $A = B_{23}/A_{21}$, α is the coefficient of nonlinearity that may be expressed as

$$\alpha = \frac{B_{12} + B_{21}}{A_{21}}. \tag{5}$$

The constants A_{ij}, B_{ij} are Einstein's coefficients of the spontaneous and the stimulated transition between the respective levels. The index 1 denotes the ground level, the indices 2, 3 denote the first and the second excited level, respectively. A solution of equation (3) for the medium with the absorption coefficient of the form (4) gives a transcendent relation for the dependence of the transmission T on the intensity u_1

$$\ln \frac{T_0}{T} = \left(\frac{\alpha}{A} - 1\right) \ln \frac{1 + ATu_1}{1 + Au_1}, \tag{6}$$

in which $T_0 = \exp(-k_0 z)$ is the transmission for the low intensities u_1. For the high intensities $u_1 \to \infty$ the transmission T acquires according to equation (6) the final value $T_2 = \exp(-k_2 z)$, where $k_2 = k_0 A/\alpha$. Such a medium is often called the medium with the residual absorption. Namely, the Bouguer law of the form (2) with a constant absorption coefficient k_2 for the high intensities of the light is valid again.

III. THE ABSORPTION OF THE GAUSSIAN BEAM

The Gaussian distribution of the intensity u_1 in the cross section is given by the form

$$u_1 = u_{01} \exp(-R^2/R_0^2), \tag{7}$$

where u_{01} is the intensity on the axis of the beam, R_0 is the radius of the beam, R is a distance from the axis of the beam. It follows from the theory of propagation of the Gaussian beam [3] that the radius R_0 is a hyperbolic function of the distance z

$$R_0^2(z) = w_0^2 \left[1 + \left(\frac{2z}{k' w_0^2}\right)^2\right], \tag{8}$$

where w_0 is the radius of the vaist of the beam in the plane $z = 0$; $k' = 2\pi/\lambda$ is the wavenumber.

We assume that the absorbing medium through which a Gaussian beam is propagating is sufficiently thin so that a change of the radius of the beam R_0 can be neglected. We shall be interested in the character of the above mentioned transmission

$$T_G = \Phi_2/\Phi_1. \tag{9}$$

The radiant power Φ_1 at the input of the absorber can be expressed analytically

$$\Phi_1 = \int_0^{2R_0} u_1 2\pi R \, dR = u_{01} \pi R_0^2 (1 - e^{-4}). \tag{10}$$

The radiant power Φ_2 at the output of the absorber can be obtained by solving the integral

$$\Phi_2 = \int_0^{2R_0} u_2 2\pi R \, dR \tag{11}$$

using the transcendental equation (6) for u_2. In both cases (10) and (11) we confined ourselves to the limiting distance $2R_0$ from the axis of the beam, since approximately 98 % of the total energy of the beam are concentrated in this region. The integral expressing the radial power Φ_2 can be solved numerically. The result of solving T_G proves beyond doubt that the value of T_G is independent of the radius R_0, it is only the function of the intensity u_{01}. The dependences of T_G and T, respectively, on the dimensionless quantity $\overline{u_{01}} = \alpha u_{01}$ for the values $T_0 = 1.58$ % and $T_2 = 63.10$ % are shown in Fig. 1. It can be seen from this graph that the character of the dependence $T_G(\overline{u_{01}})$ is similar to that of the dependence $T(\overline{u_{01}})$. The absorbing ability of the medium decreases if the intensity of light increases, i.e. the transmission is increasing until it reaches the constant value T_2 at the high intensity $\overline{u_{01}}$. The dependence of the quotient T_G/T on $\overline{u_{01}}$ is also shown in Fig. 1. It reaches its minimum value in the region of the bleaching of the absorber. Both dependences $T(\overline{u_{01}})$ and $T_G(\overline{u_{01}})$, respectively, are characteristic for the absorbing medium and they are fully determined by the constants k_0, A, α from eq. (4), which may be reversely determined from them.

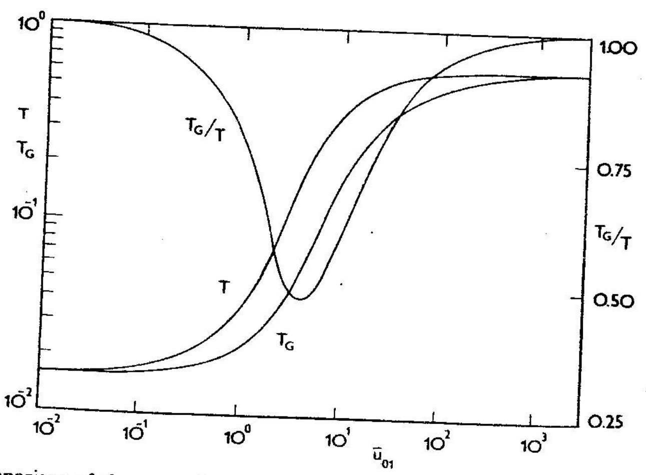

Fig. 1. The comparison of the transmission T of the light beam with a homogeneous distribution of intensity in the cross section with the transmission of the beam with the Gaussian distribution. The dimensionless quantity $\bar{u}_{01} = \alpha u_{01}$ determines the input light intensity of the nonlinear absorber.

IV. THE EXPERIMENTAL INVESTIGATION OF THE NONLINEAR ABSORPTION

The diagram of our experimental setup for the investigation of the nonlinear absorption is shown in Fig. 2. A ruby laser operating in the regime of the giant pulses was chosen as the source of the intensive radiation. The experimental arrangement permits to consider the light beam behind the lens to be Gaussian with

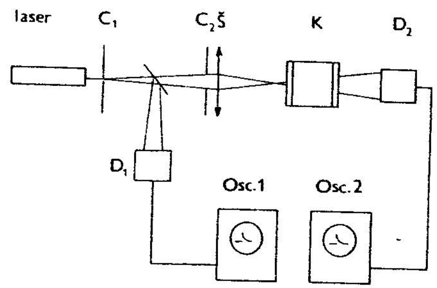

Fig. 2. The experimental arrangement of the investigation of the nonlinear absorption of the Gaussian beam. C_1, C_2 — diaphragms, S — lens, K — cell with the investigated absorber, D_1, D_2 — photoelectric detectors, Osc. 1, Osc. 2 — oscilloscopes.

the waist at the plane of the image of the circular diaphragm C_1 [4]. The value of the magnitude of the intensity u_{01} at the input to the cell may be changed by moving the cell along the beam axis and can be determined from equation (10) in the form

$$u_{01} = \frac{\Phi_1}{\pi R_0^2 (1 - e^{-4})}. \tag{12}$$

The information about the radiant power Φ_1 is given by the detector D_1, which registers the fraction of the power that is separated by the beam splitter. The radius R_0 may be calculated from eq. (8). The detector D_2 registers the radiant power at the output from the cell. The signals from the two detectors D_1 and D_2 were displayed on the oscilloscope Ocs 1, Osc 2. The transmission of the absorbing medium was determined with respect to the transmission of the cell filled with the solvent.

In the experiment we investigated the nonlinear absorption of the light in two solutions of organic dyes. The vanadyl phtalocyanine (VOPc) dissolved in nitrobenzene is widely used as a passive Q-switch for the ruby laser. BEFBTPc-chlorid dissolved in etanol may be used as the active medium of the dye lasers. We have used the VOPc as the calibrated nonlinear absorbing medium for our experiment.

The results of the experiment in the form of the dependence $T_G(u_{01})$ for BEFBTPc-chlorid are shown in Fig. 3. The transmission T_0 for the low intensities of light has been measured by a spectrometer.

V. DISCUSSION

The experimentally obtained dependence $T_G(u_{01})$ for VOPc and the known value of the coefficient $\alpha = 5 \times 10^{-10}\ W^{-1}\ m^2$ [2] for this dye enables us to calibrate the magnitude of the signals of the detector D_1 on the units of the quantity u_{01}. From the results in Fig. 3 we then get the value of the nonlinearity coefficient for

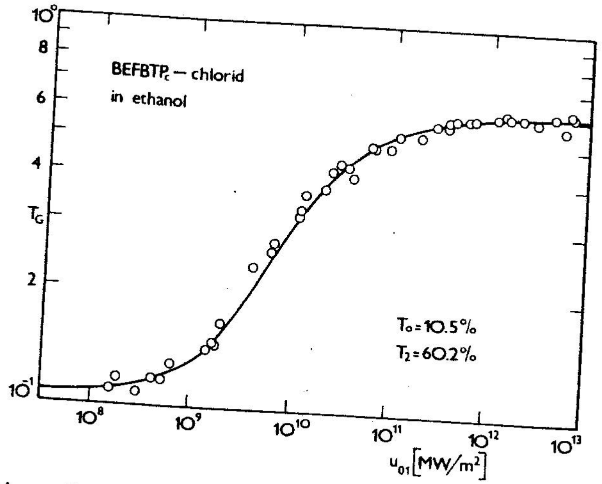

Fig. 3. The experimentally obtained dependence of transmission $T_G(u_{01})$ for the Gaussian beam in nonlinearly absorbing solution of BEFBTPc-chlorid in ethanol.

BEFBTPc-chlorid in etanol, $\alpha = 6 \times 10^{-10}\ W^{-1}\ m^2$. The transmission T_0 and T_2 together with the coefficient α make it possible to determine the value of the constant A from eq. (4), $A = 1.4 \times 10^{-10}\ W^{-1}\ m^2$. Thus the parameters characterizing the nonlinear absorption in this dye are fully determined. The theoretical curve with these parameters is in Fig. 3, drawn in full line.

The results of the measurements in Fig. 3 show a partial dispersion of the experimental points around the theoretical curve. We shall try to assess the influence of some factor on the obtained results. In the calculation of the theoretical curve the divergence of the beam on the length of the cell thickness was neglected. We used a 2 mm thick cell in the experiments. It gives a relative change of the radius 0.4 % on the length of the cell for the greatest divergence angle of the used beam of 0.8°. We did not take into account the temporal pulse shape. The detectors D_1 and D_2 detect the whole energy of the pulse in the cross section of the beam. The consideration of this fact leads to the dependence of the energy transmission $T_E = E_2/E_1$ on the light intensity. E_2 is the energy of the pulse at the output of the absorber and E_1 is the energy at the input, respectively. The

theoretical calculations of the dependence $T_E(u)$ give the same character of the dependence as that of $T_G(u)$ [5]. Then with regard to the calibration of the intensities the axis u_{01} it is not crucial for the evaluation of the results with the quantity T_E. The evaluation of the results with the quantity T_G is correct in the case of the constant duration of the pulse. However, its value may be changed and contribute to the dispersion of the measured points in this way. The nonreproducibility in the distribution of the intensity in the cross section of the beam may have the same influence. This nonreproducibility is caused by the inhomogeneous distribution of the intensity on the diaphragm C_1.

VI. CONCLUSION

Regardless of the mentioned unfavourable influence which may occur in the proposed experiment, the attained results demonstrate the convenience of the application of the Gaussian beam in the investigation of the nonlinear absorption of light. The advantage of this method is the removal of the influence of the phase modulation of the beam on the obtained results. The intensity of light in this method unlike to that of other methods is changed in a simple way by moving the cell along the beam axis. The nonlinear absorption of light may be investigated with any beam similar to the Gaussian beam but it is necessary to know the distribution of the intensity in its cross section.

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