NONLINEAR ABSORPTION OF PICOSECOND LIGHT PULSES IN SEMICONDUCTORS

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Nonlinear absorption of the radiation of a neodymium laser (λ 1.06 or 0.53 μ) with locked modes was observed in transparent semiconductors (CdS_xCdSe_{1-x} CdS, ZnS). The indication was by means of the crystal luminescence produced by two- or three-photon excitation of the carriers. The high peak intensity of the radiation S_0 leads to a noticeable nonlinear absorption accompanied by a characteristic decrease of the brightness I(z) of the luminescence track along the beam propagation direction. A comparison of the experimental and theoretical I(z) dependences in the case of two-photon (ZnS) and three-photon absorption (CdS) has made it possible to determine the moments $\langle |E|^{2n} \rangle$ of the radiation and to estimate the pulse duration and the radiation intensity.

THE purpose of the present communication is to call attention to a new method of investigating ultrashort light pulses; this method uses nonlinear absorption in transparent semiconductors. In our experiments, the radiation of a neodymium laser (λ 1.06 μ) with locked modes, or its harmonic (λ 0.53 μ) was passed respectively through CdS or ZnS crystals (the nonlinear damping in these crystals was apparently due to three- or two-photon interband transitions). A measure of the intensity of the light inside the sample was the brightness of the photoluminescence accompanying the carrier recombination. The law governing the decrease of the brightness of the luminescence track I(z) produced by the laser beam with increasing distance, was determined from photographs taken sideways.

It is easily seen that the I(z) dependence (which deviates from exponential because of the nonlinearity of the medium) contains useful information on the properties of the radiation (or, if the latter are known, on the properties of the medium). The electronic nonlinearity of semiconductors, even in the case of picosecond pulses, can be regarded as non-inertial, so that the intensity of the beam decreases in a transparent medium as a result of n-photon absorption, in accordance with the law

$$S(z,t) = S\left(0, t-\frac{z}{v}\right) \Big/ \left[1+(n-1)\beta_n z S^{n-1}\left(0, t-\frac{z}{v}\right)\right]^{1/(n-1)} \quad (1)$$

where $\beta_n \equiv S^{-n} dS/dz$ is the coefficient of n-photon absorption. We shall assume for simplicity that the luminescence brightness I(z) at the point z is proportional to a number of pairs produced there during the time of the pulse:

$$I_n(z) \sim \int S^n(z,t)\,dt \sim \langle [S^{1-n}+(n-1)\beta_n z]^{n/(1-n)} \rangle, \quad (2)$$

where $S \equiv S(0, t)$ is the envelope of the pulse at the entrance to the nonlinear medium, and the angle brackets denote averaging over the time. We expand (2) in a Taylor series:

$$I_n(z) \sim \sum \binom{n/(1-n)}{k} [(n-1)\beta_n z]^k \langle S^{kn+n-k} \rangle, \quad (3)$$

which converges when $\tilde{z} \equiv (n-1)\beta_n S_0^{n-1} < 1$, where S_0 is the peak intensity of the input pulse. Thus, the law governing the attenuation of the luminescence along the track, for a known β_n, is determined by the higher moments $\langle S^k \rangle$ (k = 2, 3, ...), knowledge of which makes it possible, in principle, to reconstruct the form of the pulse (or the density of the distribution g(S), if S(t) is assumed to be a random ergodic process).

Let us examine the form of the normalized function

$$\tilde{I}_n(\tilde{z}) \equiv \frac{I_n(\tilde{z})}{I_n(0)} = \frac{\langle (f^{1-n}+\tilde{z})^{n/(1-n)} \rangle}{\langle f^n \rangle} \quad (n = 2, 3\ldots) \quad (4)$$

in the case when $S(t) = S_0F)t)$ assumes certain simple analytic forms. Thus, in the case of a rectangular pulse (f = 1) we have

$$\tilde{I}_2 = (1+\tilde{z})^{-2}, \quad (5)$$

$$\tilde{I}_3 = (1+\tilde{z})^{-3/2}. \quad (6)$$

In the case of a Lorentz pulse ($f = [1 + (2t/\tau)^2]^{-1}$) we have

$$\tilde{I}_{2a} = (1+\tilde{z})^{-3/2}. \quad (5a)$$

In our experiments we registered the luminescence brightness integrated over the beam cross section, and therefore the angle brackets should denote in the presented formulas not only longitudinal but also transverse averaging. Putting in (4)

$$f = \frac{e^{-(r/w)^2}}{1+(2t/\tau)^2}, \quad (7)$$

we obtain

$$\tilde{I}_{2b} = -\frac{4}{\tilde{z}^2}\left(\frac{\tilde{z}+2}{\sqrt{\tilde{z}+1}} - 2\right) = 1 - \tilde{z} + 0{,}94\tilde{z}^2\ldots, \quad (5b)$$

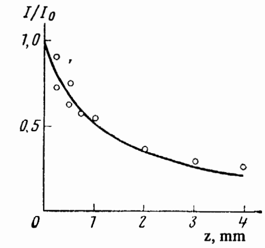
$$\tilde{I}_{3b} = \sum \frac{8(2k+1)!!(4k+3)!!}{(2k+3)(2k)!!(4k+4)!!} \times (-\tilde{z})^k = 1 - 0.66\tilde{z} + 0{,}48\tilde{z}^2. \quad (6b)$$

The second harmonic is of the form $f^1 = f^2$ and in the case of (7) we have

$$\tilde{I}_{2c} = {}^{8}/_{5}\tilde{I}_{2b} - {}^{3}/_{2} = 1 - 1{,}6\tilde{z} + 1{,}5\tilde{z}^{2} \ldots \quad (5c)$$

In the interpretation of the experimental results we used formulas (5c) and (6b). The figure shows a plot of the function (6b) (we used numerical integration to construct it at $\tilde{z} \geq 1$) and the experimental data. The best agreement is obtained from a ratio $\tilde{z}/z = 2\beta_3 S_0^2$, equal to 15 cm^{-1}. Similar experiments on two-photon damping of the second harmonic in ZnS yielded, using the expansion (5c), $\beta_2 S_0 = 4\ cm^{-1}$. Unfortunately, the values of the coefficients β_2, and particularly β_3, are known only for a few substances (see, for example, [1-4]). According to [4], β_2(ZnS; $\lambda\, 0.53\, \mu$) ~ 2 cm/GW, from which it follows that the peak intensity of the harmonic reaches ~2 GW/cm^2. Simultaneous measurement of the pulse energy ($W = 4 \times 10^{-3}$ J:20) and of the beam radius (w = 0.6 mm) has made it possible to estimate with the aid of (5c) the duration of the harmonic pulse, $\tau \sim 2W/\pi^2 w^2 S_0 \sim 5$ psec. An independent estimate of τ, which gave a value of 5 psec, was carried out with the aid of the standard method of colliding opposing beams in the same crystal (the contrast was 2–2.5).

In the case of $\lambda\, 1.06\, \mu$, similar estimates (W = 0.05 (W = 0.05 J:20, w = 0.7 mm, $\tau \sim 3$ psec; collision contrast 2.3–2.5 in a crystal of $CdS_{0.9}CdSe_{0.1}$) gave S_0 ~20 GW/cm^2, so that β_3 (CdS; $\lambda\, 1.06\, \mu$) ~ 0.02 cm^3/GW^2. The data of [3] yield a value of β_3 lower by three orders of magnitude, which seems too low to us (for comparison, we indicate that β_3 (Al_2O_3; $\lambda 0.69\, \mu$) ~ 0.1 cm^3/GW^2. We note that the estimate of β_3 obtained in the present paper gives a level of 3 GW/cm^2 for the three-photon limitation[1] in a crystal 1 cm long.



Experimental dependence of the luminescence intensity of a laser track in CdS on the distance to the input face z (points) and a plot of the function (6b) for $\tilde{z}/z$ = 1.5 (solid curve).

In conclusion, one can express assurance that a study of the nonlinear interaction of picosecond pulses of light with semiconductors is of considerable interest for the investigation of the properties of matter and of radiation.

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