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NONLINEAR OPTICAL PHENOMENA

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Two-photon absorption of light beams of variable cross section

A.A. Gordeev, V.F. Efimkov, I.G. Zubarev

Abstract. **An equation is obtained to describe two-photon absorption of light beams of variable cross section. It is shown that the experimental values of the two-photon absorption coefficients of focused laser beams, calculated in the plane-wave approximation in the focal waist region and taking into account the variable cross section of the light beam, differ by two orders of magnitude.**

Keywords: *two-photon light absorption coefficient, toluene, focused beams.*

1. Introduction

When observing various nonlinear processes in experimental practice, use is often made of the focusing of the exciting radiation to increase its intensity, because the efficiency of the observed nonlinear processes, as a rule, is determined by the intensity of the exciting radiation. This situation, in particular, takes place in observing stimulated temperature scattering caused by two-photon absorption of pump radiation by metal nanoparticles in solutions of some liquids [1–5] or in pure liquids [3, 5]. The two-photon absorption coefficient depends on the square of the intensity of the exciting radiation. Therefore, it is often assumed that absorption mainly occurs in the region of the focal waist [2–6], where the pump intensity has a maximum value. Since the wavefront of the exciting radiation is plane in this region, the plane-wave approximation can be used to describe the occurring processes. As will be shown below, when the real variable of the transverse beam structure is taken into account, the results differ significantly, at least when focusing the exciting radiation with sufficiently short-focus lenses with a numerical aperture $N \geqslant 0.1$.

2. Experimental results

The schematic of the experimental setup is shown in Fig. 1. Pumping was carried out by radiation of the second harmonic of a single-mode single-frequency pulsed neodymium laser. In the experiments, we measured the energy and shape of the radiation pulses incident on a cell with liquid toluene and of transmitted radiation pulses.

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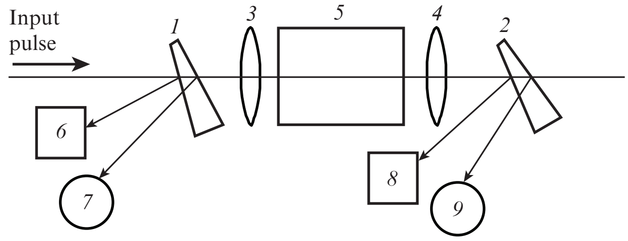

Figure 1. Experimental setup:
(*1*, *2*) glass wedges; (*3*) lens with f = 3 cm; (*4*) lens with f = 7 cm; (*5*) cell with toluene, length l = 7 cm; (*6*, *8*) calorimeters; (*7*, *9*) fast photodiodes.

The exciting radiation was focused by lens *3* with a focal length of 3 cm into a cell (*5*) with liquid toluene. Using lens *4* with a focal length f = 7 cm, the divergence of the radiation emerging from the cell was compensated for in order to deliver it without loss to the corresponding meters (*8*, *9*).

Figure 2 shows the experimental dependence of the energy of the pulses emerging from the cell on the energy of the incident pulses. When plotting it, we took into account the losses due to Fresnel reflection from all optical surfaces through which the corresponding laser pulses passed.

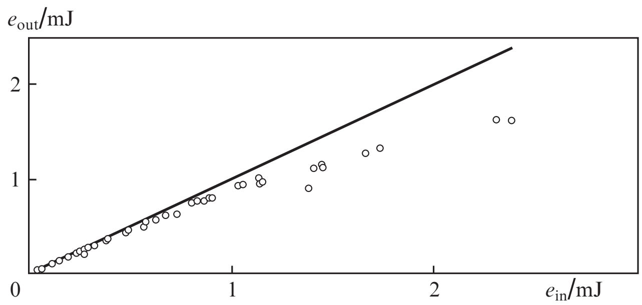

Figure 2. Dependence of the energy e_{out} of the pulses passed through the toluene cell on the energy e_{in} of the incident pulses. The straight line corresponds to the transmission of the cell in the absence of two-photon absorption.

3. Calculation of the coefficient of two-photon absorption of radiation in the plane-wave approximation

In this model, the evolution of the intensity I of the exciting radiation is described by the equation,

$$\frac{dI}{dz} = -\alpha I - \beta I^2, \tag{1}$$

which takes into account possible weak linear absorption. Here, α and β are the linear and two-photon absorption coefficients, respectively. The variables in (1) are separated, and therefore, solving this equation, we obtain

$$\int_{I_{in}}^{I_{out}} \frac{dI}{I[1+(\beta/\alpha)I]} = -\alpha\int_0^l dz,$$
$$\ln\left|\frac{I_{out}}{1+(\beta/\alpha)I_{out}}\right| - \ln\left|\frac{I_{in}}{1+(\beta/\alpha)I_{in}}\right| = -\alpha l, \tag{2}$$

and after converting, the last expression has the form

$$I_{out} = \frac{I_{in}e^{-\alpha l}}{1+(\beta/\alpha)I_{in}(1-e^{-\alpha l})}. \tag{3}$$

As can be seen from Fig. 2, $\alpha l \ll 1$ in our case, and so the solutions takes the form

$$I_{out} = \frac{I_{in}}{1+\beta l I_{in}}. \tag{4}$$

Using this expression, we calculate the two-photon absorption coefficient:

$$\beta = \frac{1}{I_{in} l}\left(\frac{I_{in}}{I_{out}} - 1\right). \tag{5}$$

To obtain the average value of the two-photon absorption coefficient β with allowance for the experimental measurement errors, we use the dependence in Fig. 2 to form Table 1 for five values of the energies of incident pulses, multiples of 0.5 mJ.

Table 1.

e_{in}/mJ	e_{out}/mJ
0.5	0.46
1.0	0.88
1.5	1.19
2.0	1.5
2.5	1.67

The FWHM duration of the exciting radiation pulses at the second harmonic frequency of the neodymium laser is $\tau_{in} = 28$ ns. The divergence of this radiation is $\theta = 3\times10^{-4}$ rad. The focal length f_n of lens *3* in the cell, taking into account the refractive index of toluene $n = 1.5$, is 4.5 cm; therefore, the diameter of the focal spot is $d_f = \theta f_n = 13.5\times10^{-4}$ cm. The radius of the Gaussian beam in the focal waist is $w_0 = (\lambda z_0/\pi)^{1/2}$, where $\lambda = 0.53$ µm is the pump radiation wavelength, and z_0 is the focal waist length. Hence, $z_0 = \pi w_0^2/\lambda$, and since $w_0 = d_f/2$, then $z_0 = 2.7\times10^{-2}$ cm. The area of the focal waist is $S_f = \pi w_0^2 = 1.43\times10^{-6}$ cm^2, the radiation intensity is $I = e_{in}/(\tau_{in}S_f)$, where $\tau_{in} = 2.8\times10^{-8}$ s. Using these values, we transform the data in Table 1 into the intensities of the corresponding pulses and then, proceeding from the fact that $l = z_0$, we calculate the two-photon absorption coefficients by formula (5). From the results given in Table 2, we obtain the average value $\beta_{av} = (2.56 \pm 0.26)\times10^{-10}$ cm W^{-1}.

Table 2.

$I_{in}/10^{10}$ W cm^{-2}	$I_{out}/10^{10}$ W cm^{-2}	$\beta/10^{-10}$ cm W^{-1}
1.26	1.15	2.8
2.5	2.2	2.0
3.75	2.97	2.59
5.0	3.75	2.47
6.24	4.17	2.95

4. Two-photon absorption of light beams of variable cross section

The power of the exciting radiation pulses propagating through a medium does not depend on their cross-sectional area. Therefore, it is necessary to obtain an equation describing the evolution of the power of an exciting beam of a variable cross section as it propagates through a nonlinear medium. To this end, we use the expression for the pulse intensity $I(z) = P(z)/S(z)$, where $P(z)$ is the power of the light pulse, and $S(z)$ is its variable cross-sectional area. Let us differentiate both sides of this equality and compare the result with equation (1) without a linear term. As a result, we obtain

$$\frac{dI}{dz} = \frac{1}{S(z)}\frac{dP}{dz} - \frac{P}{S^2}\frac{dS}{dz} = -\beta I^2 = -\frac{\beta}{S^2}P^2, \tag{6}$$

or

$$\frac{dP}{dz} - \frac{P}{S}\frac{dS}{dz} = -\frac{\beta}{S}P^2. \tag{7}$$

The change in the radius of a Gaussian beam along the length of the medium is described by the expression

$$w(z) = w_0\left[1+\left(\frac{z}{z_0}\right)^2\right]^{1/2}, \tag{8}$$

and Gaussian beam area has the form

$$S(z) = \pi w^2(z) = \pi w_0^2\left[1+\left(\frac{z}{z_0}\right)^2\right], \quad \frac{dS}{dz} = \pi w_0^2 2\left(\frac{z}{z_0}\right)\frac{1}{z_0}. \tag{9}$$

Then, denoting the coefficients in equation (7) as

$$\frac{-1}{S}\frac{dS}{dz} = \frac{-z_0^2}{\pi w_0^2(z^2+z_0^2)}\pi w_0^2 2\left(\frac{z}{z_0}\right)\frac{1}{z_0} = -\frac{2z}{z^2+z_0^2} = p(z),$$
$$-\frac{\beta}{S} = \frac{-\beta z_0^2}{\pi w_0^2(z^2+z_0^2)} = q(z), \tag{10}$$

we reduce equation (7) to the classical form of Bernoulli's equation

$$\frac{dP}{dz} + p(z)P = q(z)P^m \tag{11}$$

with the exponent $m = 2$. As is known, the general solution of Bernoulli's equation has the form [7]

$$P(z) = \left\{\exp\left[\int(m-1)p(z)dz\right]\right.$$
$$\left.\times\left\{C + \int(1-m)q(z)\exp\left[\int(1-m)p(z)dz\right]dz\right\}\right\}^{1/(1-m)}. \tag{12}$$

This expression contains the following integrals:

$$\int(m-1)p(z)\,\mathrm{d}z = -2\int\frac{z\mathrm{d}z}{z^2+z_0^2} = -\ln|z^2+z_0^2|,$$

$$\int(1-m)p(z)\,\mathrm{d}z = 2\int\frac{z\mathrm{d}z}{z^2+z_0^2} = \ln|z^2+z_0^2|, \tag{13}$$

$$\int(1-m)q(z)\exp(\ln|z^2+z_0^2|)\,\mathrm{d}z$$

$$= \frac{\beta z_0^2}{\pi w_0^2}\int\frac{\mathrm{d}z}{z^2+z_0^2}\exp(\ln|z^2+z_0^2|) = \frac{\beta z_0^2}{\pi w_0^2}\int\mathrm{d}z = \frac{\beta z_0^2}{\pi w_0^2}z.$$

Substituting these integrals into the general solution, we reduce it to the form

$$P(z) = \left(\frac{C}{z^2+z_0^2} + \frac{\beta z_0^2}{\pi w_0^2}\frac{z}{z^2+z_0^2}\right)^{-1}. \tag{14}$$

Since the exciting radiation is focused into the cell by a lens with a focal length f_n, the range of z variation is determined by the inequalities

$$-f_n \leqslant z \leqslant f_n, \tag{15}$$

where $z = -f_n$ corresponds to the input plane of the cell, and $z = f_n$ corresponds to its output plane. In this case, $z = 0$ corresponds to the focal plane of lens *3* in Fig. 1. As a result, solution (14) takes the form:

$$P_{\text{in}} = \left(\frac{C}{f_n^2+z_0^2} - \frac{\beta z_0^2}{\pi w_0^2}\frac{f_n}{f_n^2+z_0^2}\right)^{-1},$$

$$P_{\text{out}} = \left(\frac{C}{f_n^2+z_0^2} + \frac{\beta z_0^2}{\pi w_0^2}\frac{f_n}{f_n^2+z_0^2}\right)^{-1}. \tag{16}$$

We represent these expressions in the form

$$P_{\text{in}} = \frac{1}{A-B},\quad P_{\text{out}} = \frac{1}{A+B} = \frac{1}{A-B+2B}$$
$$= \frac{1}{(A-B)[1+2B/(A-B)]} = \frac{P_{\text{in}}}{1+2BP_{\text{in}}}, \tag{17}$$

and, as a result, we obtain

$$P_{\text{out}} = P_{\text{in}}\left(1 + \frac{\beta z_0^2}{\pi w_0^2}\frac{2f_n}{f_n^2+z_0^2}P_{\text{in}}\right)^{-1} = P_{\text{in}}\left(1 + \frac{\beta z_0}{\lambda}\frac{2f_n}{f_n^2+z_0^2}P_{\text{in}}\right)^{-1}. \tag{18}$$

In the limiting case, this expression turns into formula (4). Indeed, we substitute the expression $P = IS$ in (18), where $S(z) = \pi w_0^2[1 + (z/z_0)^2]$. As a result, (18) takes the form

$$I_{\text{out}} = I_{\text{in}}\left\{1 + \beta\frac{z_0}{\lambda}\frac{2f_n}{f_n^2+z_0^2}I_{\text{in}}\pi\omega_0^2\left[1+\left(\frac{z}{z_0}\right)^2\right]\right\}^{-1}. \tag{19}$$

In the plane-wave approximation, the focal waist length z_0 increases indefinitely. Then, $(z/z_0)^2 \to 0$, $f_n^2 \ll z_0^2$, and since $\lambda z_0 = \pi w_0^2$, and $2f_n = l$, we obtain (4).

Since in our case $z_0 = 2.7 \times 10^{-2}$ cm and $f_n = 4.5$ cm, then, neglecting the value of z_0^2 in comparison with f_n^2 in the denominator of expression (18), we reduce it to its final form:

$$P_{\text{out}} = P_{\text{in}}\left(1 + \frac{\beta z_0}{\lambda}\frac{2}{f_n}P_{\text{in}}\right)^{-1}. \tag{20}$$

Hence, we obtain an expression for the coefficient β of two-photon absorption of radiation in this model:

$$\beta = \left(\frac{z_0}{\lambda}\frac{2}{f_n}P_{\text{in}}\right)^{-1}\left(\frac{P_{\text{in}}}{P_{\text{out}}} - 1\right) \tag{21}$$

or, using the above values for z_0, λ, and f_n,

$$\beta = \frac{1}{2.26\times 10^2 P_{\text{in}}}\left(\frac{P_{\text{in}}}{P_{\text{out}}} - 1\right). \tag{22}$$

Let us recalculate the data of Table 1 for the pulse power, taking into account the duration of the exciting radiation pulse of 2.8×10^{-8} s. Then, using expression (22), we calculate the corresponding values of the two-photon absorption coefficient β. The results are presented in Table 3, according to which the average value is $\beta_{\text{av}} = (2.32 \pm 0.30) \times 10^{-8}$ cm W^{-1}.

Table 3.

$P_{\text{in}}/10^4$ W	$P_{\text{out}}/10^4$ W	$\beta/10^{-8}$ cm W^{-1}
1.8	1.6	3
3.6	3.1	2
5.4	4.25	2.2
7.2	5.4	2
8.9	6.0	2.4

5. Conclusions

In our previous work [6], we unfortunately made a misprint in the reduced value of the two-photon absorption coefficient: $\beta \approx (0.5 - 1) \times 10^{-9}$ cm MW^{-1}. Actually, $\beta \approx (0.5 - 1) \times 10^{-9}$ cm W^{-1}. This value was estimated in order of magnitude in the plane-wave approximation using some of the results presented in Fig. 4 from Ref. [6]. The value of obtained with this estimate differs only by two–four times from that given in Table 2.

In Ref. [6], Fig. 4 shows the dependence of the ratio $\Delta e/e_{\text{in}} = (e_{\text{in}} - e_{\text{out}})/e_{\text{in}}$ on e_{in}. Using the results presented in Fig. 2 of this work, we plotted the same dependence (Fig. 3).

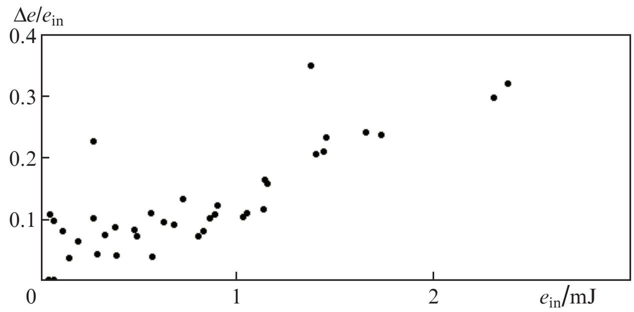

Figure 3. Dependence of $\Delta e/e_{\text{in}}$ on e_{in}, plotted using the experimental points in Fig. 2.

Here the scatter of points significantly exceeds that observed in [6]. A significant difference between these dependences is the range of pump pulse energies. The large scatter of points in Fig. 3, in our opinion, indicates that in plotting the dependence of the difference between two experimentally measured quantities, the smaller the absolute values of the quantities themselves, the greater the role of the relative measurement errors.

Let us plot one more experimental dependence of the type shown in Fig. 3. To do this, we construct the dependences

$$\frac{\Delta I}{I_{\rm in}}(I_{\rm in}) = \frac{I_{\rm in} - I_{\rm out}}{I_{\rm in}} = \frac{\beta l I_{\rm in}}{1 + \beta l I_{\rm in}},$$
$$\frac{\Delta P}{P_{\rm in}}(P_{\rm in}) = \frac{P_{\rm in} - P_{\rm out}}{P_{\rm in}} = \frac{\beta z_0}{\lambda}\frac{2}{f_n}P_{\rm in}\left(1 + \frac{\beta z_0}{\lambda}\frac{2}{f_n}P_{\rm in}\right)^{-1}. \tag{23}$$

When calculating them, we use the data from Tables 2 and 3. It turned out that the values obtained using both formulae coincide within the measurement errors. The calculation results are shown in Fig. 4. The scatter of these data is no longer as large as that in Fig. 3.

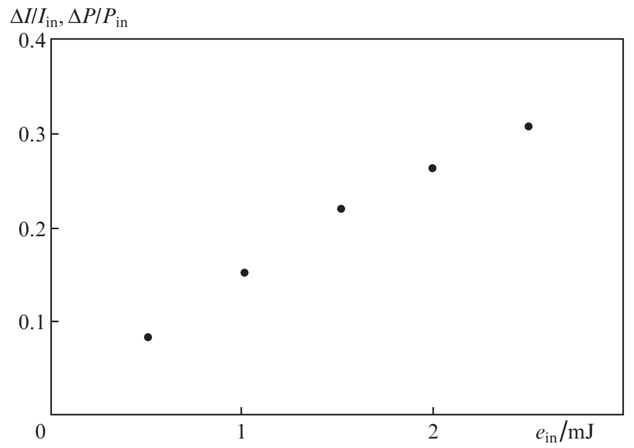

Figure 4. Dependences of the ratios $\Delta I/I_{\rm in}$, $\Delta P/P_{\rm in}$ (23) on the input pulse energy, which can be recalculated into the intensity or power of the input radiation using the data from Tables 2 and 3.

In conclusion, we note once again that to determine some nonlinear coefficients, in particular, the coefficient of two-photon absorption of radiation, the dependence of the energy of laser pulses passing through a nonlinear medium on the energy of incident pulses is usually measured experimentally. In this case, to increase the pump intensity, the exciting radiation is often focused into a nonlinear medium. Then the coefficient of two-photon absorption of radiation is calculated using expressions obtained within the framework of some theoretical models. Experimentally measured parameters of the type shown in Fig. 2 are substituted into these expressions. We have shown that the calculated two-photon absorption coefficients differ by two orders of magnitude when using expressions obtained on the basis of the following theoretical models: the plane-wave approximation, when it is assumed that all absorption occurs at the length of the focal waist, where the wavefronts of the radiation pulses are plane; and the second used approximation when the focusing of a real Gaussian beam with a variable cross section is considered.

References

1. He G.S., Yong K., Zhu J., Prassad P.N. *Phys. Rev. A*, **85**, 043839 (2012).
2. He G.S., Law W., Zhang X., Prassad P.N. *Appl. Phys. Lett.*, **101**, 011110 (2012).
3. He G.S., Law W., Baev A., Lin S., Swihart M.T., Prassad P.N. *J. Chem. Phys.*, **138**, 024202 (2013).
4. Shi I., Wu H., Yan F., Yang J., He Y. *J. Nanopart. Res.*, **18**, 23 (2016).
5. Averyushkin A.S., Bulychev N.A., Efimkov V.F., Erokhin A.I., Kazaryan M.A., Mikhailov S.I., Saraeva I.N., Zubarev I.G. *Laser Phys.*, **27**, 055401 (2017).
6. Gordeev A.A., Efimkov V.F., Zubarev I.G., Mikhailov S.I. *Quantum Electron.*, **50** (11), 1078 (2020) [*Kvantovaya Elektron.*, **50** (11), 1078 (2020)].
7. Matveev N.M. *Metody integrirovaniya obyknovennykh differentsial'nykh uravnenii* (Methods for Integration for Ordinary Differential Equations) (Moscow: Vysshaya Shkola, 1967).