SEMICONDUCTOR QUANTUM GENERATOR WITH TWO-PHOTON OPTICAL EXCITATION

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Generation was obtained and investigated in a GaAs semiconductor excited via two-photon absorption of emission from a neodymium laser. The probability and coefficient of two-photon absorption are calculated. The dependence of the excitation intensity on the depth of the excitation penetration into the semiconductor is determined, as well as the external quantum yield and its dependence on internal losses in the active substance and on the resonator length. The calculated values of the two-photon absorption coefficient, penetration depth, and external coherent quantum yield are in good agreement with the experiments.

THE excitation of semiconductors with powerful light beams makes it possible to obtain appreciable non-equilibrium concentrations of electron-hole pairs. By using laser radiation for this purpose, concentrations corresponding to degeneracy of the non-equilibrium carriers in the bands could be attained, population inversion was produced in semiconductors, and coherent generation of radiation was realized.[1,2]

In experiments of this kind, the semiconductors are excited by absorption of radiation with quantum energy exceeding the width of the forbidden band, accompanied by production of electron-hole pairs. At high incident-beam intensities, however, multiphoton absorption of light in the semiconductors should also become important; this corresponds to production of an electron-hole pair with simultaneous absorption of several photons. The absorption coefficient is proportional in such a process to the intensity of the radiation raised to the $n-1$ power, where n is the number of photons participating in the elementary act. Semiconductors are a convenient medium for the investigation of nonlinear absorption at optical frequencies, since the distance between neighboring bands is, as a rule, of the order of 1 eV, whereas the continuous spectrum of the electronic states of the atoms is separated from the ground state by about 10 eV. This facilitates experimental investigation of the nonlinear processes with a small number of quanta.

In this article we investigate two-photon absorption in a semiconductor and also a laser with two-photon optical excitation, the construction of which was reported earlier.[3] The source of excitation of gallium arsenide samples was a Q-switched neodymium-glass laser in which the photon energy was 1.17 eV (the width of the forbidden band of GaAs at 77° K is 1.51 eV).

An essential feature of the semiconductor laser with two-photon excitation (compared, say, with a p-n junction laser) is the large volume of the medium in which lasing can be produced. This is connected with the fact that the decrease in intensity of the exciting radiation with depth is smoother in two-photon absorption than in one-photon absorption (for equal values of the corresponding absorption coefficients on the surface of the medium). The increased generating volume makes it possible to obtain a much larger power and better coherence of radiation than from p-n junction lasers. In addition, a laser with two-photon excitation yields coherent radiation of higher frequency than the excitation frequency, that is, can raise the frequency of the radiation.

1. TWO-PHOTON EXCITATION OF SEMICONDUCTORS

The luminescence of CdS produced by two-photon excitation of a ruby laser was investigated by Braunstein and Ockman,[4] who also calculated the coefficient of two-photon absorption of light in type-CdS semiconductors. However, formulas (7)–(9) obtained in [4] for two-photon absorption contain several errors, making them unsuitable for the interpretation of our experiments with gallium arsenide. These errors consist in the fact that, in calculating the absorption probability, no account was taken of the intermediate states in the valence band and the interference term was left out of the

compound matrix element without sufficient justification. The theoretical analysis of multiphoton absorption is contained in a paper by Keldysh.[5] The final formula (41) of [5] provides a good approximation for the case when a large number of photons is absorbed in a single act. In our case, when the number of absorbed quanta is equal to two, Keldysh's formula gives for the transition probability a value which is too low compared with experiment. In this connection, it is advantageous to calculate the probability $W^{(2)}$ and the coefficient $K^{(2)}$ of two-photon absorption directly by perturbation theory, with due allowance for the band structure of the gallium arsenide.

The operator of interaction between the electromagnetic field and the semiconductor electrons has the usual form $H = (e/mc)\hat{\mathbf{p}}\mathbf{A}$, where p is the electron momentum operator, $\mathbf{A} = A\boldsymbol{\alpha}$ the vector potential, $\boldsymbol{\alpha}$ the polarization vector, and m the mass of the free electron. The interaction term proportional to $|\mathbf{A}|^2$ can be omitted, for the orthogonality of the wave functions causes its contribution to appear only when more than two photons are absorbed. The transition from the V_3 band to the C band (see Fig. 1) can be disregarded, since according to the data of Sturge,[6] this transition has low probability compared with the transitions from V_1 and V_2.

In second-order perturbation theory we have for the transition probability $W^{(2)}$

$$W^{(2)} = W_1^{(2)} + W_2^{(2)},$$

$$W_i = \frac{2\pi}{\hbar}\iint d^3k_C d^3k_{V_i}\left(\sum_c \frac{H_{Cc}H_{cV_i}}{E_{V_i} + \hbar\omega - E_C - \Delta_i} + \sum_v \frac{H_{V_i v}H_{vC}}{E_v + \hbar\omega - E_C - \Delta_i}\right)^2 \delta(E_C + \Delta_i - E_{V_i} - 2\hbar\omega), \tag{1}$$

where W_i is the probability of transition from the band V_i (i = 1, 2) to the conduction band C (Fig. 1), E_C and E_{V_i} are the energies of electrons in the conduction and in the valence bands, Δ_i is the width of the forbidden band, and ω is the frequency of the electromagnetic wave.

The wave functions can be written in the form

$$\Psi_{C,V} = \frac{1}{(2\pi)^{3/2}} u_{C,V}(\mathbf{r})\exp(i\mathbf{kr}),$$

so that

$$\int \Psi^*_{C,V}\Psi_{C,V}\,d\mathbf{r} = \delta_{CV}\delta(\mathbf{k}_{C,V} - \mathbf{k}'_{C,V}),$$

$$\frac{1}{l^3}\int u^*_{C,V}u_{C,V}\,d\mathbf{r} \equiv \langle u_{C,V}|u_{C,V}\rangle = \delta_{CV}. \tag{2}$$

In the last expression the integration is over the unit-cell volume $l^3 = V/M$, where V is the volume of the crystal and M the number of unit cells.

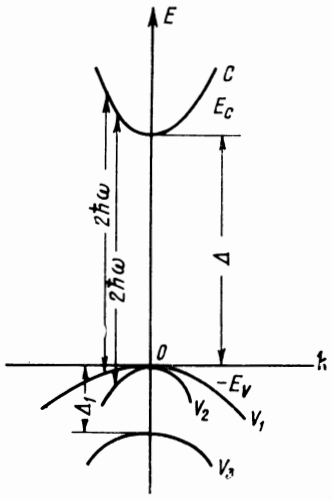

FIG. 1. Band structure of GaAs. C – conduction band, V_1, V_2, V_3 – valence bands, Δ – width of forbidden band. The arrows denote the transitions of electrons from the valence band to the conduction band, accompanied by absorption of two quanta of pump light. The energy is reckoned from the "ceiling" of the valence bands V_1 and V_2.

Using the relations (see [7, 8]

$$\langle u_C(\mathbf{k})|\hat{\mathbf{p}}|u_C(\mathbf{k})\rangle = \frac{m}{\hbar}\frac{\partial}{\partial k}E_C(\mathbf{k}) - \hbar\mathbf{k},$$

$$\langle u_C(\mathbf{k})|\hat{\mathbf{p}}|u_V(\mathbf{k})\rangle \approx \langle u_C(0)|\hat{\mathbf{p}}|u_V(0)\rangle + O(k/k_0) = p_{CV} + O(k/k_0), \tag{3}$$

and assuming the photon momentum to be small compared with the electron momentum, which in turn is small compared with the reciprocal-lattice vector $\mathbf{k}_0$, we obtain for the matrix element of the interaction operator the expressions

$$H_{CV} = \frac{eA}{mc}(\hat{\mathbf{p}}\boldsymbol{\alpha})_{CV}\,\delta(\mathbf{k}_V - \mathbf{k}_C),$$

$$H_{Cc} = \frac{eA}{m_C c}\hbar\mathbf{k}_C\boldsymbol{\alpha}\,\delta(\mathbf{k}_C - \mathbf{k}_c'),$$

$$H_{Vv} = \frac{eA}{m_V c}\hbar\mathbf{k}_V\boldsymbol{\alpha}\,\delta(\mathbf{k}_V - \mathbf{k}_v'), \tag{4}$$

where m_C and m_V are the effective masses of the electron in the conduction and in the valence bands.

After substituting formulas (4) in (1) and integrating over the momenta $\mathbf{k}_C$ and $\mathbf{k}_{V_i}$, we obtain for the transition probability the final formula

$$w_i^{(2)} = \frac{W_i^{(2)}}{V} = \frac{2^{17/2}\pi e^4}{\varepsilon c^2(\hbar\omega)^6}\frac{|(\boldsymbol{\alpha}\hat{\mathbf{p}})_{CV_i}|^2}{m^2} \times m_{CV_i}^{*1/2}(2\hbar\omega - \Delta_i)^{3/2}l^2, \tag{5}$$

where $m^{*-1}_{CV_i} = m_C^{-1} + m_{V_i}^{-1}$, ϵ is the dielectric constant, and I is the intensity of radiation in the medium, equal to

$$vN\hbar\omega = v\varepsilon E_1^2/8\pi = v\omega^2\varepsilon A_1^2/8\pi c^2$$

(here N is the photon density, $v = c/\epsilon^{1/2}$ the velocity of light in the medium, and E_1 and A_1 the amplitudes of the electric field intensity and of the vector potential in the medium). In the derivation of (5) we also took into account the relations

$$\delta^2(\mathbf{k}) = (2\pi)^{-3} V\delta(\mathbf{k}), \qquad |\alpha|^2 = 1,$$

$$E_C - E_{V_i} = E_C + |E_{V_i}| = \hbar^2k^2/2m^*_{CV_i}.$$

From (5) we obtain the absorption coefficient $K_i^{(2)}$, defined with the relation

$$K_i^{(2)} = -\frac{1}{I}\frac{dI}{dr} = \frac{2\hbar\omega w_i^{(2)}}{I}$$

$$K_i^{(2)} = \frac{2^{19/2}\pi e^4}{\varepsilon c^2(\hbar\omega)^5}\frac{|(\hat{\alpha p})_{CV_i}|^2}{m^2} m^{*1/2}_{CV_i}(2\hbar\omega - \Delta_i)^{3/2} I, \qquad (6)$$

where i = 1, 2. The total absorption coefficient is $K^{(2)} = K_1^{(2)} + K_2^{(2)}$.

The probabilities and absorption coefficients of single-photon and two-photon processes can be represented by means of the general formulas:

$$w_i^{(n)} = \frac{(8\pi)^n e^{2n}\Xi^2 m^{-2}|(\hat{\alpha p})_{CV_i}|^2}{\pi\hbar^3 c^n \varepsilon^{n/2}(\hbar\omega)^{2n-1}\omega^{2n-1}}$$

$$\times 2m^*_{CV_i}[2m^*_{CV_i}(n\hbar\omega - \Delta_i)]^{n-1/2} I^n \qquad (7)$$

$$K_i^{(n)} = w_i^{(n)} n\hbar\omega / I, \qquad (8)$$

where

$$n = 1, 2, \qquad \Xi = \sum_{j=0}^{n-1} \frac{1}{m_C^{n-1-j} m_{V_i}^{\ j}}$$

The dependence of the intensity I of the irradiation of the depth of penetration x into the medium can be readily determined from the equation

$$dI/dx = -2\hbar\omega w^{(2)} - \varkappa I = 2\hbar\omega a I^2 - \varkappa I, \qquad (9)$$

where $w^{(2)} = aI^2$ is the two-photon absorption probability defined by formula (5); κ is the single-photon absorption coefficient, which takes into account the linear losses in the medium (the absorption of the radiation by free carriers, defects, etc.); a is the factor preceding I^2 in formula (5). Integrating (9) we obtain

$$I = \frac{I_0 \exp(-\varkappa x)}{1 + (2\hbar\omega a I_0/\varkappa)[1 - \exp(-\varkappa x)]} \qquad (10)$$

where I_0 is the initial intensity of the exciting radiation of the medium.[1)]

When $\kappa x \ll 1$ expression (10) takes the form

$$I(x) = I_0/(2\hbar\omega a I_0 x + 1). \qquad (11)$$

We see from (11) that the decrease of the intensity I with increasing depth of penetration x is slower than for single-photon absorption, for which $I = I_0 \exp(-K^{(1)}x)$, if $K^{(1)} = K_0^{(2)} = 2\hbar\omega a I_0$. Here $K^{(1)}$ is the total coefficient of single-photon absorption.

2. EXTERNAL QUANTUM YIELD IN THE LASING MODE

To estimate the external quantum yield we start from the fact that in the lasing mode the excitation rate in excess over threshold is compensated by the stimulated emission in the active medium. Indeed, in the lasing mode, the following relation is always satisfied

$$\gamma = -\frac{1}{2L}\ln r_1 r_2 + \varkappa = \text{const}, \qquad (12)$$

where γ is the gain, r_1 and r_2 are the reflection coefficients of the resonator mirrors, the distance between which is L, and κ is the loss coefficient. According to [9], in the case of direct transition in an undoped semiconductor the gain can be written in the form

$$\gamma(J) = K^{(1)}\frac{1}{4kT}(\mu - \hbar\omega_g)\left[1 + \frac{K^{(1)}J}{4kTB\hbar\omega_g}\right]^{-1}, \qquad (13)$$

where ω_g is the generation frequency; μ is the sum of the Fermi quasilevels of the electrons (μ_e) and of the holes (μ_h); $J/\hbar\omega_g$ is the flux density of the generated photons; B is the derivative of the recombination rate R with respect to the sum of the Fermi quasilevels μ at the point corresponding to the self-excitation threshold μ_{thr}; T is the carrier temperature.

Equating the gain $\gamma(J)$ to the gain at the self-excitation threshold

$$\gamma_{thr} = K^{(1)}(\mu_{thr} - \hbar\omega_g)/4kT,$$

we obtain for the rate of stimulated recombination $\gamma_{thr} H/\hbar\omega_g$

$$\gamma_{thr} J/\hbar\omega_g = R - R_{thr}, \qquad (14)$$

where R and R_{thr} are the recombination rates in

[1)] One must not forget that $I_0 = I_{tot}(1 - r)$, where I_{tot} is the total intensity of the exciting radiation incident on the surface of the medium and r is the reflection coefficient.

the lasing mode and at the self-excitation threshold, equal respectively to the excitation rates $w^{(2)}$ and $w_{thr}^{(2)}$ of the electron-hole pairs.

Thus, the flux of generated photons P, with due allowance for the dependence of the intensity of the exciting radiation on the depth of penetration x, is given by the equation

$$\frac{1}{\hbar\omega_g}\frac{dP}{dx} = S(w^{(2)} - w_{thr}{}^{(2)}) = Sa(I^2 - I_{thr}{}^2), \quad (15)$$

where S is the area of the irradiated surface of the semiconductor (see Fig. 4 below), I_{thr} is the density of the exciting flux at the self-excitation threshold. Solving (15) together with (9), we obtain for the generated power P

$$P = S\frac{\omega_g}{2\omega}\Big(I_0 - I_{thr} - \frac{2\hbar\omega a I_{thr}^2}{\varkappa}\ln\frac{1+\varkappa/2\hbar\omega a I_{thr}}{1+\varkappa/2\hbar\omega a I_0} + \frac{\varkappa}{2\hbar\omega a}\ln\frac{I_0+\varkappa/2\hbar\omega a}{I_{thr}+\varkappa/2\hbar\omega a}\Big). \quad (16)$$

In the case when $2\hbar\omega a I_{thr} \gg \kappa$, formula (16) takes the simpler form

$$P = S\frac{\omega_g}{2\omega}I_0(1 - I_{thr}/I_0)^2. \quad (17)$$

The total generated power leaving the resonator is $P_{1,2} = P_1 + P_2$. Here P_1 and P_2 are the fluxes passing through the mirrors with reflection coefficients r_1 and r_2, which, allowing for the internal losses in a resonator of length L, are equal to

$$P_1 = \frac{1}{2}P\frac{1-\exp(-\varkappa L)}{\varkappa L}\frac{(1-r_1)[1+r_2\exp(-\varkappa L)]}{1-r_1r_2\exp(-2\varkappa L)}$$

$$P_2 = \frac{1}{2}P\frac{1-\exp(-\varkappa L)}{\varkappa L}\frac{(1-r_2)[1+r_1\exp(-\varkappa L)]}{1-r_1r_2\exp(-2\varkappa L)} \quad (18)$$

Formulas (16)–(18) allow us to estimate η –the relative external quantum yield of the generator, that is, the ratio of the total generation power $P_{1,2}$ to the excitation power I_0S. Thus, for example, for $\hbar\omega_g = 1.51$ eV, $\hbar\omega = 1.17$ eV and $r_1 = r_2 = 0.3$, we obtain ($\kappa = 10$ cm^{-1}, L = 0.5 cm):

1) $\eta = 0.82\%$ for $\varkappa L > 1$; $I_0/I_{thr} = 1.4$,
2) $\eta = 5.8\%$ for $\varkappa L \ll 1$; $I_0/I_{thr} = 1.4$,
3) $\eta = 9\%$ for $\varkappa L = 5$; $I_0/I_{thr} \gg 1$,
4) $\eta = 64\%$ for $\varkappa L \ll 1$; $I_0/I_{thr} \gg 1$.

3. EXPERIMENTAL RESULTS

We investigated spontaneous and stimulated recombination radiation in gallium arsenide under two-photon optical excitation. The source of the exciting radiation was a Q-switched neodymium–glass laser with maximum power 15 MW (the light pulse had an energy 0.6 J and a duration 40 nsec). A GaAs crystal of n-type with area 5 × 8 mm and thickness 5 mm, with impurity density 1.7×10^{17} cm^{-3} and mobility 4400 cm^2/V-sec (300° K), was secured to a bulky cold finger placed in liquid nitrogen (Fig. 2). The laser radiation was focused on one face of the sample with the aid of a cylindrical lens. The other two faces, perpendicular to the foregoing, were finished to form a planar resonator. The radiation from one of the faces was photographed through an ISP-51 spectrograph. The radiation from the other faces was aimed directly on photographic film to determine the directivity pattern.

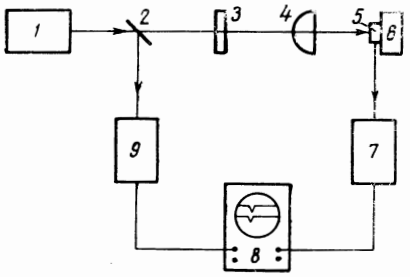

FIG. 2. Block diagram of experimental setup. 1 – laser (neodymium glass); 2 – glass plate diverting part of the energy to the control photomultiplier 9; 3 – calibrated light filter; 4 – cylindrical lens, 5 – gallium-arsenide sample of n-type with carrier density 1.7×10^{17} cm^{-3} and mobility 4450 cm^2/V-sec (at T = 300°C); 6 – cold finger of cryostat; 7 – calibrated photomultiplier for the measurement of the radiation energy (measuring photomultiplier); 8 – two-beam oscilloscope.

Figure 3 shows the emission spectrum of the GaAs crystal. At an excitation flux density I_{tot}

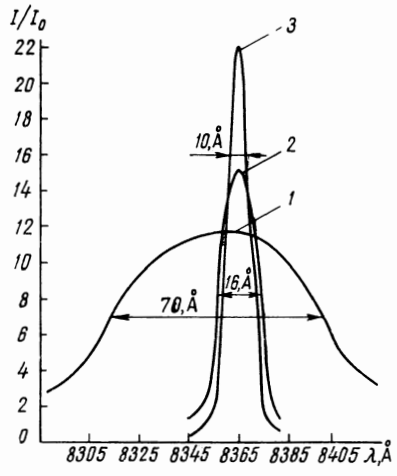

FIG. 3. Spectral emission lines of GaAs at different values of the pump power density. The ordinate scales are different for different curves: the maximum of curve 3 is many times larger than that of curve 1.

= 6 MW/cm^2 the width of the spectral line was 70 Å (curve 1). When the flux density was increased to 8 MW/cm^2 the spectral line width decreases to 16 Å (curve 2) and its spectral intensity increased abruptly. At a flux density of 16 MW/cm^2, generation at a wavelength λ = 8365 Å was observed. The width of the generation spectral line was in this case 10 Å (curve 3). The resolution of the spectrograph (5 Å) did not make it possible to distinguish between the different modes, the distance between them in wavelengths being

$$d\lambda = \frac{\lambda^2}{2L(n - \lambda dn/d\lambda)} = 0.24\text{Å}$$

(here n = 3.5 is the refractive index and L = 0.5 cm is the length of the resonator). The width of the directivity pattern was 5°. To investigate the depth of penetration of the exciting light in the sample, and also the distribution of the intensity of luminescence of the GaAs sample along the x axis, we projected the sample face lying in the zx plane on the spectrograph slit so that the latter coincided with the direction of the x axis (Fig. 4). This made it possible to determine simultaneously from the spectrogram the radiation line width, the distribution of the sample luminescence intensity along x, and the depth of penetration of the exciting radiation into the sample.[2)] The results of processing one of the typical spectrograms obtained by the foregoing method, at an excitation power below threshold, are shown in Fig. 5. The continuous curve is experimental, obtained at I_0 = 7 MW/cm^2. The dashed curve is theoretical, calculated by formulas (6) and (10) with the following values of the quantities contained in them: I_0 = 10 MW/cm^2, $m_C = 0.07m$, $m_{V_1} = 0.5m$, $m_{V_2} = 0.01m$ (m—mass of the free electron), $\epsilon = 11.8$,[10] $\Delta = 1.51$ eV (at T = 77° K), $m^{-2}|(\alpha\hat{p})_{CV}|^2 = 3\Delta/2m_C$,[11] and $\omega = 1.88 \times 10^{15}$ rad/sec.

Thus, formulas (6) and (10) give a satisfactory interpretation of the experimental results.

Figure 5 shows for comparison also the theoretical plots of I(x) for single-photon absorption of the exciting radiation at $K^{(1)} = 80$ cm^{-1} (dash-dot curve).

In those cases when the excitation power exceeds the threshold, the dependence I(x) can not

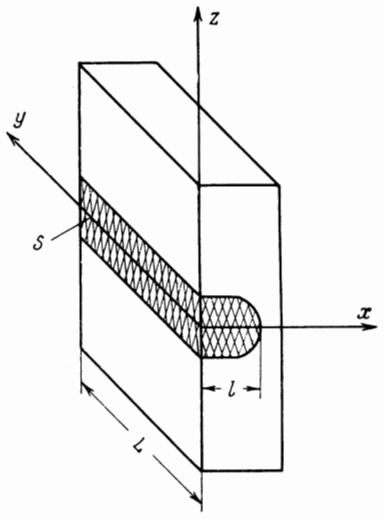

FIG. 4. S – area of irradiated semiconductor surface, l – depth of penetration of exciting radiation, L – distance between the sample faces that form the resonator.

be determined from the generation intensity in the semiconductor, since the latter is closely related to the optical homogeneity of the medium and to the resonator mode structure. The distribution of the intensity along x in the generation mode is shown in Fig. 6. The intensity oscillations are apparently connected with the inhomogeneity of the sample.

We also determined the dependence of the gallium-arsenide radiation energy on the intensity of the exciting radiation, which was regulated by va-

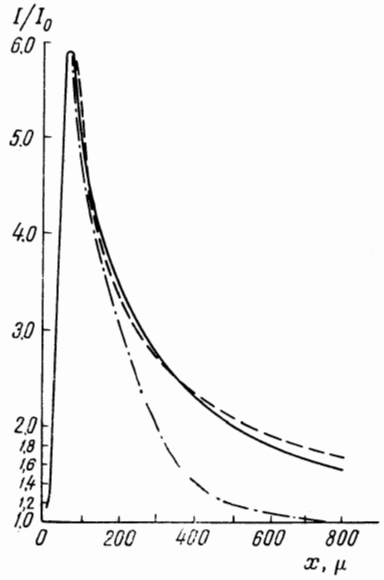

FIG. 5. Distribution of the intensity of recombination radiation at exciting-radiation power below the threshold of self-excitation: dashed – theoretical curve calculated by formula (10) at $K_0^{(2)} = 63$ cm^{-1} ($I_0 = 7$ MW/cm^2) solid curve – experimental relation; dash-dot curve – theoretical curve for single-photon absorption (n = 1).

[2)]Owing to diffusion of the nonequilibrium carriers, the luminescence intensity at each point depends on the degree of excitation of the neighboring regions of the sample. However, if the depth of penetration of the exciting external radiation is much larger than the diffusion length (as is the case for two-photon excitation), the diffusion can be neglected.

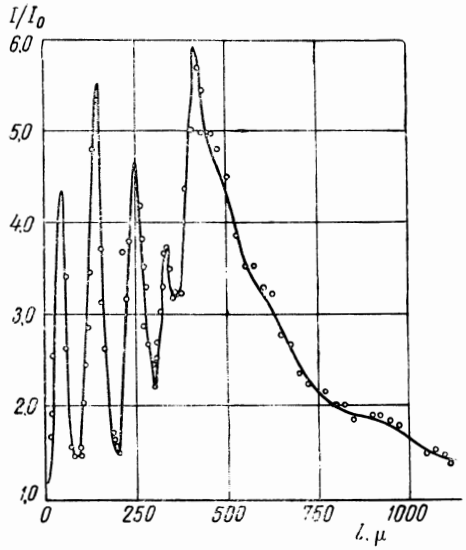

FIG. 6. Distribution of luminescence intensity through the depth of the sample during generation.

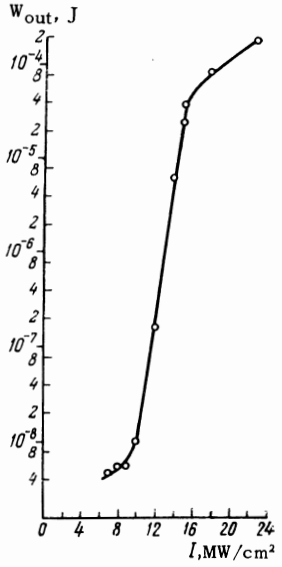

FIG. 7. Dependence of the energy W of the generated light pulse on the density of the exciting light flux I. The energy of the exciting light pulse is constant at 0.07 J.

rying the distance between the cylindrical lens and the sample. Part of the exciting-radiation pulse energy was diverted to a control photomultiplier. The radiation from the sample was guided to a second photomultiplier, the energy of which was calibrated beforehand at 8281 Å. The photomultiplier was calibrated against the Stokes component of the stimulated Raman scattering of a ruby laser in liquid nitrogen.[11] The signals from the outputs of both photomultipliers were fed to a two-beam oscilloscope. The results of the processing of the oscillograms are shown in Fig. 7. As can be seen from this figure, the process of soft excitation has a clearly pronounced threshold character: when the flux density of the exciting radiation changes from 10 to 16 MW/cm^2 (start of generation) the output energy increases by three orders of magnitude. The energy in the exciting-radiation pulse penetrating into the semiconductor was constant and amounted to 0.07 J.

It follows from Fig. 7 that the relative external quantum yield of a semiconductor laser in one of the two directions in which the radiation is emitted during generation, for example, along the y axis (Fig. 4), was 0.25%. The total external quantum yield, with account of the two propagation directions of the generated radiation, was 0.5%, that is, of the same order as the theoretical value 0.82%, as determined by formulas (16) and (18) at $I_0/I_{thr} = 1.4$, $\kappa = 10\ cm^{-1}$, $L = 0.5$ cm, and $r_1 = r_2 = 0.3$.

[1] N. G. Basov, A. Z. Grasyuk, and V. Katulin, DAN SSSR **161**, 1306 (1965), Soviet Phys. Doklady **10**, 343 (1965).

[2] R. J. Phelan and R. H. Rediker, Appl. Phys. Lett. **6**, 70 (1965).

[3] N. G. Basov, A. Z. Grasyuk, I. G. Zubarev, and V. A. Katulin, JETP Letters **1**, No. 4, 29 (1965), transl. **1**, 118 (1965).

[4] R. Braunstein and N. Ockman, Phys. Rev. **134**, 499 (1964).

[5] L. V. Keldysh, JETP **47**, 1945 (1964), Soviet Phys. JETP **20**, 1307 (1965).

[6] M. D. Sturge, Phys. Rev. **127**, 768 (1962).

[7] C. Kittel, Introduction to Solid State Physics, Wiley, 2d ed. (Russ. Transl., Fizmatgiz, 1963).

[8] A. I. Ansel'm, Vvedenie v teoriyu poluprovodnikov (Introduction to Semiconductor Theory), Fizmatgiz, 1962.

[9] O. N. Krokhin, FTT **7**, 2612 (1965), Soviet Phys. Solid State **7**, 2114 (1966).

[10] R. Philipp and H. Ehrenreich, Phys. Rev. **129**, 1550 (1963).

[11] L. V. Keldysh, JETP **45**, 364 (1963), Soviet Phys. JETP **18**, 253 (1964).

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