Self-Induced Birefringence of Infrared Light in n-Ge

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(Received 6 July 1993)

We report the experimental proof of the self-induced birefringence of infrared light in many-valley cubic semiconductors. The effect is connected with the redistribution of free electrons between the equivalent valleys on account of heating by the infrared wave. The agreement of the obtained data with the theoretical prediction is very good.

PACS numbers: 77.30.+d, 72.20.Ht, 72.80.Cw

Nonlinear optical effects associated with free carriers in semiconductors have been investigated, e.g., in [1–5]. The frequency mixing and the optical phase conjugation of CO_2 laser beams arising from the nonparabolicity of the conduction band were investigated in III-V compounds and in $Hg_{1-x}Cd_xTe$ [1,2] . The third harmonic in the far IR region due to both the nonparabolicity of the energy band and the energy dependence of the scattering events was observed in Ge and Si [5].

In the cubic many-valley semiconductors strong nonlinearity of optical characteristics in the IR region is connected with the redistribution of the carriers between the equivalent valleys. There are two reasons for this redistribution: (a) the energy displacement of a valley, which is caused by the oscillatory character of the carrier motion in the electric field $\mathbf{E}$ of the light wave and therefore depends on the orientation of $\mathbf{E}$ to the axes of the valley; (b) the different carrier heating in the various valleys. As shown theoretically the latter mechanism dominates over the former one [6].

As a consequence of the redistribution, the contribution of the free carriers to the dielectric function becomes anisotropic and dependent on the intensity of the light, causing self-induced effects of laser beams.

Some phenomena in n-Ge for $\mathbf{E}$ of a CO_2 laser wave parallel to $\langle 111 \rangle$ were theoretically investigated in [7]. As well known for this orientation of the electric field the redistribution of electrons in Ge has a maximum. Nonetheless, it is reasonable to consider the other directions of $\mathbf{E}$ with respect to the crystallographic axes. For such nonsymmetric directions self-induced birefringence of light waves takes place. The investigation of this effect is the subject of the present paper.

The self-induced change of the light polarization in n-Ge has been measured for the linearly polarized IR light with $\lambda = 10.6$ μm at 300 and 80 K. The samples were cut from Sb-doped Ge single crystals in the form of plates of known thickness. Slight wedge angles were used between the opposite surfaces of the plate to avoid Fabry-Pérot effects. The carrier concentration was measured by the Hall effect and was about 3×10^{12} cm^{-3} (intrinsic) and 5×10^{16} cm^{-3}, respectively.

The multimode TEA-CO_2 laser (where TEA is transversely excited at atmospheric pressure) was used to provide the IR radiation. The laser produced a maximal peak power of about 1 MW in a 100 nsec FWHM single pulse. Pyroelectric detectors with a response time of about 3 nsec were used to measure the peak power of the incident and transmitted light pulses and their shapes. The variation of the output peak power was about $\pm 15\%$ from one pulse to the other, but no change of the pulse shape was observed. Therefore in the experiment the measurements were performed with pulses of the same peak power within an accuracy of 3%.

The propagation vector of the incident radiation was along the $\langle \bar{1}10 \rangle$ axis. The rotation of the sample around this axis served to change the orientation of $\mathbf{E}$ in the crystal.

We routinely measured the polarization of the output wave using a GaAs analyzer and a quarter-wave plate. With respect to the lateral distribution of the intensity by using a diaphragm of 4 mm diameter a central portion of the beam was selected within which the peak power varied no more than 10%. This radiation was collimated by a telescope to an intended spot size of 1 mm^2 on the sample. A set of calibrated CaF_2 attenuators allowed us to vary the incident light intensity on the sample surface in the range of 1 MW/cm^2 to 20 MW/cm^2.

Within the accuracy of the measurement, a linear-polarized light of a small intensity ($I_{\text{inc}} \leq 10$ kW/cm^2) passes through the samples of n-type Ge with no change of polarization. Contrary to this, increasing the light intensity we obtained elliptically polarized outgoing light with the major axis of the ellipse along the polarization vector of the incident light. This means that the light propagates within the crystal in the form of an ordinary and an extraordinary beam with a certain difference of the phase φ_d and polarized at right angles to each other.

The ratio between the light intensities perpendicular and parallel to the major axes of the ellipse of polarization (the coefficient of ellipticity ξ) depends on the orientation of the electric field of the incident wave $\mathbf{E}$ relative to the crystallographic axes of the sample. It is shown in Fig. 1 with Θ denoting the angle between $\mathbf{E}$

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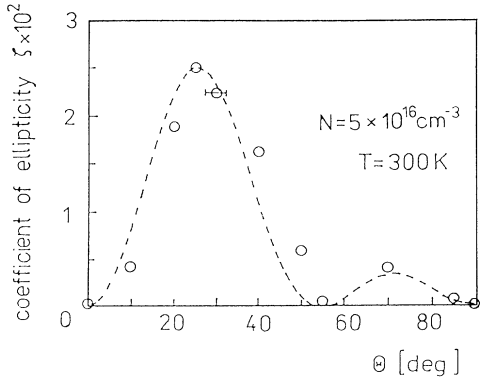

FIG. 1. The ellipticity coefficient ξ as a function of the electric field direction of the laser wave in the $(\bar{1}10)$ plane for an incident intensity of 15 MW/cm^2. ○ denotes experimental values. The error of the angle is $\pm 1°$. Dashed line according to Eq. (9) with E_0 and ϵ_2 as given in the text.

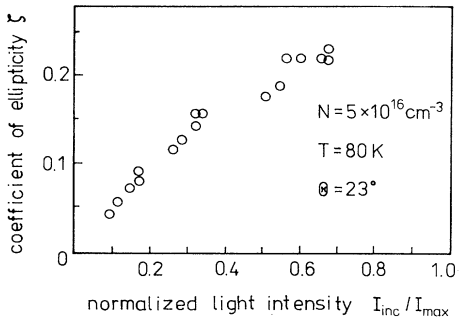

FIG. 2. Dependence of the ellipticity coefficient ξ on the normalized light intensity ($I_{\max} = 30$ MW/cm^2).

and the $\langle 001 \rangle$ direction in the $(\bar{1}10)$ plane. At a constant light intensity this coefficient rises sharply as the sample temperature is decreased. For example, changing the temperature from 300 to 80 K the value of ξ for the sample with the carrier concentration of about 5×10^{16} cm^{-3} increases from 2.5×10^{-2} to 1.7×10^{-1} at $I_{\text{inc}} = 15$ MW/cm^2 and $\Theta = 23°$. For this case the differences of phase φ_d measured by a quarter-wave plate are equal to 17° and 45° at 300 and 80 K, respectively, for a sample thickness of about 0.32 cm. The coefficient ξ increases with increasing light intensity in the beginning and then saturates as shown in Fig. 2 for 80 K.

We would like to emphasize that within the limits of experimental error for intrinsic Ge the polarization of the light remains unchanged up to the maximum intensity used in the experiment.

Based on the fact that the change in the polarization is only observed in doped Ge and that it increases sharply with decreasing sample temperature, we conclude that this effect is due to the redistribution of free electrons between the equivalent valleys.

As is well known the dielectric displacement in nonlinear systems can be described in powers of the components of the electric field of an electromagnetic wave

$$D_i = \epsilon_{ij} E_j + \epsilon_{ikj} E_k E_j + \epsilon_{iklj} E_k E_l E_j + \cdots . \tag{1}$$

In cubic crystals $\epsilon_{ij} = \epsilon_0 \delta_{ij}$ and in the presence of an inversion center $\epsilon_{ijk} = 0$, too. In the crystallographic coordinate system the tensor ϵ_{iklj} has four nonvanishing independent components of the types $\epsilon_{iiii}, \epsilon_{iijj}, \epsilon_{ijij}$, and ϵ_{ijji} for the classes T_h, O, O_h [8]. Furthermore, the tensor ϵ_{iklj} has to be symmetric with respect to the permutation of the inner or outer index [9]. As a consequence, the number of independent tensor components is reduced to 3.

Equation (1) can be rewritten as

$$D_i = \epsilon_{ij}(\boldsymbol{E}) E_j , \tag{2}$$

where the tensor ϵ_{ij} has the form

$$\begin{aligned} \epsilon_{ij} &= 2\epsilon_3 E_i E_j, \quad i \neq j , \\ \epsilon_{ii} &= \epsilon_0 + (\epsilon_1 - \epsilon_2) E_i^2 + \epsilon_2 E^2 . \end{aligned} \tag{3}$$

Here, ϵ_1, ϵ_2, and ϵ_3 denote ϵ_{iiii}, ϵ_{ijji}, and ϵ_{ijij}, respectively.

It is convenient to use a new coordinate system with $\bar{x} \parallel \langle 110 \rangle$, $\bar{y} \parallel \langle \bar{1}10 \rangle$, and $\bar{z} \parallel \langle 001 \rangle$ for the electric vector lying in the $(\bar{1}10)$ plane in correspondence with the experimental condition. Then the $\hat{\epsilon}$ tensor reads

$$\hat{\epsilon} = \begin{bmatrix} \epsilon_{11} + \epsilon_{12} & 0 & \sqrt{2}\,\epsilon_{13} \\ 0 & \epsilon_{11} - \epsilon_{12} & 0 \\ \sqrt{2}\,\epsilon_{13} & 0 & \epsilon_{33} \end{bmatrix} . \tag{4}$$

Here, ϵ_{ij} are the components of the $\hat{\epsilon}$ tensor in the crystallographic coordinate system given by Eq. (3), in which the electric field components E_i are $E_1 = E_2 = E \sin\Theta$ and $E_3 = E \cos\Theta$. The values of $\hat{\epsilon}$ along the main axes are

$$\epsilon_y = \epsilon_0 + \frac{E^2}{2} \left[\sin^2\Theta \, (\epsilon_1 + \epsilon_2 - 2\epsilon_3) + 2\epsilon_2 \cos^2\Theta \right] , \tag{5}$$

$$\epsilon_{x,z} = \epsilon_0 + \frac{E^2}{2} \left(\epsilon_1 + \epsilon_2 + \left(\epsilon_3 - \frac{1}{2}\epsilon_1 + \frac{1}{2}\epsilon_2 \right) \sin^2\Theta \pm \left\{ \left[\left(\frac{3}{2}\epsilon_1 - \frac{3}{2}\epsilon_2 + \epsilon_3 \right) \sin^2\Theta - \epsilon_1 + \epsilon_2 \right]^2 + 16\,\epsilon_3^2 \sin^2\Theta \cos^2\Theta \right\}^{1/2} \right) .$$

One of the main axes of the $\hat{\epsilon}$ tensor forms the angle α with the $\langle 001 \rangle$ direction determined by

$$\tan 2\alpha = \frac{4\epsilon_3 \sin\Theta\cos\Theta}{(\epsilon_1-\epsilon_2)(1.5\sin^2\Theta-1)+\epsilon_3\sin^2\Theta}\,. \quad (6)$$

From Eq. (6) it is to be seen that only for $\mathbf{E}\parallel\langle 001\rangle$ or $\mathbf{E}\parallel\langle 110\rangle$ as well as $\mathbf{E}\parallel\langle 111\rangle$ one of the main axes of the $\hat{\epsilon}$ tensor coincides with the given crystallographic orientation, respectively.

Consequently, when the laser wave is incident perpendicularly to the $(\bar{1}10)$ plane and the electric vector is not coinciding with one of the indicated crystallographic axes two waves with refractive indexes $n_x=\sqrt{\epsilon_x}$ and $n_z=\sqrt{\epsilon_z}$ propagate through the crystal with their polarization along the main axes of the $\hat{\epsilon}$ tensor; i.e., the crystal exhibits self-induced birefringence of the electromagnetic waves.

The difference of the phases between these two waves will be

$$\varphi_d = \frac{2\pi}{\lambda}\int_0^d (n_x-n_z)\,dy \approx \frac{\pi}{\lambda\sqrt{\epsilon_0}}\int_0^d(\delta\epsilon_x-\delta\epsilon_z)\,dy\,, \quad (7)$$

where d is the thickness of the sample.

We assume that the direction of the heating electric field of the waves coincides with that of the incident wave, if φ_d is sufficiently small. The constants ϵ_2 and ϵ_3 are the same for this case. Obviously the constant ϵ_1 has to be zero for n-type Ge if the nonlinearity stems from the redistribution of electrons only. From Eq. (6) it follows that α does not depend on the components of the $\hat{\epsilon}$ tensor then and is only determined by Θ. Therefore, the axes of the tensor do not change along the direction of the wave propagation.

It is easy to show that in the lowest order

$$\varphi_d = \frac{2\pi E_0^2\epsilon_2}{\lambda\kappa(\sqrt{\epsilon_0}+1)^2}\,[1-\exp(-\kappa d)] \times\sqrt{(1-\tfrac{1}{2}\sin^2\Theta)^2+16\sin^2\Theta\cos^2\Theta}\,. \quad (8)$$

Here, κ is the free electron absorption coefficient and $\mathbf{E}_0$ the amplitude of the laser wave.

The ratio of the outgoing light intensities for two analyzer orientations coinciding with the polarization of the incident light and the orientation perpendicular to it is given by

$$\xi = \frac{\sin^2[2(\Theta+\alpha)]\,(1-\cos\varphi_d)}{2-\sin^2[2(\Theta+\alpha)]\,(1-\cos\varphi_d)}\,. \quad (9)$$

The constant ϵ_2 can be expressed by the redistribution of electrons between the valleys for any polarization of the incident light. As an example we consider the case when the electric field of the wave is orientated along the $\langle 111\rangle$ direction, for which self-induced birefringence is absent as can be seen from Fig. 1. It is known (e.g., [10]) that for $\mathbf{E}\parallel\langle 111\rangle$ the concentration of the electrons in the valley located on this crystallographic axis n_1 is increased, but in the other three valleys the concentration of the electrons n_3 is decreased.

Let the carrier concentration in each of these three valleys be $n_3=\beta n_0$, where $\beta\le 1$ is the degree of the redistribution of the electrons and $4n_0=N$ is the total concentration. In this case the change of the dielectric constant $\delta\epsilon$ along the electric field $\mathbf{E}$ can be expressed by

$$\delta\epsilon = \frac{8\pi e^2}{3\omega^2}N(1-\beta)\left(\frac{1}{m_\perp}-\frac{1}{m_\parallel}\right)\,. \quad (10)$$

On the other hand, as can be seen from Eq. (5) $\delta\epsilon_x=\epsilon_x-\epsilon_0=E_0^2\epsilon_2$. The degree of the redistribution of the electrons is then

$$\beta = 1-\epsilon_2 E_0^2\,\frac{3\omega^2}{8\pi e^2 N(\frac{1}{m_\perp}-\frac{1}{m_\parallel})}\,. \quad (11)$$

The system of equations (6), (8), and (9) describes the angular dependence of ξ in the regime of small φ_d. From these equations it is seen that the position of the extrema given by $d\xi/d\Theta=0$ does not depend on the components of the $\hat{\epsilon}$ tensor, but only on Θ, whereas the value of ξ depends on ϵ_2. The maximum of the ellipticity coefficient of the outgoing light is expected to occur at $\Theta\simeq 25°$ and $\Theta\simeq 71°$ as the electric field of the incident wave is rotated in the $(\bar{1}10)$ crystallographic plane. Besides, the self-induced birefringence must be absent (i.e., the ellipticity coefficient is equal to zero) for polarization along the $\langle 001\rangle$, $\langle 110\rangle$, as well as $\langle 111\rangle$ directions in this plane ($\Theta=0$, 90°, and 53°).

The experimental results are in very good agreement with the predictions of the theory. As seen in Fig. 1, the ellipticity coefficient has its maximum values at the predicted orientations. Using the experimental value of the ellipticity coefficient $\xi(\Theta)$ and Eq. (9) we may determine the phase difference between the ordinary and extraordinary beams, after evaluating α from Eq. (6). With the incident intensity $I_{\rm inc}=15$ MW/cm^2 and $\Theta=23°-25°$ (experimental uncertainty) for a crystal with a carrier concentration $n=5\times10^{16}$ cm^{-3} and a length $d=0.32$ cm, φ_d was calculated to be 18° at room temperature. This value agrees very well with the experimental one, which is equal to 17°. The constant ϵ_2 obtained from Eq. (8) is then $\epsilon_2=2.68\times10^{-8}$ esu^{-2} when the value of the absorption coefficient $\kappa=2.15$ cm^{-1} determined by absorption measurements and $\epsilon_0=16$ are used.

Using Eq. (10) and the value of ϵ_2 at room temperature for $\mathbf{E}\parallel\langle 111\rangle$ the redistribution β can be estimated: If $E_0=1.06\times10^5$ V/cm ($I_{\rm inc}=15$ MW/cm^2), then $\beta=0.91$, when $m_\perp=0.18m_0$ and $m_\parallel=1.588m_0$ according to [11]. With this value of β and the condition $n_1+3n_3=4n_0$ the ratio n_1/n_3 is determined and equals 1.4. In analogy at 80 K from Fig. 2 in the region of small φ_d the value $\epsilon_2=1.25\times10^{-7}$ esu^{-2} is obtained with $\kappa=1$ cm^{-1}. For $E_0=6.1\times10^4$ V/cm (i.e., $I_{\rm inc}=5$

MW/cm^2) β equals 0.856 and $n_1/n_3 = 1.65$. These results can be compared with data determined in dc fields. It should be remembered that the effective heating fields in both cases differ by the factor $\omega\tau$ with τ being the momentum relaxation time. For our case we estimate τ to be about 10^{-13} and 4×10^{-13} sec at 300 and 80 K, respectively, using the measured values of the mobilities. These values yield equivalent dc fields of 4600 and 860 V/cm, respectively. It should be mentioned that the carrier redistribution between the valleys is a function of the carrier density, because it strongly diminishes with increasing electron-electron interaction [12]. Consequently, for 5×10^{16} cm^{-3} this effect is important for 80 K and only weakly pronounced for 300 K. As there are only data published for purer samples such results [13] agree with our value for 300 K, but yield a significant higher one [14] for 80 K as expected.

Note that self-induced birefringence of microwaves (at 9.4 GHz) connected with the hot carrier redistribution between the equivalent valleys in n-Ge and -Si was investigated in [15] for $\omega\tau \ll 1$.

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