NONLINEAR ABSORPTION AND SUSCEPTIBILITY OF THE FIFTH ORDER IN InSb AT THE WAVELENGTH OF 10.6μm

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It is shown, that nonlinear absorption of laser radiation at the wavelength of 10.6 μm in band-gap semiconductor InSb is caused by two-photon absorption and the constant of nonlinear absorption is measured in samples n-type with a different degree alloying. It is established, that the basic mechanism of nonlinearity responsible for reflection at degenerate four-wave mixing is caused by free electron generation at two-photon absorption. Growth of efficiency at four-wave mixing in band-gap semiconductors is limited to nonlinear absorption of interacting waves.

One of the major problems of applied nonlinear optics is search of materials with possible great values of nonlinear susceptibilities. In this respect semiconductors as have shown experiments, are one of the most perspective media [1]. Big nonlinearity of the last leaves that fact what exactly semiconductors with their rather small band gap E_g are characterized by low enough internal fields $E_0 = \frac{mE_g^3}{e^2\hbar^2}$ determining bond forces of optical electron. Therefore, at all too high laser fields already should provide the big contribution to a susceptibility of electronic nonlinear polarization.

Studying of nonlinear susceptibilities is- the central question of nonlinear spectroscopy. The effects caused by a nonlinear susceptibility, underlie such methods of nonlinear spectroscopy as two-photon spectroscopy, spectroscopy of saturation, and also allow, will solve such important practical problem - as correction of phase distortions by a method of four-wave mixing [2,3].

For reception of high efficiency of reflection of radiation on length of a wave of the CO_2-laser at four-wave mixing significant interest, represent band-gap semiconductors [2].

In the given work at temperature T=300 K samples InSb of n-type with concentration donor impurity of~10^{14}-10^{16}cm^{-3}, thickness ℓ=0.5 mm were investigated.

For all scopes of semiconductors, it is important to know a limit of their serviceability on intensity of laser radiation. A surface breakdown threshold of a material usually imposes this limit. Measurement of a threshold of breakdown on a surface of all samples investigated in our work has shown that this size lies in a range 3÷4·10^7Vt/cm^2. As show measurements transmission of samples InSb, as against wide-gap semiconductors (for example, Ge) still at intensities of radiations are lower than a threshold of breakdown on a surface in them appreciable reduction transmission is observed (fig. 1).

This reduction transmission is convertible, is shown at intensity of radiation I ≳10^5Vt/cm^2, and caused by processes of nonlinear absorption of radiation in researched semiconductors. The compared sizes of quantum of radiation of the CO_2-laser (ħω ≃0,117 eV) with InSb band gap (~0,17eV at room temperature) allows to assume, that nonlinear absorption in them is caused by two-photon process. For definition from the data on transmission constants of absorption, we consider a problem about dependence transmission from intensity in view of linear and nonlinear absorption. In a stationary case change of intensity at passage of light to semiconductors at presence of effects of two-photon absorption can be written down as follows

$$-dI = \alpha I(x)dx + \beta I^2(x)dx + \gamma I^3(x)dx \qquad (1)$$

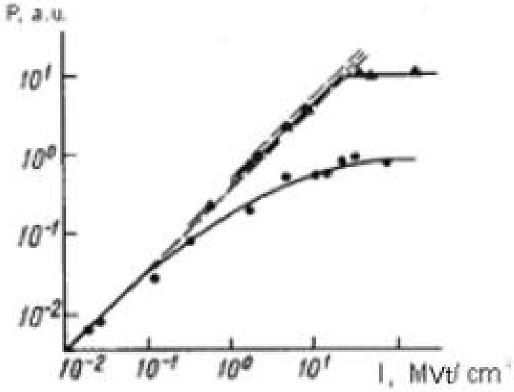

Fig. 1. Dependence of the past pulse power from intensity of a falling pulse. (Ge - triangles, InSb - dark points.)

Here α is the linear optical absorption coefficient, β is the two-photon absorption coefficient, γ is the coefficient of absorption free hole, arising as a result of two-photon absorption. γ it is connected to the β following parity

$$\frac{\gamma}{\beta} = \frac{q\tau}{2\hbar\omega}, \qquad (2)$$

where q is section of absorption free hole, τ is time of a life of nonequilibrium carriers. In compounds of type A^{III}B^{IV} the section of absorption free hole appears so big [4] that already at moderate intensities laser radiation by the second member in the right part (1) it is possible to neglect. In this case, expression for transmission in view of two-photon absorption depending on intensity of falling radiation I_0 becomes samples

$$T^{(2)} = (1-r)\sqrt{\frac{\alpha e^{-2\alpha\ell}}{\alpha + \gamma I_0^2(1-e^{-2\alpha\ell})}}, \qquad (3)$$

where r is coefficient of Fresnel reflection from a sample surface, ℓ is sample length.

Comparison of results of experimental researches (fig. 2) with the data of calculation by formula (3) allows defining sizes γ directly. In view of disorder of experimental data for InSb it is received $\gamma = 500 \pm 200 \frac{cm^3}{MVt^2}$ also this value is constant for all samples.

The process of phase conjugation via four-wave mixing involves the incidence of three input waves (E_1,E_2, E_3) onto a medium, with the nonlinear generation of an output field (E_4) whose amplitude is proportional to the complex conjugate of one of the input fields (fig.3). The mechanism of occurrence of the inverted wave in such circuit most simply to explain, proceeding from holographic interpretation of optical phase conjugation [5]. Let on the nonlinear medium the any wave $E_3(r)$ that is required to be turned and a pump wave $E_1(r)$ from a constant on cross-section section amplitude falls. If waves $E_1(r)$ and $E_3(r)$ are coherent then they write down in nonlinear medium interference disturbance of dielectric permeability (hologram). If this hologram to illuminate from the opposite side with a pump wave $E_2(r)$, is exact a passer in relation to a pump wave $E_1(r)$, such, that $E_2(r) = E_1^*(r)$ the hologram will restore the inverted wave

$$E_4(r)=const\cdot E_1(r)E_2(r)E_3^*(r)=const\cdot|E_1|^2 E_3^*(r) \quad (4)$$

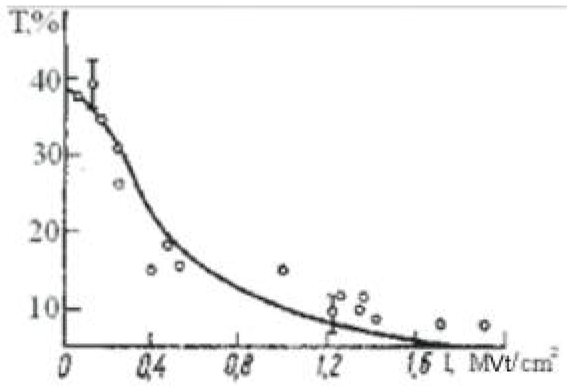

Fig. 2. Dependence transmission *T* from intensity falling radiations in InSb.

In the volumetric nonlinear medium record interference pictures waves $E_2(r)$ and $E_3(r)$ and reading by its wave $E_1(r)$ from restoration of the same wave $E_4(r)$ is possible. In the traditional experimental geometry, the second pump wave is created by reflection of the first by a flat mirror back.

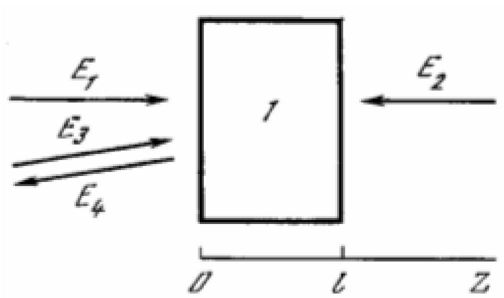

Fig.3. Backward-going nonlinear optical phase conjugation via degenerated four-wave mixing

The measured dependence of efficiency of reflection at four-wave mixing from intensity of first pump wave $R\ (I_1)$ is resulted on fig.4. Data processing in the field of $I_1 \approx$10-20kVt/cm^2 on a method of the least squares has shown, that in logarithmic scale they are described by a straight line with an inclination corresponding to dependence $R\sim I_1^4$ with factor of correlation 0,85. At I_1>20kVt/cm^2 dependence R(I_1) starts to deviate already essentially dependence of kind R ~ I_1^4 (dotted line). Such behavior of an experimental curve at degenerate four-wave mixing in InSb on 10,6 μm at coherent probe and pump waves specifies that the effective nonlinear susceptibility responsible for phase conjugation , is caused by generation free electrons. As this process goes by two-photon absorption it is possible to draw a conclusion that in observable the dominating contribution is brought with a nonlinear susceptibility of the fifth order $\chi^{(5)}$. The continuous curve on fig.4 puts dependence $R(I_1)$ in view of nonlinear absorption under the formula [6]

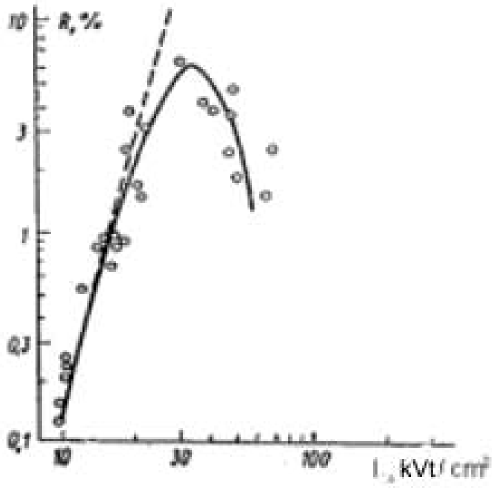

Fig. 4. Dependence of efficiency of reflection at four-wave mixing from intensity

$$R = \left[(1-r)^3 \frac{2\pi\omega}{cn} \chi^{(5)} E_1^4 \frac{e^{-\alpha_n \ell}(1-e^{-\alpha_n \ell})}{\alpha_n} \right]^2, \quad (5)$$

where $\alpha_n = \alpha + \gamma_3 I_\Sigma^2$, $I_\Sigma = \sum_{i=1}^{4} a_i |E_i|^2$; ω is frequency of laser radiation; c is speed of light in vacuum; n = 3.95 - a linear refractive index ; α=5cm^{-1} and $\gamma = 500 \pm 200 \frac{cm^3}{MVt^2}$ are coefficients of linear and nonlinear absorption in InSb; r is coefficient of Fresnel reflection from a surface of a sample; a_I are the factors which are taking into account the contribution of each interacting waves in nonlinear absorption.

In area intensities I_1 from 10 up to 30 - 40 kVt/cm^2, i.e. down to a maximum of dependence $R(I_1)$, the settlement curve well describes results of experiment. Substituting experimentally measured R and I_1 at I_1<40kVt/cm^2 in the formula (5), we get a nonlinear susceptibility of the fifth order in InSb: $\chi^{(5)} \approx 8\cdot 10^{-8}$ esu.

Proceeding from the assumption, that at $I_1 \lesssim$40kVt/cm^2 the basic mechanism of the nonlinearity responsible for phase conjugation via four-wave mixing is caused by generation free electrons we shall estimate $\chi^{(5)}$. It is known [4], that at generation free electrons in semiconductors the nonlinear additive to dielectric permeability

$$\Delta\varepsilon = -\frac{\omega_p^2}{\omega^2} = -\frac{4\pi^2 Ne^2}{m_{eh}^* \hbar\omega^2} \ , \qquad (6)$$

where e is a charge of electron, N is concentration nonequilibrium free electrons in a zone of conductivity, m_{eh}^* - the reduced mass of electron-hole pair. Generally N it is defined by speeds of generation electron-hole pair (in our case as a result of two-quantum absorption), them recombination and diffusion:

$$\frac{\partial N}{\partial t} = \frac{\eta\beta I^2}{\hbar\omega} - \frac{N}{\tau} + D_a \frac{\partial^2 N}{\partial x^2} \ , \qquad (7)$$

where η is quantum efficiency of generation electron-hole pair, β is the two-photon absorption coefficient, τ is time of life electron-hole pair, D_a is coefficient of ambipolar diffusions. As in InSb at the contribution in R is given only with the rare lattice which is written down by passing waves, diffusion member in (7) can be neglected. Taking into account also, that in InSb at T=300 K $\tau\approx 10^{-8}$ sec with [7], and duration of a laser pulse exceeds ~10^{-7} sec with, we get

$$N = \frac{\eta\beta I^2\tau}{\hbar\omega} \qquad (8)$$

Substituting (8) in (6) and defining connection $\Delta\varepsilon$ with $\chi^{(5)}$ as $\Delta\varepsilon = 4\pi\chi^{(5)}\sum E_i E_j E_k E_\ell$ we get

$$\chi^{(5)} = -\frac{\eta\beta n^2 c^2 e^2 \tau}{64\pi^2 m_{eh}^* \hbar\omega^3} \qquad (9)$$

Substituting in (9) known parameters InSb (η=0.5, n=3.95, $m_{eh}^* \approx 0.012$m, $\beta\approx 5.5$cm/MVt [6], $\tau \approx 10^{-8}$ sec), we get $\chi^{(5)}=5\cdot 10^{-8}$ esu, that will not bad be coordinated to experimentally measured value.

From results of researches of temperature dependence τ in InSb [3,6] follows, that reduction T from ~300K up to 200 K in samples with concentration of impurity less allows to increase τ than 10^{15}cm^{-3} up to $(0.5\text{-}1)\cdot 10^{-6}$ sec with. Accordingly, taking into account, as $\chi^{(5)}\sim\tau$ lowering temperature of a sample, it is possible to increase $\chi^{(5)}$ on 1, 5 - 2 order.

Thus, direct experiments confirm, that the basic contribution to four-wave mixing in InSb on 10,6 μm is given with a nonlinear susceptibility of the fifth order, and $\chi^{(5)}$ in InSb can be considerably increased.

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InSb – DƏ İNFRAQIRMIZI 10,6 μm DALĞA UZUNLUQ OBLASTINDA QEYRİ-XƏTTİ UDULMA VƏ BEŞİNCİ TƏRTİB QAVRAYICILIQ

Dar qadağan zonalı yarımkeçirici InSb-da 10,6mkm dalğa uzunluğunda qeyri-xətti optik udulmanın ikifotonlu udulma nəticəsində baş verdiyi göstərilmiş və müxtəlif dərəcəli leqirə olunmuş n – tip nümunələrdə qeyri-xətti optik udulma sabiti ölçülmüşdür. Dörddalğalı qarşılıqlı təsir prosesində qayıtmanın baş verməsinə səbəb olan əsas mexanizm ikifotonlu udulmada sərbəst elektronların generasiyasıdır. Bu prosesin effektivliyini məhdudlaşdıran əsas səbəb qeyri-xətti udulmadır.

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НЕЛИНЕЙНОЕ ПОГЛОЩЕНИЕ И ВОСПРИИМЧИВОСТЬ ПЯТОГО ПОРЯДКА В InSb В ИНФРАКРАСНОЙ ОБЛАСТИ ПРИ ДЛИНЕ ВОЛН 10,6μm

Показано, что нелинейное поглощение на длине волны 10,6мкм в узкозонном полупроводнике InSb обусловлено двухфотонным поглощением и измерена константа нелинейного поглощения в образцах n-типа с разной степенью легирования. Установлено, что основной механизм нелинейности, ответственный за отражение при вырожденном четырехволновом взаимодействии обусловлен генерацией свободных электронов при двухфотонном поглощении. Рост эффективности при четырехволновом взаимодействии в узкозонных полупроводниках ограничивается нелинейным поглощением взаимодействующих волн.

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