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Infrared phase-conjugate reflection by hot electron-induced optical nonlinearity in n-Ge

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Abstract. We have studied backward-degenerate four-wave mixing at CO_2 laser wavelengths in n-type Ge having a free electron density of $N = 5 \times 10^{16}\,\mathrm{cm}^{-3}$. The phase conjugation due to the redistribution of free electrons between the equivalent valleys was observed. The effect occurs only when the electric field $\boldsymbol{E}$ of the light wave is aligned nonsymmetrically relative to the long axes of the valleys in the crystal, and is related to carrier heating by the infrared radiation.

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The phase conjugation of infrared CO_2 laser beams has been observed in direct narrow band-gap semiconductors, such as InSb, InAs, and $Hg_{1-x}Cd_xTe$ ($0.217 < x < 0.232$), this being associated with free carrier optical nonlinearity [1–3]. The most notable origin of this nonlinearity is the nonlinear dependence of carrier velocity on momentum, due to the nonparabolicity of the conduction band.

In cubic many-valley semiconductors, the strong nonlinearity of the dielectric permittivity in the IR region is related to the redistribution of the electrons between the equivalent valleys [4, 5]. The dominant reason for this redistribution is the different carrier heating in the various valleys by infrared radiation, and therefore the redistribution occurs only when the electric field $\boldsymbol{E}$ of the light wave is aligned nonsymmetrically relative to the long axes of the valleys in the crystal.

As a consequence of the electron redistribution, the contribution of the free carriers to the dielectric permittivity becomes anisotropic and dependent on the intensity of the light, initiating light-induced changes in the optical constants.

The existence of optical nonlinearity caused by the redistribution of the hot electrons in many-valley semiconductors has been confirmed successfully in our previous works [6, 7], in which a self-induced birefringence of intense IR CO_2 laser radiation in n-Ge was observed. It was demonstrated that this mechanism of optical nonlinearity dominates over any other for a relatively narrow interval of free carrier concentration $N_e \sim (3-5) \times 10^{16}\,\mathrm{cm}^{-3}$. For more lightly doped samples, the contribution of the free carriers to the susceptibility in Ge is small. However, with a carrier concentration larger than $5 \times 10^{16}\,\mathrm{cm}^{-3}$, the energy exchange between the electrons from different valleys caused by electron–electron collisions becomes essential. This type of interaction decreases the difference between the electron temperatures in various valleys and leads to the suppression of the redistribution of the electrons between the equivalent valleys. The magnitude of the nonlinearity was found to rise sharply as the sample temperature is decreased. The measured values of the third-order nonlinear susceptibilities were 2×10^{-9} esu at 300 K and 1×10^{-8} esu at 80 K.

In this paper we describe the observation and study of another nonlinear effect caused by hot carrier optical nonlinearity in n-Ge, namely a phase conjugation of CO_2 laser IR radiation via degenerate four-wave mixing.

The backward-degenerate four-wave mixing (BDFWM) in n-Ge has been measured for IR light with a CO_2 laser wavelength of $\lambda = 10.6\,\mu\mathrm{m}$ at 300 and 80 K. The samples were cut from Sb-doped Ge single crystals in the form of slabs with thickness $\ell = 0.32$ cm. The carrier concentration was measured by the Hall effect and was about $5 \times 10^{16}\,\mathrm{cm}^{-3}$ at both 300 K and 80 K.

A TEA-CO_2 laser (where TEA is transversely excited at atmospheric pressure) was used as a light source. The laser produced a maximal peak power of about 2 MW in a 100-ns FWHM single pulse. The laser beam was split into two beams; the pump beam with intensity I_1 and the probe beam with intensity $I_3 < I_1$ (Fig. 1a). These beams were incident on the same spot on the sample with an angle of 3° between them. The optical path difference between the pump and the probe beams did not exceed 1 cm. The counter-propagating pump beam with intensity I_2 was obtained by the Fresnel reflection of the first pump beam from the rear face of the sample.

The phase-conjugate beams (I_4) were observed on the same side of the sample as the first pump and probe beams by introducing a ZnSe beam splitter into the probe beam. The phase-conjugate signals were measured using a HgCdTe detector at 300 K and displayed on an oscilloscope. Pyroelectric detectors with a response time of about 3 ns were used to measure the peak power of the incident pump and probe light pulses and their shape.

The propagation vectors of the incident beams were along the $\langle 110 \rangle$ axis. BDFWM experiments have been per-

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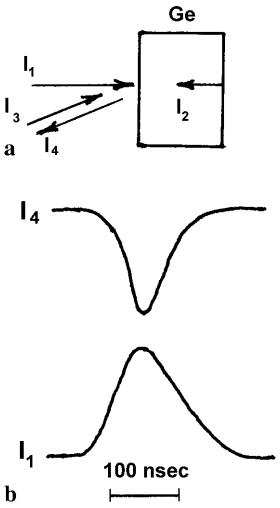

Fig. 1. a Setup for backward-degenerate four-wave mixing in a Ge crystal. I_1, I_2 denote pump waves, I_3 is the probe wave, I_4 is the phase-conjugate signal. **b** Time traces of pump wave I_1 and phase-conjugate wave I_4

formed for two orientations of the electric field $\boldsymbol{E}$ of the incident beams in the crystal, $\boldsymbol{E}//\langle 111\rangle$ and $\boldsymbol{E}//\langle 001\rangle$. For the $\boldsymbol{E}//\langle 111\rangle$ orientation, the effective mass of the electrons along the $\boldsymbol{E}$ direction is higher in the valley located on the $\langle 111\rangle$ crystallographic axis than in the other three valleys, and the carrier heating by the electric field of the light wave is smaller in it. Due to the different mean carrier energies in the valleys, the redistribution of the electrons between them takes place [8]. However, for the $\boldsymbol{E}//\langle 001\rangle$ orientation, all the valleys are settled down symmetrically to the field direction. The carrier heating is the same in them and therefore no redistribution of electrons between the valleys occurs.

It should be noted that the redistribution of hot electrons does not result in birefringence for $\boldsymbol{E}$ oriented along the $\langle 111\rangle$ axis in n-Ge [6]. Therefore, the polarization of the pump and probe beams remains unchanged with the propagation.

Figure 1b represents the time traces of the first pump wave I_1 and back-reflected signal wave I_4 for $\boldsymbol{E}$ along the $\langle 111\rangle$ crystallographic axis. As expected, the pulse duration of the back-reflected signal wave I_4 is smaller than that of the pump wave I_1. At the same time, no delay is observed in the position of the signal pulse maximum as compared to that of the pump pulse maximum. Thus the conclusion can be made that the response time of the nonlinearity is at least much shorter than the pulse duration, i.e. the saturation of the nonlinear response is reached during the pulse.

Figure 2 shows a log–log plot of the normalized intensities I_4/I_1 of the back-reflected wave (BDFWM, peak power reflectivity P_c) versus the incident pump intensity I_1 for $\boldsymbol{E}$ along the $\langle 111\rangle$ crystallographic axis. The reflectivity increases quadratically with the pump intensity I_1 (the slope of the lines in a log–log plot is approximately two), thus indicating that only the third-order interaction contributes significantly to the BDFWM signals. The reflectivity rises sharply as the sample temperature is decreased at constant pump beam intensity. By changing the temperature from 300 to 80 K, for example, the phase-conjugate reflectivity can be increased from 1% to 64% at $I_1 = 40\,\mathrm{MW/cm^2}$.

It should be emphasized that for $\boldsymbol{E}//\langle 001\rangle$, where the inter-valley redistribution of the hot electrons does not occur,

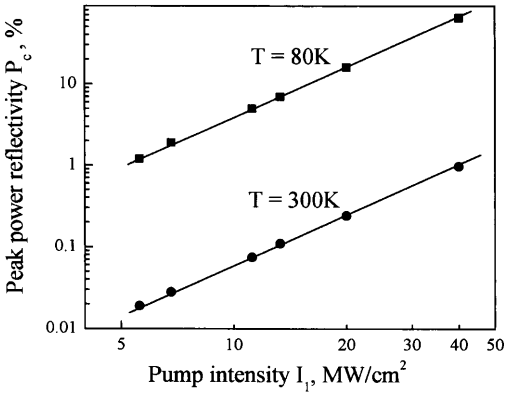

Fig. 2. BDFWM peak power reflectivity P_c as a function of the incident pump wave intensity

only a very weak phase-conjugate signal has been detected at the maximum pump intensity used in the experiment. The estimated BDFWM reflectivity, P_c, is about 2×10^{-2}% for both 300 K and 80 K at $I_1 = 40\,\mathrm{MW/cm^2}$.

Based on the fact that the BDFWM reflectivity increases sharply with decreasing sample temperature for the orientation of the electric field $\boldsymbol{E}$ along the $\langle 111\rangle$ crystallographic axis, we conclude that the observed phase-conjugate reflectivity is due to the redistribution of the hot electrons between the equivalent valleys. The inter-valley transition time in n-Ge is essentially smaller than the used laser pulse duration, about 100 ns [9]. Therefore, the optical nonlinearity connected with the redistribution of free electrons is fast enough to produce the observed steady-state BDFWM.

BDFWM reflectivity can be expressed in terms of the third-order nonlinear susceptibility $\chi^{(3)}$ [10]. Taking into account the Fresnel reflection R of incident and output waves from the crystal faces and the light absorption in the crystal, the BDFWM reflectivity is given by:

$$P_c = \frac{1024\pi^4\omega^2 \left|\chi^{(3)}\right|^2 L^2}{n^4c^4} I_1^2 R(1-R)^4\,, \tag{1}$$

where ω is the optical frequency, I_1 is the intensity of the incident pump beam and L is the effective overlap length given by:

$$L^2 = \left(\frac{1-\mathrm{e}^{-\alpha\ell}}{\alpha}\right)^2 \mathrm{e}^{-2\alpha\ell}\,, \tag{2}$$

with α being the absorption coefficient.

By using (1) and (2), together with experimentally determined values for P_c, we may evaluate the third-order nonlinear susceptibility $\chi^{(3)}$. For $\boldsymbol{E}//\langle 111\rangle$ we obtain 1.9×10^{-9} esu and 9×10^{-9} esu at 300 and 80 K, respectively, with the measured absorption coefficients $\alpha_{300\,\mathrm{K}} = 2.15\,\mathrm{cm^{-1}}$ and $\alpha_{80\,\mathrm{K}} = 1\,\mathrm{cm^{-1}}$ and $R = 0.36$. These values for the third-order susceptibilities associated with the redistribution of the hot electrons agree very well with those obtained using the self-induced birefringence experiments [6, 7].

The BDFWM reflectivity, measured for $\boldsymbol{E}//\langle 001\rangle$, corresponds to the third-order nonlinear susceptibility, $\chi^{(3)} \sim 10^{-10}$ esu, for both 300 K and 80 K, which is close to the value of $\chi^{(3)}$ due to the anharmonic motion of bound electrons in Ge [11].

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