NONLINEAR ABSORPTION AND LIMITATION OF LIGHT INTENSITY IN SEMICONDUCTORS

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The nonlinear absorption of the emission of a ruby laser in CdS, SiC, CdS_xCdSe_{1-x}, ZnSe, and $LiNbO_3$ crystals and of a neodymium laser in GaAs is studied. The two-photon absorption coefficient of these crystals is measured. The contribution to nonlinear absorption from free-carrier absorption and second-harmonic generation (with subsequent absorption) is estimated. The feasilibity of nonlinear absorption for use in light intensity limiters is considered and the observation of the limiting effect is reported for CdS and SiC. The emission is focused in the specimen to reduce the limiting level; a limiting level of 10 kW is obtained in CdS. The limiting power (~2.5 kW) of a GaAs injection semiconductor laser determined by the two-photon absorption of the generated emission is estimated.

LASERS make it possible to measure a number of new parameters of materials; the two-photon absorption coefficient is such a parameter. In addition to spectroscopic applications, the study of multi-photon absorption is of practical interest. Multi-photon processes may affect the power limit of lasers[1-3] and the laser-induced mechanism of damage to transparent materials. Semiconductor lasers with two-photon pumping have been built[4-9]. The two-photon absorption in CdS was recently used to increase the pulse length of a Q-switched laser[10]. A number of papers have been published so far on the measurement of the two-photon absorption coefficient β in the following semiconductors: CdS[11-13], SiC[11], ZnS[14], and CdSe[15], and in some liquids[16].

The present paper reports on the measurement of the coefficient β in a number of crystals, and on the observation of light intensity limitation due to the absorption in semiconductor crystals. The experimental results were used to estimate the power limit of a semiconductor laser. The preliminary results of our experiments were given in[11].

1. LIGHT INTENSITY LIMITATION IN MULTI-PHONON ABSORPTION

We consider the variation of intensity S of a plane wave in an isotropic material with linear absorption coefficient α and N-photon absorption coefficient β (N = 2, 3, ...). We have

$$dS/dz = -\alpha S - \beta S^N. \tag{1}$$

The solution of (1) has the form

$$S(z) = S_0 e^{-\alpha z}\left\{1 + \frac{\beta}{\alpha} S_0^{N-1}[1 - e^{(1-N)\alpha z}]\right\}^{1/(1-N)}. \tag{2}$$

It follows from (2) that the wave intensity does not depend on S_0 when $S_0^{N-1} \gg (\alpha/\beta)[1 - e^{(1-N)\alpha z}]$. The limiting level is determined by the expression

$$S_{max} = e^{-\alpha z}\left[\frac{\beta}{\alpha}(1 - e^{(1-N)\alpha z})\right]^{1/(1-N)}. \tag{3}$$

If the linear absorption is small, so that $\alpha z \ll 1$ (the material is transparent in the ordinary sense), then

$$S_{max} \approx [(N-1)\beta z]^{1/(N-1)}; \tag{4}$$

and in two-photon absorption

$$S_{max} \approx 1/\beta z. \tag{5}$$

According to the experiments with GaAs crystals described below the coefficient $\beta \sim 0.8$ cm/Mw at the wavelength of a neodymium laser ($\lambda = 1.06\ \mu$), i.e., the intensity of emission that passed through a crystal 1 cm long does not exceed 1.25 MW/cm^2.

Consequently a material, whose two-quantum transition absorption is intensive in the range $\omega_1 - \omega_2$, can serve as a light limiter for the frequencies within the range $\omega_1/2 - \omega_2/2$, and the maximum output emission density is $1/\beta z$ for a plane wave.

We should thus expect a reduction in the limiting level due to single-quantum absorption of light by free carriers formed in two-photon transitions, and also due to second harmonic generation in crystals:

$$S_{max} \approx 1/(\beta + \beta_c + \beta_{2\omega})z, \tag{6}$$

where the coefficient β_c is due to free carrier absorption and $\beta_{2\omega}$ to the generation of the second harmonic and its subsequent one-photon absorption.

We evaluate the magnitude of β_c assuming that the carrier lifetime is much shorter than the laser pulse length. In this case the carrier concentration excited in two-photon absorption is of the order of $S(\beta/4\hbar\omega\sigma v)^{1/2}$ and

$$\beta_c \approx \frac{e^3\lambda^2\beta^{1/2}}{2m^{*2}c^3 n\mu}(\hbar\omega\sigma v)^{-1/2},$$

where μ is the mobility, σ is the cross section of direct electron-hole recombination, and v is the mean thermal carrier velocity. For an effective mass m* equal to the electron mass, $\lambda = 0.7\ \mu$, n = 2.5, $\mu = 200$ cm^2/V·sec, $\sigma = 10^{-17}$ cm^2, and $v = 10^7$ cm/sec, the value of $\beta_c \sim 10^{-3}$ cm/MW[1]. We note that this evaluation is fairly approximate. In real semiconductors free carrier absorption can deviate by many orders in either direction.

We can readily show that in crystals opaque at the

[1] The above evalutaion is in good agreement with Zubarev's [15] experimental results for CdSe.

frequency of 2ω

$$\beta_{2\omega} \sim \frac{(4\pi)^5\chi_{2\omega}}{cn_\omega^2 n_{2\omega}\lambda^2}\frac{\alpha_{2\omega}/2}{(\alpha_{2\omega}/2)^2+\Delta^2}$$

where $\chi_{2\omega}$ is the quadratic polarizability, n_ω and $n_{2\omega}$ are refraction indices at the frequencies of ω and 2ω, $\Delta = 4\pi(n_{2\omega} - n_\omega)/\lambda$, and λ is the wavelength of incident radiation. For the CdS crystal the coefficient $\beta_{2\omega}$ ~ 1 cm/GW (for $\chi_{2\omega} = 10^{-7}$ abs. units, $n_\omega^2 n_{2\omega} \sim 10$, $\alpha_{2\omega} \sim \Delta \sim 10^4$ cm^{-1}, and $\lambda = 0.7$ μ) which is by an order lower than the value of β observed by us.

2. MEASUREMENT OF NONLINEAR ABSORPTION COEFFICIENTS

The widths of the forbidden bands of CdS, SiC, CdS_xCdSe_{1-x}, ZnSe, and GaAs semiconductor crystals satisfy the condition $\hbar\omega < \Delta < 2\hbar\omega$ that is necessary to observe two-photon interband transitions in which $\hbar\omega = 1.78$ eV (1.17 eV) is the energy of the ruby (or neodymium for GaAs) laser.

In the experiments with CdS and SiC (type 6H) we compared the amplitude and shape of two laser pulses, before and after they passed through the investigated specimen, at various intensities of radiation incident on the crystal. We used a Q-switched ruby laser (20 MW, 30 nsec). The Q-switching was accomplished with a saturable filter (vanadium phthalocyanide in nitrobenzene). The radiation density incident on the specimen varied within the interval from 5 to 100 MW/cm^2. After passing through removable calibrated neutral filters and the specimen, the light beam entered the FÉU-22 photomultiplier where it produced a signal supplied to the S1-11 high-speed oscilloscope; the time constant of the indicating system was 5 nsec. The optical delay line allowed us to observe both pulses with the same photomultiplier, thus improving the accuracy of recording the change in pulse shape.

Figure 1 shows a typical oscilloscopic trace for a CdS crystal 4.5 mm long. The photograph shows the change in pulse shape due to the limiting effect: the first pulse has a flatter shape. A similar effect was observed in the SiC crystal 1.4 mm long.

To determine β the energy of the incident pulse and of the pulse transmitted through the specimen was measured with calibrated calorimeters. At the same time we monitored the pulse shape and the nonuniform laser intensity distribution over the beam cross section. The linear and two-photon absorption coefficients of the crystals were determined from the experimental relationships $S'(S_0')$ using the following expression obtained from (2) for N = 2:

$$S' = \frac{S_0' T^2 e^{-\alpha z}}{1+(\beta/\alpha)S_0' T(1-e^{-\alpha z})}, \qquad (7)$$

where $T = 4n/(n+1)^2$ is the transmission coefficient.

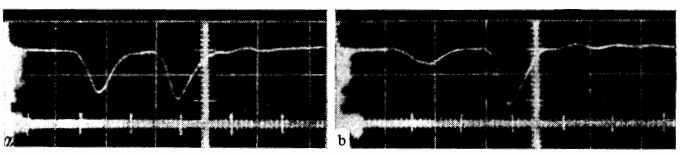

FIG. 1. Oscilloscopic traces of a laser pulse incident upon and transmitted through a CdS crystal for incident radiation intensity of 10 MW/cm^2 (a) and 100 MW/cm^2 (b).

As an example, Fig. 2 shows the results of measuring the intensity ratio $y \equiv S_0'/S'$ at the input and output of the specimen for different intensities of the incident radiation $x \equiv S_0'$ on the $CdS_{0.9}CdSe_{0.1}$ crystal. The straight line was obtained from a statistical treatment of the experimental points according to the least squares method, assuming the relation $y = a + bx$. According to (7) we have $\alpha = z^{-1}\ln(aT^2)$ and $\beta = b\alpha T/(e^{\alpha z} - 1)$.

The results of measurements are given in the table. The error of measurement amounted to $\sim 30\%$ and was mainly due to the error in the measurement of the absolute value of energy with the IMO-1 calorimeter.

We note the fairly high two-photon absorption coefficient of the ZnSe crystal which would seem suitable for a semiconductor laser. Akmanov and others[18] observed the superradiance regime (emission wavelength of 0.46 μ) with two-photon optical pumping of a ZnSe crystal. We also note that the magnitude of β in $LiNbO_3$ may limit the use of this material in nonlinear optics.

3. THE USE OF FOCUSING TO REDUCE THE LIMITING LEVEL

The indicent light can be focused to reduce the limiting level in beams with small divergence. If the divergence of a beam with diameter D is diffraction limited the ratio of the focal region cross section A to its length, $A/d \sim \lambda/3.5n$ does not depend on D and the focal length of the lens. In this case the output power of the limiter[16][2)]

$$P_{max} \approx \frac{A}{\beta d} \approx \frac{\lambda}{3{,}5n\beta}. \qquad (8)$$

We observed the limiting of a ruby laser intensity focused in a CdS crystal. The laser emission was focused in a specimen 4 cm long by a short-focus lens (F = 2 cm). The lens was placed close to the crystal to reduce the radiation density at the surface of the specimen and to protect it from damage. The power and shape of the laser pulse transmitted through the crystal were investigated. The top of the pulse was flat. The power remained practically constant ($P_{max} \sim 10$ kW) as S_0 varied within 1–10 MW/cm^2. The value of P_{max} computed from (8) at $\beta = 0.03$ cm/MW was ~ 30 times lower than the experimental value; this is apparently due to the fact that the beam divergence in a crystal with internal inhomogeneities is much larger than the diffraction-limited value.

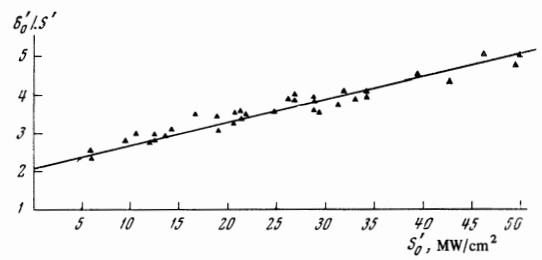

FIG. 2. Relation between intensities of incident S_0' and transmitted S' light for the case of the $CdS_{0.9}CdSe_{0.1}$ crystal.

2) A significant increase in the ratio d/A and consequently a reduction in power P_{max} should be expected from the self-focusing effect.

Linear and two-photon absorption coefficients at the ruby (neodymium for GaAs) laser frequency

Sample	α cm^{-1}	β, cm/MW
CdS *	0.23	0.03
SiC	0.43	0.2
$CdS_{0,9}CdSe_{0,1}$	0.49	0.07
$CdS_{0,8}CdSe_{0,2}$	0.25	0.13
ZnSe	0.32	0.04
GaAs **	4	0.8
$LiNbO_3$ ***	0.08	0.01

*Laser emission is polarized perpendicularly to the optical axis of the crystal (β is maximum here [17]).

**Crystal is of n-type with impurity concentration $\sim 1 \times 10^{16}$ cm^{-3}.

***Direction of laser beam did not coincide with the matching direction for second harmonic generation.

4. CONCLUSION

The above experiments show that transparent semiconductors with a high β value can be used to limit and to effect a temporal and spatial stabilization of the laser beam intensity. The main disadvantage of semiconductor limiters is their low threshold of damage by laser radiation. Liquids are more promising from the viewpoint of the maximum input emission intensity. However the β coefficient seems to be lower in liquids than in solids (for example in pyrene $\beta \approx 0.01$ cm/GW[16]).

Other possibilities of limiting light intensity are noted. For example, Tomlinson[19] observed the limiting effect with a maximum input power of ~ 0.2 MW in gas breakdown. Parametric processes, and stimulated Raman, Brillouin, and Rayleigh scattering should also produce this effect[20].

The measured two-photon absorption coefficients allow us to evaluate the power limit of the generators determined by multi-photon absorption[1]. The expression for the emission density maximum inside the semiconductor laser has the form $S_{max} = \tilde{\alpha}/\beta$, where $\tilde{\alpha}$ is the gain (the value of $\tilde{\alpha}$ accounts for the linear energy losses). For a GaAs injection laser ($\tilde{\alpha} \sim 200$ cm^{-1}) S_{max} is estimated at 250 MW/cm^2 and the power limit of such a laser $P_{max} = 2.5$ kW (p-n junction end face area $\sim 10^{-5}$ cm^2); these figures take into account the value of the two-photon absorption coefficient $\beta \sim 0.8$ cm/MW at the neodymium laser frequency. The accounting for dispersion β apparently decreases somewhat the value of P_{max}. Bunkin and Prokhorov[1] computed the value of β from eq. (41) in a paper by Keldysh[21], in order to evaluate the power limit of a semiconductor injection laser. This equation is not entirely appropriate to the case of two-photon absorption processes since it is derived on the condition that $I \gg \hbar\omega$ (I is the ionization potential) which does not hold for semiconductors.

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