Cyclotron resonance of the nonlinear optical susceptibility of *n*-type InSb

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An investigation was made of the cyclotron resonance of the nonlinear optical susceptibility $X^{(3)}(-\omega_3, \omega_1, \omega_1, -\omega_2)$, responsible for the frequency shift of the $\omega_3 = 2\omega_1 - \omega_2$ type in *n*-type InSb. The experiments were carried out using a *Q*-switched CO_2 laser emitting simultaneously at two frequencies $\omega_1 \approx 944$ cm^{-1} and $\omega_2 \approx 1044$ cm^{-1}. A semiclassical theory based on the solution of the equation of motion of a conduction electron in a nonparabolic band with the Kane dispersion law described well the position and profile of the cyclotron resonance line of $X^{(3)}(-\omega_3, \omega_1, \omega_1, -\omega_2)$. It was established that, in contrast to the linear cyclotron resonance case, the resonance of the nonlinear susceptibility of a doped semiconductor in the Voigt experimental geometry is not governed by the frequency of magnetoplasma oscillations $(\omega_p^2 + \omega_c^2)^{1/2}$ but by the cyclotron frequency of one-particle excitations ω_c. The electron relaxation time at the difference frequency $\omega_2 - \omega_1$, as well as the magnitude and sign of the nonlinear optical susceptibility of bound electrons in InSb were determined experimentally.

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INTRODUCTION

Nonlinear optical phenomena have been used successfully in the generation and conversion of electromagnetic radiation, and in obtaining information on the energy spectra of solids.

Studies of the phenomenon of frequency mixing in narrow-gap semiconductors[1] have established that the main source of the nonlinearity of such materials is the conduction band nonparabolicity.

In the case of cubic crystals it is convenient to investigate the nonlinear process of frequency mixing of the $\omega_3 = 2\omega_1 - \omega_2$ type, which is described by the third-order nonlinear susceptibility $\chi^{(3)}(-\omega_3, \omega_1, \omega_1, -\omega_2)$. For a combination of frequencies such that ω_1 is close to ω_2, the frequency ω_3 lies in the same spectral range and the phase-matching condition is satisfied well, so that relatively weak nonlinearities can be detected.

From the quantum point of view the generation of radiation of frequency ω_3 is a coherent four-photon process occurring via three intermediate states. In this process the initial and final states of the electron system are identical and the photons participating in the conversion obey the law of conservation of energy. When the difference between the photon energies $\hbar(\omega_2 - \omega_1)$ is close to the separation between discrete or quasidiscrete energy levels of electrons, the probability of transition to the second intermediate state increases resonantly and this gives rise to a resonance of the nonlinear optical susceptibility. If a CO_2 laser emitting at two frequencies $\omega_1 \approx 944$ and $\omega_2 \approx 1044$ cm^{-1} is used, the difference $\omega_2 - \omega_1 \approx 100$ cm^{-1} corresponds to the separation between the electron Landau levels of InSb in relatively weak magnetic fields of ~20 kOe. Yablonovich, Bloembergen, and Wynne[2] were the first to observe a resonance of the nonlinear optical susceptibility $\chi^{(3)}(-\omega_3, \omega_1, \omega_1, -\omega_2)$ in the case when the difference frequency $\Delta\omega = \omega_2 - \omega_1$ was equal to the cyclotron frequency or twice that frequency. For brevity, we shall refer to this effect as the cyclotron resonance of the nonlinear optical susceptibility. Subsequently, Wynne suggested a semiclassical theory[3] based on the solution of the equation of motion of an electron in the conduction band of InSb subjected to a magnetic field; this theory explains the principal experimental results of Ref. 2. A more rigorous analysis of the behavior of the nonlinear optical susceptibility in a magnetic field by solving the Boltzmann transport equation[4] does not give any significantly different results but explains the meaning of the electron momentum relaxation time introduced phenomenologically by Wynne in the final result.

Yablonovich *et al.*[2] investigated only one sample of *n*-type InSb and, therefore, in comparing the theory with experiment Wynne varied several parameters at the same time in an arbitrary manner. We carried out a detailed investigation of the cyclotron resonance of the nonlinear optical susceptibility of *n*-type InSb in a wide range of electron densities.

On the basis of general considerations we can expect that, in addition to the resonance at the cyclotron frequency $\omega_c = eH/m^*c$ (m^* is the effective electron mass), the nonlinear optical susceptibility $\chi^{(3)}(-\omega_3, \omega_1, \omega_1, -\omega_2)$ may also exhibit a resonance when the frequency difference $\omega_2 - \omega_1$ becomes comparable with the spin frequency of the conduction electrons. This resonance has indeed been observed.[5,6]

We shall now consider the theory of the cyclotron resonance of the nonlinear optical susceptibility.

THEORY

We shall use the results of the simpler semiclassical analysis of the behavior of the nonlinear optical susceptibility in a magnetic field based on the solution of the equation of motion of an electron in the conduction band of a narrow-gap semiconductor.[3] The application of the semiclassical approach is justified by the fact that in the case of resonance at twice the cyclotron fre-

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quency $\Delta\omega = 2\omega_c$, which will be the only case considered here, the inequality $\hbar\omega_c \ll \varepsilon_F$ is satisfied for practically all the electron densities of interest to us; here, ε_F is the Fermi energy of the electron system (at the temperature of liquid helium the electron gas in the conduction band is degenerate in all cases when the nonlinearity is not too weak). We shall consider the motion of an electron in a band described by the Kane Hamiltonain

$$\varepsilon = (\varepsilon_g/2)\,[(1+2p^2/m^*\varepsilon_g)^{1/2}-1], \tag{1}$$

where ε_g is the band gap; m^* is the effective mass at the bottom of the conduction band; p is the electron momentum.

In this case the electron velocity is

$$\mathbf{v}(\mathbf{p}) = (\mathbf{p}/m^*)\,(1+2p^2/m^*\varepsilon_g)^{-1/2}, \tag{2}$$

i.e., the electron velocity is a nonlinear function of the momentum because of the conduction band nonparabolicity. It is this factor which is responsible for the nonlinear optical phenomena in narrow-gap semiconductors of the InSb type.

In the presence of a static magnetic field $\mathbf{H}$ the equation of motion of an electron acted upon by an external high-frequency electric field $\mathbf{E}$ is

$$\dot{\mathbf{p}} = e\mathbf{E} - (1+2p^2/m^*\varepsilon_g)^{-1/2}[\mathbf{p}\omega_{c0}], \tag{3}$$

$$\mathbf{E} = \sum_j \mathbf{E}_j e^{-i\omega_j t},$$

where $\omega_{c0} = -e\mathbf{H}/m^*c$ is the cyclotron frequency of an electron at the bottom of the conduction band.

It is convenient to seek the solution of Eqs. (2) and (3) by assuming that the nonlinearity is weak and expanding the momentum as a series in powers of the electric field

$$\mathbf{p} = \mathbf{p}^{(0)} + \mathbf{p}^{(1)} + \mathbf{p}^{(2)} + \ldots, \tag{4}$$

where $\mathbf{p}^{(0)}$ is the electron momentum in the absence of the electric field (Fermi or thermal momentum in the absence of degeneracy); $\mathbf{p}^{(1)}, \mathbf{p}^{(2)}, \ldots$ are the corrections which are linear, quadratic, etc. in respect of the electric field.

The solution of Eq. (3) for the linear correction $\mathbf{p}^{(1)}$ describes the usual cyclotron resonance at the frequency $\omega_j = \omega_c$ in the $\mathbf{E}_j \perp \mathbf{H}$ geometry. This resonance may occur also for the nonlinear optical susceptibility (because of an increase in the probability of transitions to the first intermediate state), but in the case of n-type InSb illuminated with CO_2 laser radiation it occurs in fields of the order of 100 kOe (Ref. 7).

However, we can see that in addition to the nonlinearity occuring in Eq. (2) and applicable to the velocity which results in frequency mixing in zero magnetic field, there is an additional nonlinearity which occurs in the expression for the Lorentz force in Eq. (3) and which appears in a magnetic field.

In the quadratic approximation with respect to $\mathbf{E}$ the nonlinear correction to the Lorentz force in Eq. (3) contains a component at the frequency $\Delta\omega$ (whose amplitude is proportional to ω_c) and this gives to a resonance of $\mathbf{p}^{(2)}$ at the frequency $\Delta\omega = \omega_c$. The nonlinear term in the expression for the velocity results in mixing of the contribution $\mathbf{p}^{(2)}$ at the frequency $\Delta\omega$ with $\mathbf{p}^{(1)}$ at the frequency ω_1, which produces a correction to the frequency at $\omega_3 = 2\omega_1 - \omega_2$ that resonates at the frequency $\Delta\omega = \omega_c$. However, this type of cyclotron resonance of the nonlinear optical susceptibility is not the only one possible: we can also have a resonance at $\Delta\omega = 2\omega_c$.

Although these two types of cyclotron resonance can be derived in a natural manner from the quasiclassical theory,[3] it is more convenient to account for their origin and for the geometry in which they are observed by a quantum expression for the nonlinear current

$$\mathbf{j}(\omega_3) \sim \sum_{i,j,k,l} \frac{\rho_i \langle i|\mathbf{pA}_1|l\rangle\langle l|\mathbf{pA}_2^*|k\rangle\langle k|\mathbf{p}|j\rangle\langle j|\mathbf{pA}_1|i\rangle}{(\varepsilon_i-\varepsilon_l+\hbar\omega_1)\,[\varepsilon_i-\varepsilon_k-\hbar(\omega_1-\omega_2)]\,(\varepsilon_i-\varepsilon_j\pm\hbar\omega_1)}. \tag{5}$$

The summation is carried out over all the states i, j, k, and l and over all the transpositions of the photons participating in the many-photon process; however, by way of illustration only the contribution to the resonance at the frequency $\Delta\omega$ is included above (the other contributions do not resonate).

In a magnetic field the electron states i, j, k, and l are described by the Landau wave functions. It is known that for $\mathbf{E} \perp \mathbf{H}$ the operator p shifts—as a result of an electron transition—the Landau quantum number by unity $N' = N \pm 1$, whereas in the $\mathbf{E} \parallel \mathbf{H}$ case the selection rule is $N' = N$. Therefore, the energy of the state k may differ from the energy of the state i either by $2\hbar\omega_c$ or by $\hbar\omega_c$, depending whether the electric fields are directed so that $\mathbf{E}_1, \mathbf{E}_2 \perp \mathbf{H}$ or whether they have components parallel to $\mathbf{H}$.

Both types of cyclotron resonance were observed in Refs. 2 and 3; we shall consider only the purely transverse geometry $\mathbf{E}_1, \mathbf{E}_2 \perp \mathbf{H}$, which is not only simpler to realize experimentally but also more convenient for the comparison with the theory because in this case there is no superposition of two resonance contributions in the range of fields between the resonances. Going back to the quasiclassical approach, we shall now write down the final expression[3] for the nonlinear current at the frequency ω_3 in the transverse experimental geometry $(\mathbf{E}_1, \mathbf{E}_2, \mathbf{E}_3 \parallel x \perp \mathbf{H} \parallel z)$:

$$\begin{aligned} j_x(\omega_3) = &-(3/m^*\varepsilon_g)\,(ie^3E_{1x}^2E_{2x}/\omega_1^2\omega_2)\,\{[D^{-1/2}-8D^{-1/2}(p^{(0)2}/2m^*\varepsilon_g) \\ &+16D^{-1/2}(p^{(0)2}/2m^*\varepsilon_g)^2]-(16/3)\,\omega_{c0}\omega_c'[(\Delta\omega)^2 \\ &-(2\omega_c')^2]^{-1}(p_\perp^{(0)2}/2m^*\varepsilon_g)D^{-3}[1-3D^{-1}(p_\perp^{(0)2}/2m^*\varepsilon_g)]^2\}, \end{aligned} \tag{6}$$

where

$$D = 1+2p^{(0)2}/m^*\varepsilon_g, \quad \omega_c' = \omega_{c0}D^{-1/2}[1-D^{-1}(p_\perp^{(0)2}/2m^*\varepsilon_g)],$$

ω_c' is the cyclotron frequency of an electron with the momentum $\mathbf{p}^{(0)}$ in the nonparabolic case, and $p_\perp^{(0)}$ is the transverse component of the electron momentum.

The first term in the brackets in Eq. (6) represents the nonresonance contribution to the frequency mixing in the absence of a magnetic field (nonresonance background), whereas the second term describes the resonance in a magnetic field at $\Delta\omega = 2\omega_c'$.

We can find the nonlinear current by summing the velocity expression (6) over all the electrons in the

conduction band:

$$\mathbf{j}(\omega_3)=\frac{e}{4\pi^3}\int_0^\infty p^{(0)2}dp^{(0)}f_0(\varepsilon)\int_{4\pi}d\Omega\,\mathbf{v}(\omega_3). \quad (7)$$

Here, $f0(\varepsilon)$ is the Fermi distribution function.

By definition, the nonlinear optical susceptibility $\chi^{(3)}(-\omega_3,\omega_1,\omega_1,-\omega_2)$ is the coefficient relating the nonlinear current to the electric fields exciting it:

$$\mathbf{j}(\omega_3)=i\omega_3\chi^{(3)}\mathbf{E}(\omega_1)\mathbf{E}(\omega_1)\mathbf{E}(-\omega_2). \quad (8)$$

The power of the observed signal is proportional to $|\mathbf{j}(\omega_3)|^2$. It follows from the expression (6) that the resonance contribution to the nonlinear current vanishes in the absence of a magnetic field. This makes it possible to "calibrate" the experiment, i.e., to eliminate the influence of such factors as the change in the laser power from one run to another, mode structure of the laser radiation, thickness of the samples, etc.

The resonance amplitude, relative to the nonresonance spectrum, can be estimated allowing for the relaxation processes which limit the resonance amplitude. In the Wynne theory they are allowed for phenomenonologically by replacing $\Delta\omega$ with $\Delta\omega - i/\tau$, where τ is the relaxation time of the same order of magnitude as the momentum relaxation time.

In the weak nonparabolic case (when the Kane dispersion law can be expanded as a series), we have $D\approx 1$ and for the ratio ξ of the cyclotron resonance amplitude to the background we have the estimate

$$\xi\sim\frac{4}{9}\frac{\varepsilon_F}{\varepsilon_g}\omega_c\tau<1$$

for moderately high conduction electron densities.

It should be pointed out that because of the condition $\omega_1\tau$, $\omega_2\tau\gg 1$ the relaxation processes are unimportant for the nonresonance part of the nonlinear susceptibility χ_{nr}, which is a real quantity; the resonance susceptibility χ_r has real χ_r' and imaginary χ_r'' parts. It follows from the above inequality that $|\mathbf{j}_r(\omega_3)|\ll|\mathbf{j}_{nr}(\omega_3)|$; hence, the ratio of the signal powers at the frequency ω_3 in the presence and absence of a magnetic field is

$$\frac{P(\omega_3,H)}{P(\omega_3,0)}=\frac{j_{nr}^2+2\mathbf{j}_{nr}\mathbf{j}_r'+j_r'^2+j_r''^2}{j_{nr}^2}\approx 1+2\frac{\chi_r'}{\chi_{nr}}. \quad (9)$$

The real part of the resonance contribution to the susceptibility χ_r' exhibits dispersion and, consequently, if $\chi_r<\chi_{nr}$, one can expect dispersion (frequency dependence) of the radiation power near the resonance.

We shall compare the experimental results for the power $P(\omega_3)$ normalized to zero H field with the results of calculations of $1+2\chi_r'/\chi_{nr}$ based on Eq. (9).

EXPERIMENTS

A Q-switched laser emitted at two frequencies $\omega_1\approx 944$ and $\omega_2\approx 1044$ cm^{-1}, corresponding to the wavelengths 10.6 and 9.6 μ. The pulse duration was about 250 nsec and the repetition frequency was 250 Hz. Since the active-medium gain at ω_1 was higher than at ω_2, the maximum of a pulse at ω_1 was observed several tens of nanoseconds earlier than at ω_2. The pulses were made to coincide in time by introducing a dispersive element into the laser resonator: this was a NaCl wedge with a vertex angle of 1.2° (Ref. 8). This wedge initially facilitated the emission of radiation at the frequency ω_2. Rotation of the wedge made it possible to ensure that the radiation pulses at the two frequencies coincided and this produced a strong mixed radiation signal.

The expression for the output radiation at the mixing frequency is[9]

$$P_3=\frac{256\pi^2\omega_3^2P_1^2P_2(1-R_1^2)(1-R_2)}{(1+R_3)n_1^2n_2n_3c^4}(\chi^{(3)})^2\times\frac{4w_3^2}{w_1^4w_2^2}\left|\frac{e^{i(\Delta k)l}e^{-(\Delta\alpha)l}-1}{i\Delta k-\Delta\alpha}\right|^2e^{-\alpha_3 l}, \quad (10)$$

where R is the reflection coefficient; n is the refractive index; w is the size of a focused spot produced by a Gaussian beam; α is the absorption coefficient; k is the wave vector; the indices 1, 2, and 3 refer to the waves with the frequencies ω_1, ω_2, and ω_3, respectively; $\Delta\alpha=\alpha_1+(\alpha_2/2)-(\alpha_3/2)$; $\Delta k=2k_1-k_2-k_3$. It follows from Eq. (10) that, firstly, in the case of small absorption coefficients at the frequencies ω_1, ω_2, and ω_3 the optimal thickness of the sample l corresponding to the maximum output radiation at the mixing frequency is equal to the coherence length (for electron densities in n-type InSb up to about 9×10^{16} cm^{-3}) and $l\approx 1/\alpha$ at higher densities (it is assumed that $\alpha_1\approx\alpha_2\approx\alpha_3$). Secondly, the optimal ratio of the laser powers at the frequencies ω_1 and ω_2 for a constant total laser power $P=P_1+P_2$ should be 2:1. The optimal ratio of the powers P_1 and P_2 was ensured by a suitable alignment of the laser resonator and selection of the composition of the CO_2-He-N_2 gaseous mixture.

The laser radiation was focused by a BaF_2 lens onto samples which were immersed directly in liquid helium kept at $T=1.8°$ K. These samples were inside a superconducting solenoid capable of generating magnetic fields up to 75 kOe. Measurements were carried out on n-type InSb single crystals of 6×4 mm dimensions and thickness selected on the basis of the considerations mentioned above. Since a study was made of the carrier-density dependences of the position, amplitude, and line profile of the cyclotron resonance of the susceptibility $\chi^{(3)}$, it was important to select samples which had homogeneous carrier-density distributions. The electron density in our samples was determined from the Hall effect at liquid nitrogen temperature after grinding and polishing, and a correction was made for the geometric factor.[9] The homogeneity was deduced from the identity of the Hall emf's in identical but oppositely directed magnetic fields; disagreement between these two values was typical of an inhomogeneous material. The electron density in our samples ranged from 1.7×10^{16} to 2.5×10^{17} cm^{-3} and the mobility from 7×10^4 to 4×10^4 cm$^2\times V^{-1}\times$ sec^{-1}. Use was made of samples with the [100]‖H, [111]‖H orientations as well as of unoriented samples. However, since there were no effects associated with anisotropy, all the results on the oriented and unoriented samples were analyzed together.

Radiation at the mixing frequency $\omega_3=2\omega_1-\omega_2$ emerging from a sample was separated from the high-power

pump radiation transmitted by the sample; this was done using a two-step reflecting sapphire filter[10] and a prism monochromator. Then, the radiation at the mixing frequency was recorded with a Ge:Hg detector cooled with liquid helium and a pulsed synchronous detector. (The time constant of this detector ranged from a few tenths of a second to 2.5 sec.) The reference signal for the synchronous detector was the radiation reflected from one of the Brewster windows of the CO_2-laser discharge tube. The output of the synchronous detector was applied to an X-Y plotter; the second input of this plotter received a signal proportional to the magnetic field created inside the superconducting solenoid. The measurements were carried out in the Voigt experimental geometry, i.e., the wave vectors at all the frequencies $(\omega_1, \omega_2, \omega_3)$ were perpendicular to a static magnetic field H.

RESULTS AND DISCUSSION

Figure 1 shows typical magnetic-field dependences of the power $P(\omega_3)$ at the mixing frequency corresponding to the wavelength of 11.8 μ. It is clear from this figure that the dependences do indeed show dispersion in accordance with the above theory. An increase in the electron density enhances the resonance intensity and shifts the resonance toward stronger magnetic fields. In the case of a pure dispersion curve the resonance position corresponds to the point of inflection. In the case of a Lorentzian line profile the separation from the minimum to the maximum of the dispersion curve plotted against the magnetic field corresponds to the half-width of the double cyclotron resonance. The amplitude of this resonance is taken to be the separation of the minimum to the maximum of the dispersion curve plotted as a function of the power. We shall mean this type of curve when we shall consider later the position, half-width, and amplitude of the double cyclotron resonance of the nonlinear optical susceptibility. We shall consider the dependences of the position, half-width, and amplitude of the double cyclotron resonance on the Fermi energy of the conduction electrons in InSb, i.e., on the electron density. In the case of the Kane dispersion law, we have

$$\varepsilon_F = (\varepsilon_g/2)\{[1+(2\hbar^2/m^*\varepsilon_g)(3\pi^2 n)^{2/3}]^{1/2}-1\}.$$

Figure 2 shows the experimental results and the theoretical dependence of the position of the cyclotron resonance of the susceptibility $\chi^{(3)}(-\omega_3, \omega_1, \omega_1, -\omega_2)$ on the electron Fermi energy. The calculated curve is based on Eqs. (7) and (6) and it applies to magnetic fields close to the resonance value; the position of the resonance is deduced from the point of inflection of the calculated curve. Averaging of $v(\omega_3)$ over the angles was replaced by the averaging of $p_{12}^{(0)}$, and integration with respect to the momenta was carried out on a computer. In this calculation use was made of the following band structure parameters: $m^* = 0.0144 m_0$, $\varepsilon_g = 235.5$ meV (according to Pidgeon and Brown[11]). We can see that the agreement between the theory and experiment is quite satisfactory.

We have considered so far only the theory (which describes well the experimental results) that ignores the interaction between electrons, i.e., the collective plasma oscillations of the electron gas. It is known[12] that in the case of the linear cyclotron resonance in the Voigt geometry ($\mathbf{k} \perp \mathbf{H}$, where $\mathbf{k}$ is the wave vector of the laser radiation) the resonance does not occur at the cyclotron frequency ω_c corresponding to one-particle excitations (conduction electrons) but at the frequency $\omega = (\omega_p^2 + \omega_c^2)^{1/2}$ of a mixed magnetoplasma wave resulting from the interaction between the collective plasma oscillations of the electron gas and the cyclotron motion of the carriers ($\omega_p^2 = 4\pi e^2/\varkappa m^*$, where ω_p is the plasma oscillation frequency, $\varkappa$ is the permittivity and m^* is the effective electron mass at the Fermi level).

For the Fermi energy $\varepsilon_F = 40$ meV corresponding to the middle part of the range of carrier densities investigated by us, the plasma frequency is $\omega_p = 161.5$ cm^{-1}, which is considerably greater than $\Delta\omega$. It follows that the cyclotron resonance at the magnetoplasma wave frequency cannot be observed at this carrier density. However, the experimental results indicate that the nonlinear cyclotron resonance is insensitive to the plasma oscillations.

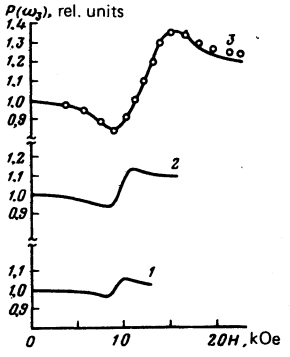

FIG. 1. Dependences of the radiation power at the mixing frequency $P(\omega_3)$ on the magnetic field applied to n-type InSb samples with various conduction electron densities n(cm^{-3}): 1) 1.96×10^{16}; 2) 4.8×10^{16}; 3) 2.4×10^{17}. The points are the results of calculations carried out allowing for the nonlinear susceptibility of the bound electrons.

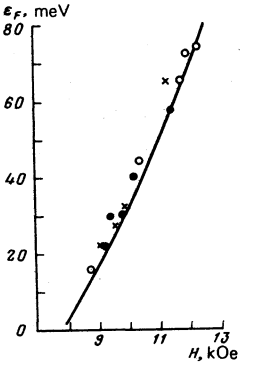

FIG. 2. Position of the cyclotron resonance of the nonlinear optical susceptibility $\chi^{(3)}(-\omega_3, \omega_1, \omega_1, -\omega_2)$ as a function of the Fermi energy of the conduction electrons in n-type InSb. The curve is calculated using Eqs. (6) and (7); the points are the experimental results: ×) [100] ∥ H; •) [111] ∥ H; ○) unoriented.

In fact, if we use the equations of motion of an electron, we can show that the nonlinear macroscopic current at the frequency $\omega_2-\omega_1$ vanishes for a material with a spherically symmetric Kane band and, consequently, it cannot excite coupled magnetoplasma oscillations so that the nonlinear cyclotron resonance corresponds to a transition between unrenormalized one-particle states and is governed by the frequency ω_c.

As mentioned above, the relaxation time τ which has to be allowed for to limit the resonance amplitude, is introduced phenomenologically in Eq. (6) by replacing $\Delta\omega$ with $\Delta\omega - i/\tau$. In Wynne's paper the relaxation time is used as one of the adjustable parameters and is varied in a fairly wide range. However, before Wynne's paper, Rustagi[13] developed a theory of the nonlinear optical susceptibility in the absence of a magnetic field by solving the Boltzmann transport equation. Rustagi showed that the correction to the distribution function due to the nonlinear force at the frequency $\omega_2-\omega_1$ does not decay at a rate given by the relaxation time τ_1 occurring in the expression for the mobility but at a rate characterized by a different time τ_2 corresponding to the relaxation of the second (and not the first) Legendre polynomial in the expansion of the nonequilibrium distribution function in terms of spherical harmonics. A similar result was obtained by Almazov and Dykman[4] in their analysis of the nonlinear susceptibility in a magnetic field. According to Rustagi,[13] in the case of elastic scattering by ionized impurities, we have

$$\frac{\tau_2(k)}{\tau_1(k)}=\frac{\ln(1+z)-z/(1+z)}{(3+6/z)\ln(1+z)-6}. \tag{11}$$

Here, k is the electron wave vector, $z=4k^2R^2$, and R is the Debye screening length

$$R=[(\pi/3n)^{1/3}(\hbar^2\varkappa/4e^2m^*)]^{1/2}, \tag{12}$$

n is the electron density, and $\varkappa$ is the permittivity. The expression (12) applies to a degenerate Fermi gas. The conduction band nonparabolicity can be allowed for approximately if the mass at the bottom of the conduction band m^* in Eq. (12) is replaced with the density-of-states effective mass (mass at the Fermi level).

In the investigated range of densities from 2×10^{16} to 2×10^{17} cm^{-3} the relaxation time deduced from the mobility varies slowly and, on the average, it is equal to 8.0×10^{-13} sec (in this estimate an allowance is again made for the dependence of the effective mass on the electron energy). In the same range of carrier densities the ratio τ_2/τ_1 is almost constant and equal to 0.53; consequently, the relaxation time of the correction to the distribution function at the frequency $\omega_2-\omega_1$ is $\tau_2\approx4.2\times10^{-13}$ sec. A calculation of τ_2 from the experimentally determined relaxation time τ_1 makes it possible to allow for any compensation of impurities in InSb.

The correction for the relaxation time has little influence on the position of the double cyclotron resonance of the susceptibility $\chi^{(3)}(-\omega_3,\omega_1,\omega_1,-\omega_2)$ and shifts the resonance by not more than 1%, which is difficult experimentally.

On the other hand, the half-width of the resonance line is related directly to the relaxation time τ_2. An analysis of the resonance function

$$1/[(\Delta\omega-i\nu_2)^2-(2\omega_c)^2],$$

where $\nu_2=\tau_2^{-1}$, we can easily show that the position of a maximum and a minimum of the dispersive part of the resonance function is governed by the conditions

$$2\omega_{c\,max,\,min}=\Delta\omega\pm\nu_2$$

and

$$\Delta H=m^*c/e\tau_2.$$

Here, ΔH is the resonance half-width defined as above; at low carrier densities (Fermi energies) it is governed only by the relaxation time (m^* is the effective mass at the bottom of the band); at higher densities, m represents the cyclotron effective mass which depends on the Fermi energy.

The experimental values of the half-width of the double cyclotron resonance of $\chi^{(3)}(-\omega_3,\omega_1,\omega_1,-\omega_2)$ are compared in Fig. 3 with the theoretical calculations on Eqs. (6) and (7). The experimental points fit well the curve calculated for $\tau=\tau_2=4.2\times10^{-13}$ sec, which conforms that the relaxation time of the double cyclotron resonance differs from the relaxation time of the momentum which occurs in the mobility.

The amplitude of the double cyclotron resonance depends not only on the relaxation time τ_2 but also on several other factors. In our experiments a CO_2 laser generated radiation at several lines of the rotational spectrum of the CO_2 molecule near the frequencies $\omega_1\approx944$ and $\omega_2\approx1044$ cm^{-1}. This operation produced several spectral lines in the range $\omega_3\approx844$ cm^{-1}, which could be observed directly or in the spin resonance spectra of the nonlinear optical susceptibility $\chi^{(3)}(-\omega_1,\omega_1,\omega_1,-\omega_2)$ (Ref. 6). This will be considered in detail later; here, we shall point out that the spin resonance spectra make it possible to determine the strongest lines corresponding to the frequency $\omega_2-\omega_1$. There are five such lines at frequencies 99.1, 100.9, 102.7, 104.5, and 106.3 cm^{-1}; their intensities (averaged over the results of several measurements) are in the ratio 0.58:0.95:1:0.95:0.58. In fact, the number of the rotational lines is greater but in theoretical calculations of the amplitude of the double cyclotron resonance and line profile only these five lines with the power weighting factors are considered. Allowance for multifrequency laser emission does not affect significantly the position

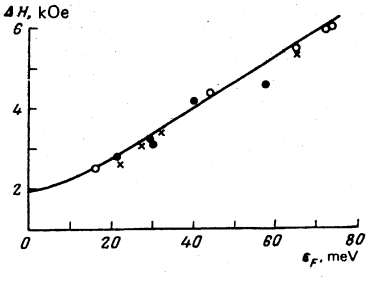

FIG. 3. Dependence of the line width of the cyclotron resonance of $\chi^{(3)}(-\omega_3,\omega_1,\omega_1,-\omega_2)$ on the Fermi energy of the conduction electrons in n-type InSb. The curve is calculated and the points are experimental results: ×) [100] ∥ H; ●) [111] ∥ H; ○) unoriented.

and half-width of the resonance, which can be explained by the strong homogeneous broadening of the resonance as a result of the relatively short relaxation time τ_2.

It follows from Eq. (6) that the resonance amplitude should be determined unambiguously by the relaxation time τ_2 (for a given carrier density n). However, it is clear from the dependence of the resonance amplitude on the electron Fermi energy (Fig. 4) that curve 1 calculated on the basis of Eqs. (6) and (7) fails to describe satisfactorily the experimental data because it gives overestimated values. This discrepancy between the calculations and experiment can be removed allowing for the contribution made to the nonresonance part of the susceptibility $\chi^{(3)}(-\omega_3, \omega_1, \omega_1, -\omega_2)$ by the bound electrons in InSb, i.e., by the electrons in the valence and deeper bands.

In the absence of a magnetic field, we have

$$P(\omega_3) \propto |\chi^{(3)}|^2 = |\chi_b + \chi_f|^2,$$

where χ_b and χ_f are the contributions of the bound and free electrons; the calculations given so far apply only to χ_f. If allowance is made for the contribution of the bound electrons, the amplitude of the double cyclotron resonance can be calculated from

$$A = A_0 \frac{\Phi_f}{\Phi_f + 2(\Phi_f \Phi_b)^{1/2} + \Phi_b}$$

where $\Phi_f \propto \chi_f^2$ and $\Phi_b \propto \chi_b^2$ represent the nonresonance background of the free and bound electrons, respectively; A_0 is the amplitude calculated from Eqs. (6) and (7); the quantity Φ_b can be regarded as an adjustable parameter. It is assumed that Φ_b is constant throughout the investigated range of free-electron densities. A comparison with the experimental results shows that a satisfactory description of the carrier-density dependence of the resonance amplitude is obtained when the contribution of the bound electrons to the nonlinear susceptibility $\chi^{(3)}$ is equal to the contribution of the free electrons of density $n = 5.5 \times 10^{15}$ cm^{-3} and of the same sign as χ_f.

The dependence of the resonance amplitude A on the Fermi energy of the conduction electrons calculated allowing for the bound-electron background is in satisfactory agreement with the experimental data (Fig. 4). Moreover, a good description is then obtained of the experimentally observed profile of the double cyclotron resonance [the calculated values of the ratio $P(\omega_3, H)/P(\omega_3, 0)$ are represented by open circles on curve 3 in Fig. 1].

The nonlinear susceptibility of the bound electrons is in agreement with the estimate given in the literature,[1,14] but its sign is opposite to the sign of $\chi_b^{(3)}$ obtained by Jha and Bloembergen[14] by the bonding orbital method. It should be noted that in the case of germanium and gallium arsenide the sign of $\chi_b^{(3)}$ also differs from that predicted by the bonding orbital model, which suggests inadequacy of the model.

Our investigation of the double cyclotron resonance of the nonlinear optical susceptibility $\chi^{(3)}(-\omega_3, \omega_1, \omega_1, -\omega_2)$ as a function of the conduction electron density in InSb thus shows that a simple quasiclassical theory based on the solution of the equations of motion of a conduction

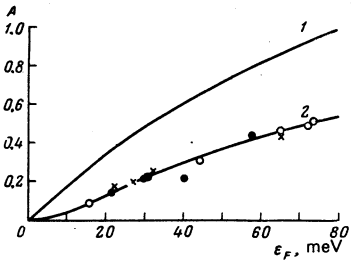

FIG. 4. Dependence of the amplitude of the cyclotron resonance of $\chi^{(3)}(-\omega_3, \omega_1, \omega_1, -\omega_2)$ on the Fermi energy of the conduction electrons in n-type InSb. Curve 1 is calculated using Eqs. (6) and (7); curve 2 is calculated allowing for the nonlinear susceptibility of the bound electrons; the points are the experimental results: ×) [100] ∥ H; ●) [111] ∥ H; ○) unoriented.

electron in a nonparabolic band with the Kane dispersion law describes satisfactorily all the experimental results if we bear in mind that the relaxation times of electrons at the difference frequency $\omega_2 - \omega_1$ is not equal to the relaxation time of the momentum of these electrons. The sign and magnitude of the nonlinear optical susceptibility $\chi_b^{(3)}(-\omega_3, \omega_1, \omega_1, -\omega_2)$ are determined for the bound electrons in InSb.

The adopted investigation method can also be applied to study the nonlinear optical susceptibility of other narrow-gap semiconductors, including those of lower symmetry than InSb.

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