REFRACTIVE OPTICAL NON-LINEARITY IN Ge CRYSTALS CAUSED BY INTER-VALLEY REDISTRIBUTION OF "HOT" ELECTRONS

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A non-linearity of the refractive index in germanium crystals for infrared light caused by the redistribution of free electrons among equivalent valleys due to their heating by light waves has been calculated. The dependences of the non-linearity magnitude on the light wavelength and intensity, as well as on the electron concentration and the crystal temperature, have been given. The results of calculations give a proper explanation of the experimental dependence of the refractive index on the CO_2-laser radiation intensity obtained earlier.

In cubic many-valley semiconductors (Ge, Si), the strong optical refractive non-linearity in the infrared (IR) range is related to the redistribution of free electrons among equivalent valleys in a light wave field **E**. In the CO_2 laser radiation ($\lambda = 10.6$ μm), we observed the birefringence [1] and the degenerate four-wave interaction [2] caused by such nonlinear mechanism in Ge crystals with the carrier concentration $N_0 = 5\times10^{16}$ cm^{-3} at temperatures of 300 and 77 K. It was found that, when the light intensity I is less than a certain value (12 MW/cm^2 at 300 K and 8 MW/cm^2 at 77 K), the refractive index variation $\delta n_\parallel$ shows a linear increase with I. This is characteristic of the third-order non-linearity. At higher light intensities, the dependence of $\delta n_\parallel$ on I deviates from the linear behavior and becomes weaker.

In the present paper, we have carried out calculations of the intervalley electron redistribution and the corresponding variation of the refractive index in Ge crystals in the field of IR electromagnetic radiation in order to explain such a dependence. In addition, the dependences of the non-linearity magnitude on the radiation wavelength, free carrier concentration, and crystal temperature, which are needed for practical use, are also considered.

The intervalley redistribution of electrons occurs only at a non-symmetric orientation of the light-wave electric field **E** in regard to the axes of ellipsoidal valleys, when carriers have different vibrational movement energies in different valleys because of different effective masses (along the field) and achieve different heatings during light absorption. Both reasons of the redistribution were considered theoretically in [3]. Estimation carried out for Ge crystals with a carrier concentration of the order of 10^{16} cm^{-3} and radiation with a wavelength of 10.6 μm and the 40 MW/cm^2 intensity evidenced that the effect was caused mainly by the heating of carriers.

In order to calculate it, one needs to know the function of electron distribution in the electromagnetic radiation field. In general, it should be determined from the Boltzmann kinetic equation. However, in the range of electron concentrations where the optical non-linearity under consideration was observed and at not too strong heating, this function can be used in the Maxwell form with different electron temperatures in different valleys $T^{(i)}$. At that, we determined the electron temperatures as usual (see, e.g., [4, 5]) from the balance of the electromagnetic wave power absorbed by electrons in a given valley and that lost due to the electron interaction with phonons and at inter-electron collisions.

The energy absorbed by electrons for unit time at different scattering mechanisms equals to the product of the incident electromagnetic field energy flux and the light absorption coefficient. The expressions for the coefficient of light absorption by "hot" electrons in many-valley semiconductors at various mechanisms of carrier scattering depending on the electron concentration N_i in the i-th valley, their temperature $T^{(i)}$, and the light-wave electric field orientation in regard to the valley axis are given in [6, 7].

To calculate the mean rate of energy losses by electrons at their scattering by acoustic and optical phonons, we used the expressions in the form given in [8]. The energy exchange among electrons of different valleys at collisions was calculated according formulae given in [5].

The electron concentrations in the valleys were determined from the equation of particle balance in each valley. It was assumed that the redistribution of "hot" electrons among the valleys occurs due to the scattering by phonons ("inter-valley phonons"). In this case, the equations of particle balance have the following form being convenient for numerical calculations [9]:

$$N_i\gamma_i^{3/2}\Big[e^{Z_M(1-\gamma_i)}+1\Big]\int_0^\infty x^{1/2}(x+Z_M)^{1/2}\,e^{-\gamma_i x}dx =$$

$$= N_j\gamma_j^{3/2}\Big[e^{Z_M(1-\gamma_j)}+1\Big]\int_0^\infty x^{1/2}(x+Z_M)^{1/2}\,e^{-\gamma_j x}dx. \quad (1)$$

Here, $\gamma_i = T/T^{(i)}$, $Z_M = \hbar\omega_M/kT$, T is the lattice temperature, and $\hbar\omega_M$ is the "inter-valley phonon" energy.

Calculations were carried out at first for the experimental conditions at which we observed the considered kind of optical non-linearity in Ge [1, 2] ($N_0 = 5{\times}10^{16}$ cm^{-3}, $T = 300{\div}$ 77 K, $\lambda = 10.6$ μm) at the polarization of IR radiation coinciding with the [111] crystal axis. Here, we take into account the light absorption by electrons at the scattering by acoustic and optical phonons and impurities. The constants characterizing the intravalley electron scattering, parameter of scattering anisotropy for acoustic phonons, optical phonon frequencies, and germanium energy band parameters were taken like in [4]. For the intervalley electron scattering, the following phonon energies were used: $\hbar\omega_M$= 320 K and $\hbar\omega_M$= 120 K [4]. It should be noted that, for this direction of the light-wave electric field, all the valleys in germanium form 2 groups in which electron concentrations and electron temperatures are different. The first group consists of one valley with the long axis parallel to the light-wave electric field direction ([111]-valley). The second group is formed by three valleys located on the axes [$\bar{1}$11], [11$\bar{1}$], and [1$\bar{1}$1].

The results of calculations for the crystal temperature of 77 K are depicted in Fig. 1. It is seen that, with the light intensity growth, the electron temperature in the [111]-valley increases slower as compared to the second-group valleys. At an intensity of 20 MW/cm^2, it turns out to be about 83 K and it is about 121 K for other three valleys. Because of the energy exchange at collisions of electrons from different valleys, the temperature in the "cold" valley increases up to 105 K, while it decreases down to 115 K in the "hot" valleys. It is worth noting that the probability of inter-electron interaction increases with decrease in the average energy of carriers. Therefore, its influence on the electron heating in the valleys is more significant at small light intensities.

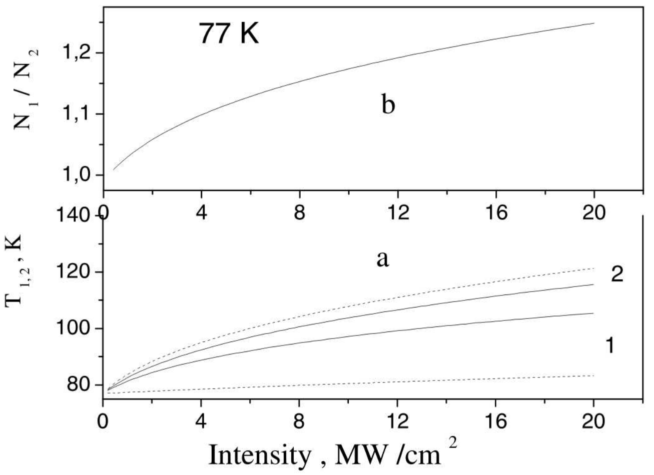

Fig. 1. Dependences of the electron temperature in the "cold" (*1*) and "hot" (*2*) valleys (*a*) and the ratio of the electron concentrations in them (*b*) for Ge on the CO_2-laser radiation intensity calculated with (solid lines) and without (dotted lines) account of the inter-electron interaction. $N_0 = 5{\times}10^{16}$ cm^{-3}

A similar behavior of electron temperature takes place also at a crystal temperature of 300 K. In this case, the magnitude of electron heating-up turns out to be somewhat larger because of a stronger light absorption by carriers [9]. The influence of the inter-electron interaction on the electron temperatures in the valleys becomes weaker.

The unequal heating of carriers leads to the violation of their uniform distribution over the valleys; they become more numerous in the less heated [111]-valley as compared to the more heated valleys of the second group. It is caused by the fact that the probability of electron scattering by the "intervalley phonons" leading to their transition from one valley to another depends on the carrier average energy and considerably increases with its growth. We note that the symmetry of the electron distribution in the k-space becomes different as compared to the initial cubic one.

Figure 1,*b* depicts the calculated ratio of electron concentrations in the "cold" and "hot" valleys, N_1/N_2, depending on the IR radiation intensity at a crystal temperature of 77 K. This ratio increases with the intensity growth and achieves the value of $\sim$ 1.26 at 20 MW/cm^2. It corresponds to the fact that about 6% of electrons from each of three valleys come to the [111]-valley. At 300 K, this value is less almost twice.

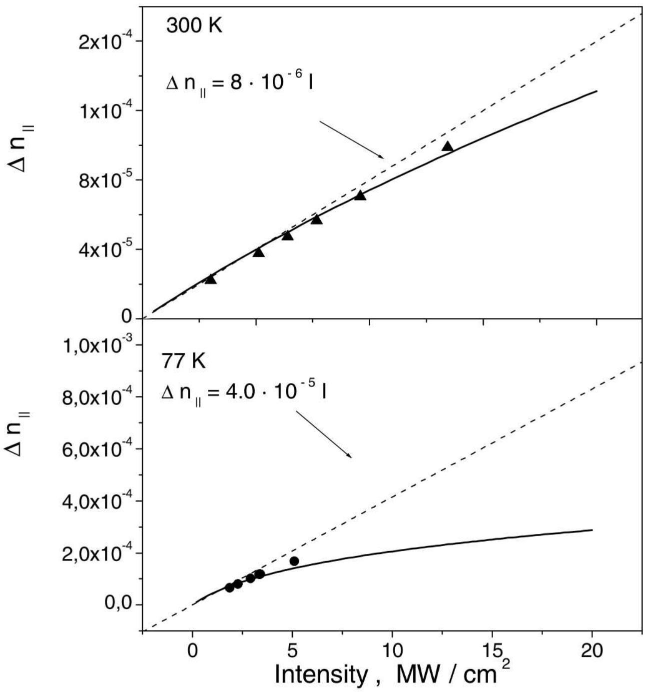

Fig. 2. Variation of the refractive index $\delta n_{\|}$ in Ge versus the CO_2-laser IR-radiation intensity. ($\mathbf{E} \,\|\, [111]$). Solid lines: calculations; dashed lines: linear fitting; circles and triangles: experiment

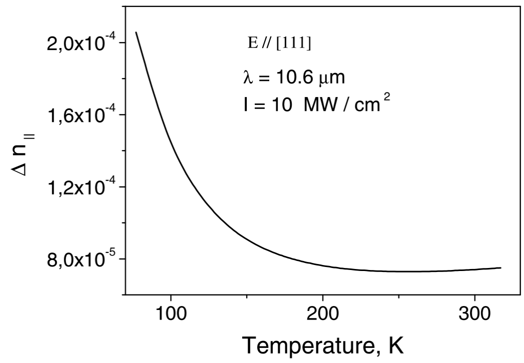

Fig. 3. Temperature dependence of the refraction index variation in Ge with $N_0 = 5 \times 10^{16}$ cm^{-3}

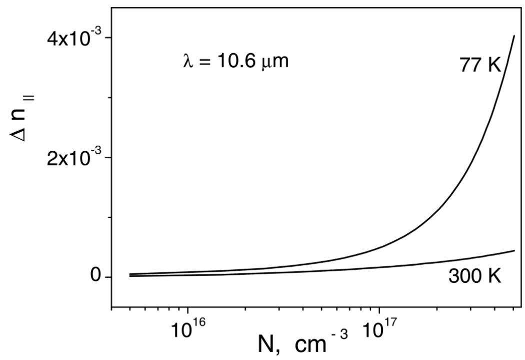

Fig. 4. Variation of the refractive index in Ge for the CO_2-laser radiation with an intensity of 20 MW/cm^2 versus the free carrier concentration

As a consequence of the redistribution, the contribution of free electrons to the dielectric permittivity becomes anisotropic and dependent on the intensity of light, inducing a non-linearity and an anisotropy of the light absorption coefficient and the refraction index. We observed these effects for the CO_2-laser radiation absorption in the germanium crystals; and they turned out to be small. With the variation of the radiation intensity from 2 to 29 MW/cm^2, the absorption coefficient varied only by 15% at 300 K and by 10% at 77 K, while its anisotropy at the maximal intensity was about 3 and 7%, respectively [9].

Let us calculate the dependence of the refractive index variation along the light-wave electric field $\delta n_{\|}$ on the IR radiation intensity in correspondence with the experimental conditions in [2]. At the radiation polarization under consideration, the value of $\delta n_{\|}$ is connected with the ratio of the electron concentrations in the "cold" and "hot" valleys, $\alpha = N_1/N_2$, as follows [10]:

$$\delta n_{\|} = \frac{4\pi^2 N_0 e^2}{3n\omega^2}\left(\frac{1}{m_t} - \frac{1}{m_l}\right)\left(\frac{\alpha - 1}{\alpha + 3}\right). \qquad (2)$$

Here, ω is the radiation frequency, m_t and m_l are, respectively, the transverse and longitudinal effective electron masses, and n is the refractive index.

Figure 2 shows the results of calculations for $\delta n_{\|}$ in Ge under the CO_2-laser irradiation. It is seen that the results give a good explanation for the experimental value of $\delta n_{\|}$ and its dependence on the radiation intensity. The dependence is, at first, linear $\delta n_{\|} = n_2 I$ with the values of the non-linearity parameter n_2 equal to 8×10^{-6} (MW/cm^2)$^{-1}$ at 300 K and 4.0×10^{-5} (MW/cm^2)$^{-1}$ at 77 K. Therefore, the refractive index variation can be described taking into account only the third-order non-linearity. With the further increase in the radiation intensity, the dependence deviates from the linear behavior and becomes weaker. Such a behavior of the refractive index cannot be described only by a cubic non-linearity, and one seems to need to take into account the non-linearity of the fifth order.

Let us consider now the main properties of the refractive non-linearity related to the inter-valley redistribution of electrons in the many-valley semiconductors: the dependences of its magnitude on the crystal temperature, carrier concentration, and IR-radiation wavelength. They are calculated for Ge and depicted in Figs. 3–5. As follows from the data presented

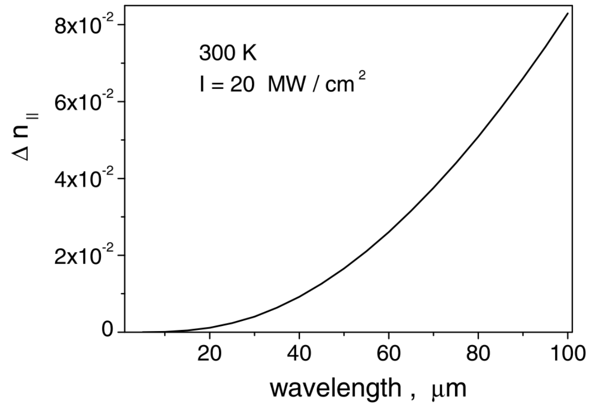

Fig. 5. Typical dependence of the refractive non-linearity related to the inter-valley "hot" electron redistribution in Ge ($N_0 = 5 \times 10^{16}$ cm^{-3}) on the radiation wavelength

earlier, the non-linearity magnitude increases by 5 times with lowering the temperature from 300 down to 77 K. As seen from Fig. 3, the changes occur basically at temperatures below 200 K. The dependence $\delta n_{\|}$ on the free electron concentration is stronger as compared to the linear dependence (Fig. 4). It is related to a significant role of the inter-electron interaction for the carrier concentrations under consideration, especially at 77 K. An important feature of the non-linearity of this kind is its strong increase with the IR radiation wavelength (Fig. 5). For example, if one uses the far IR-radiation with the wavelength $\lambda = 90.6$ μm (ammonia laser) instead of CO_2-laser radiation, then, at the same light intensity $I = 20$ MW/cm^2 at 300 K, the value of $\delta n_{\|}$ increases by almost 800 times and is about 6×10^{-2}. The effect is caused by the growth of the contribution of free carriers to the refractive index ($\delta n_{\|} \sim \lambda^2$) and their intervalley redistribution.

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РЕФРАКТИВНА ОПТИЧНА НЕЛІНІЙНІСТЬ У КРИСТАЛАХ ГЕРМАНІЮ, ПОВ'ЯЗАНА З МІЖДОЛИННИМ ПЕРЕРОЗПОДІЛОМ "ГАРЯЧИХ" ЕЛЕКТРОНІВ

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Р е з ю м е

Розраховано нелінійність показника заломлення n-Ge для інтенсивного інфрачервоного випромінювання, яка зумовлена перерозподілом вільних електронів між еквівалентними долинами внаслідок розігрівання їх світлом. Наведено залежності величини нелінійності від довжини хвилі та інтенсивності випромінювання, концентрації електронів і температури кристала. Результати обчислень добре пояснюють отриману раніше експериментальну залежність зміни показника заломлення від інтенсивності випромінювання CO_2-лазера.