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THE EFFECT OF STIMULATED FREE-CARRIER ABSORPTION ON TWO-PHOTON PHOTOCONDUCTIVY IN SEMICONDUCTORS

A. M. DANISHEVSKIĬ, A. A. PATRIN, S. M. RYVKIN, and I. D. YAROSHETSKIĬ

A. F. Ioffe Physico-technical Institute, U.S.S.R. Academy of Sciences

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The problem of two-photon volume excitation of a semiconductor is considered taking stimulated absorption of light by nonequilibrium holes into account. It is shown that such an absorption causes a significant attenuation of light intensity with increasing depth of penetration of the crystal and, as a result, that the average concentration of nonequilibrium carriers due to two-photon absorption is no longer a quadratic function of intensity at high excitation levels. According to an experiment in which InSb was excited (dark electron concentration $n = 4 \times 10^{14}$ cm^{-3}) at 90°K by a Q-switched CO_2 laser the observed dependences are in good agreement with theoretical computations. The experimental data were used to compute the value of two-photon absorption cross-section in InSb that is close to the theoretical value obtained by a second-order perturbation theory of a two-band model.

THE investigation of two-photon absorption and the related photoconductivity has recently evoked considerable interest due to the study of the laws governing these phenomena and to their application for high-intensity volume generation of carriers in semiconductors.[1-5] Since the two-photon absorption coefficient is usually small, both the equilibrium and nonequilibrium free-carrier absorption can prove significant in two-quantum excitation.

As we know the cross section of absorption by free holes can be much larger with long wavelengths in many semiconductors, and particularly in $A_{III}B_V$ type compounds, than the electron absorption cross section.[1)] This is due to the presence of transitions between subbands v_1 and v_2 of the valence band.[7] Consequently light quanta with relatively low energy capable of causing two-photon conductivity should be appreciably absorbed by free holes in semiconductors with a narrow forbidden band.

In the present paper we consider the problem of two-photon volume excitation of a semiconductor taking into account stimulated absorption of light by nonequilibrium holes. We show below that such an absorption leads to a significant attenuation of light intensity with increasing depth of penetration of the crystal and that, as a result, the average concentration of nonequilibrium carriers generated by two-photon absorption ceases to be a quadratic function of intensity at high excitation levels.

In this case the variation of light intensity in the specimen can be written as follows:[2)]

$$-dI = k_1 I(x)\,dx + k_2 I(x)\,dx + k_p I(x)\,dx, \qquad (1)$$

where

$$k_2 = WI(x)\sqrt{\varepsilon}/c, \qquad (2)$$
$$k_p = -qWI(x)\sqrt{\varepsilon}\tau/c. \qquad (3)$$

The third term in (1) determines absorption due to the generated concentration of nonequilibrium holes.

Here k_2 and W are the coefficient and cross section of two-photon absorption, k_p and q are the coefficient and cross section of absorption by free holes that were created as a result of two-photon absorption, k_1 is a coefficient of absorption by crystal defects, inhomogeneities, etc., ϵ is dielectric permittivity, τ is the lifetime of nonequilibrium carriers, and c is the velocity of light in vacuum.

After suitable transformations (1) is reduced to a

[1)]For example, in InSb at $\lambda = 10\mu$ the free hole absorption cross section is 230 times larger than electron absorption cross section and reaches the value of 3×10^{-15} cm^{-2} [6].

[2)]Absorption by free electrons and equilibrium holes is neglected here.

transcendental equation that is not convenient for analysis. However, in the most interesting case of high concentration of nonequilibrium holes, beginning with sufficiently high light intensities when

$$q\tau I(x) \gg 1, \tag{4}$$

we can neglect the first and second terms in (1) as compared to the third. Then

$$-dI = \frac{qW\sqrt{\varepsilon}\tau}{c} I^3(x)\,dx, \tag{5}$$

and hence

$$I^2(x) = I_0^2 \Big/ \left[1 + \frac{2qW\tau\sqrt{\varepsilon} I_0^2}{c} x \right], \tag{6}$$

where I_0 is the light intensity at the surface of the specimen.

If the electron mobility μ is substantially greater than hole mobility, photoconductivity $\Delta\sigma$ is determined only by the electron component; in particular this is true of narrow band semiconductors of the $A_{III}B_V$ type and has the following form

$$\Delta\sigma = e\mu \int_0^d \Delta n(x)\,dx = \frac{e\mu\tau\sqrt{\varepsilon}W}{c} \int_0^d I^2(x)\,dx, \tag{7}$$

where $\Delta n(x)$ is the concentration of nonequilibrium electrons at a depth x, and d is the specimen thickness at a normal incidence of light. Substituting (6) into (7) and integrating we obtain

$$\Delta\sigma = \frac{e\mu\tau\sqrt{\varepsilon}WI_0^2}{c} \frac{\ln(1+\beta d)}{\beta d} \tag{8}$$

where

$$\beta = \frac{2q\tau\sqrt{\varepsilon}W}{c} I_0^2. \tag{8a}$$

Given moderate light intensities when the relations $\beta d < 1$ and $\ln(1 + \beta d) \approx \beta d$ hold along with (4) it follows from (8) that

$$\Delta\sigma = \Delta\sigma^{(2)} = \frac{e\mu\tau\sqrt{\varepsilon}W}{c} I_0^2. \tag{9}$$

Consequently when absorption by nonequilibrium holes does not yet cause a significant attenuation of light intensity with increasing penetration of the specimen, but the absorption coefficient due to this mechanism does grow stronger than that due to two-quantum excitation losses, the photoconductivity varies with intensity according to the quadratic law as in the case of the ordinary two-photon photoconductivity in the absence of absorption by nonequilibrium carriers.

Taking (9) into account, the photoconductivity can be written as follows

$$\Delta\sigma = \Delta\sigma^{(2)} \frac{\ln(1+\beta d)}{\beta d}. \tag{10}$$

According to (10), with further increase of light intensity, when βd exceeds unity and accordingly

$$\frac{\ln(1+\beta d)}{\beta d} < 1, \tag{10a}$$

the observed photoconductivity $\Delta\sigma$ is always less than the corresponding quantity $\Delta\sigma^{(2)}$ due to light attenuation by absorption by nonequilibrium carriers; in this case the carriers are generated by the same light.

EXPERIMENTAL RESULTS AND DISCUSSION

The experiment was performed on n-type InSb specimens with a dark concentration of free electrons $n = 4 \times 10^{14}$ cm^{-3} at 80°K and mobility $\mu = 3.4 \times 10^5$ $cm^2/v \cdot sec$. Typical dimensions of the specimens were 1.1 × 1.0 × 0.7 mm. A CO_2 laser served as the light source ($\lambda = 10.6\ \mu$). The laser was suitable for Q-switched operation with a pulse length $t_p = 3 \times 10^{-7}$ sec and pulse repetition frequency of 300 Hz, a pulsed pumping Q-switched operation (frequency of 10 Hz), and pulsed pumping free running operation[8] with $t_p = 6-10\ \mu sec$ and frequency of 1–25 Hz. The output pulse power in Q-switched operation reached 3 kW. The pulse amplitude and shape was monitored by a Ge:Hg pickup at 78°K. Figure 1 shows a diagram of the experimental setup.

We measured the function $\Delta n(I_0)$ for unfocused laser beam 6 mm in diameter (Fig. 2) and a total uniform illumination of the specimen and for the case when the laser beam was focused in a 1 mm^2 spot (Fig. 3) to increase intensity. The analysis of experimental data was

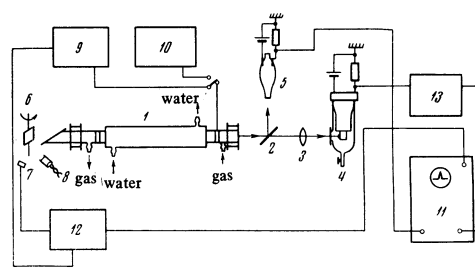

FIG. 1. Diagram of the experimental setup. 1—discharge tube with electrodes and alignment heads; 2—NaCl beam splitter; 3—BaF_2 lens; 4—nitrogen cryostat with NaCl window; 5—Ge pickup; 6—Q-switch assempy; 7—photodiode; 8—illuminator; 9—pulsed power supply; 10—15 kV DC voltage source; 11—two-beam broadband oscilloscope; 12—synchronizer; 13—broadband amplifier.

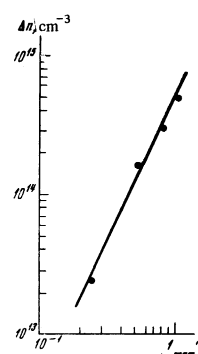

FIG. 2

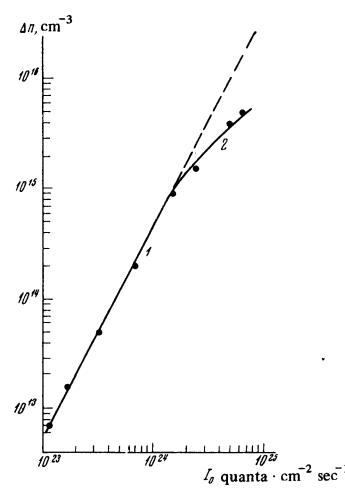

FIG. 3

FIG. 2. Nonequilibrium electron concentration as a function of excitation intensity for unfocused beam. Points designate experimental data, solid line represents the quadratic function of $\Delta n(I_0)$. Here $I_0^{max} = 1 \times 10^{24}$ $quanta/cm^2 \cdot sec$.

FIG. 3. Nonequilibrium carrier concentration in focused beam as a function of excitation intensity. Points designate experimental data, dashed line is the extension of the quadratic dependence.

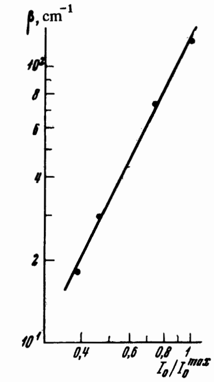

FIG. 4. Parameter β as a function of relative intensity of incident light. Points are plotted from sector 2 of the curve in Fig. 3. The solid straight line represents the theoretical function (8a). Here $I_0^{max} = 6 \times 10^{24}$ quanta/cm² · sec.

performed with a rigorous allowance for contact resistance and dark portions of the specimen. All the measurements were performed at $T = 90°K$.

As we see relatively low intensities of the incident light I_0 (Fig. 2 and the initial region of Fig. 3) result in a quadratic dependence of nonequilibrium electron concentration on light intensity. We observed, however, as expected, a departure from the quadratic dependence with high excitation levels.

In order to verify the fact that the observed effect is really due to the above mechanism we proceed as follows: equating $\Delta\sigma^{(2)}$ with values obtained by extrapolation from the quadratic dependence region to the case of high intensities (dashed line in Fig. 3) and setting up the ratio $\Delta\sigma_{ext}/\Delta\sigma^{(2)}$ for these intensities we find the value of β from the expression

$$\frac{\Delta\sigma_{ext}}{\Delta\sigma^{(2)}} = \frac{\ln(1+\beta d)}{\beta d}. \qquad (10b)$$

The thus obtained values of β as functions of the relative intensities of the incident light are given in Fig. 4. We see that the quantity β is indeed a quadratic function of I_0 in accordance with (8a).

Next, it follows from (8a) that the two-photon absorption cross section W is expressed by the β parameter in the following manner:

$$W = \beta c / 2q\tau\sqrt{\varepsilon} I_0^2. \qquad (8b)$$

But the same quantity can be found directly from the initial quadratic region of the function $\Delta\sigma(I_0)$ according to the formula

$$W = c\Delta n / \sqrt{\varepsilon}\tau I_0^2. \qquad (11)$$

Their ratio W''/W' for two light intensities, I_0' in the first region and I_0'' in the second region respectively (Fig. 3), equals[3)]

$$\frac{W''}{W'} = \frac{\beta}{2q\Delta n}\left(\frac{I_0'}{I_0''}\right). \qquad (12)$$

If the above model is valid then the ratio W''/W' should be identically equal to unity. Substituting I_0' and I_0'' and the experimentally determined values of Δn and β into (11) and using the value $q = 3.2 \times 10^{-15}$ cm² [6] we indeed find that $W''/W' = 1$ with an accuracy to 1%.[4)]

[3)]In the specimens used τ was practically independent of intensity and amounted to $\sim 10^{-7}$ sec.

[4)]Such a high accuracy in the determination of W''/W' is possible because only relative intensity figures in (12).

The adequate agreement of experimental results with the conclusions of the proposed model seems to verify the validity of the above approximations.

We note that the deviation of $\Delta\sigma(I_0)$ from the quadratic law can in principle be due to the presence of other mechanisms, such as the reduction of two-photon absorption cross section caused by the nonequilibrium Burshtein effect, or a reduction in nonequilibrium electron mobility in scattering by nonequilibrium holes. According to numerical data however both effects are significant only for $\Delta n > 5 \times 10^{16}$ cm⁻³ which is appreciably higher than the maximum concentrations achievable in our experiment.

In conclusion we consider the important problem of the absolute magnitude of two-photon absorption cross section in InSb. According to the expression obtained in [9] for two-photon absorption cross section in the two-band model approximation

$$W = \frac{2^{13/2}\pi e^4 m_{cv}\Delta_i(2\hbar\omega - \Delta_i)}{3(\hbar\omega)^4 \varepsilon^{3/2} c m_c}, \qquad (13)$$

where $m_{cv} = m_c m_v/(m_c + m_v)$, Δ_i is the width of the forbidden band, m_c and m_v are the effective masses of electron and hole, and $\hbar\omega$ is the quantum energy.

In writing (13) it was assumed that for the $A_{III}B_V$ type compounds and polarized laser light used in this work

$$P_{cv} / m_0^2 = \Delta_i / 2m_c, \qquad (14)$$

where P_{cv} is the matrix element of interband transition.

Substituting the values of $\Delta_i = 0.228$ eV, $\hbar\omega = 0.117$ eV, $m_{cv} = 1.5 \times 10^{-2} m_0$ and $\epsilon = 16$ into (13) we obtain $W = 1.5 \times 10^{-16}$ cm². The two-photon absorption cross section determined from experimental data according to (9) yielded the value of $W = (8 \pm 2) \times 10^{-17}$ cm² which is obviously in good agreement with theory.

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