Dispersion of the Nonlinear Optical Susceptibility in n-InSb†

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Using a CO_2 laser, we have observed the dispersive behavior of the third-order nonlinear optical susceptibility, $\chi^{(3)}(-\omega_3, \omega_1, \omega_1, -\omega_2)$, in n-InSb in a magnetic field. The resonances occur when the difference frequency $\omega_2 - \omega_1$ is near the cyclotron frequency, or twice the cyclotron frequency. A connection is drawn between the dispersion of $\chi^{(3)}$ and the spontaneous and stimulated Raman scattering from Landau levels. A simple semiclassical theory shows the correct polarization properties and resonance behavior of $\chi^{(3)}$ and is qualitatively in agreement with experiment.

I. INTRODUCTION

The dispersive behavior of nonlinear optical susceptibilities has been known for some time.[1] In particular, the third-order nonlinear susceptibility $\chi^{(3)}(-\omega_3, \omega_1, \omega_1, -\omega_2)$, responsible for optical-frequency mixing[2] of the form $\omega_3 = 2\omega_1 - \omega_2$, is known to have a resonant behavior not only when ω_1, ω_2, or ω_3 is close to a resonant frequency of the material system, but also when the intermediate sum frequencies $2\omega_1$ or $\omega_2 - \omega_1$ are near such a resonance. Here ω_1 and ω_2 are the frequencies of two incident light waves with $\omega_2 > \omega_1$. The resonant term at $\omega_2 - \omega_1$ which is generally superimposed on a nonresonant background, may be classified as a Raman-type susceptibility. Measured relative to ω_1, ω_2 is equivalent to the anti-Stokes frequency, and ω_3 is the Stokes frequency.[3]

We have observed the dispersive resonance of $\chi^{(3)}(-\omega_3, \omega_1, \omega_1, -\omega_2)$ at $\omega_2 - \omega_1$, in n-type InSb where the material resonance can be tuned in a magnetic field. The effect is closely related to the spontaneous and stimulated Raman effect in this material which is due to transitions between Landau levels of the conduction electrons and which has been studied extensively by Patel and coworkers.[4] But whereas the stimulated Raman scattering is related to the imaginary part of the Raman susceptibility $\chi^{(3)}(-\omega_2, \omega_1, \omega_2, -\omega_1)$, our results are described by the real part of the Raman-type susceptibility $\chi^{(3)}(-\omega_3, \omega_1, \omega_1, -\omega_2)$. This part is a superposition of the real nonresonant susceptibility associated with the nonparabolicity of the conduction band and the real part of the resonant susceptibility associated with transitions between Landau levels. Since, as we will show below, the resonant part vanishes at zero magnetic field, one may calibrate this resonant Raman-type susceptibility relative to the nonresonant part.

II. EXPERIMENT

A Q-switched CO_2 laser, of a type described earlier,[5] was used to repetitively generate pulses of radiation about 500 nsec long. It was equipped with a rotating mirror at one end and a dielectric-coated output mirror at the other end. It oscillated simultaneously on several rotational-vibrational transitions near 944, 1047, and 1082 cm^{-1}. Since the laser tube had NaCl Brewster-angle windows, the direction of linear polarization could be changed by rotating the laser tube within the cavity formed by the mirrors. The radiation was focused into the sample with an $f/8$ BaF_2 lens. The sample of n-InSb was mounted on a copper cold-finger in a KBr window-equipped Dewar which was placed between the tapered pole pieces of a 12-in. electromagnet. Temperatures of $T \approx 15\,°K$ and $T \approx 77\,°K$ and magnetic fields up to 23 kG could be obtained. New frequencies of the form $\omega_3 = 2\omega_1 - \omega_2$ were generated in the InSb due to three-wave mixing. Although the large number of closely spaced output frequencies of the CO_2 laser makes it possible to observe the dispersion directly by varying $\omega_2 - \omega_1$ and keeping the magnetic field fixed, it was much simpler to fix ω_1 and ω_2 and to sweep the magnetic field. Actually, two frequency combinations were studied: $2\times(944\ cm^{-1}) - 1047\ cm^{-1}$, and $2\times(944\ cm^{-1}) - 1082\ cm^{-1}$. The newly generated frequency components were separated from the incident laser radiation with interference filters and a $\frac{1}{4}$-m grating monochromator. These frequencies were detected with a Ge:Cu photoconductor, and the signal was averaged over many laser pulses with a boxcar integrator.

The carrier concentration of the single-crystal n-InSb sample was nominally $n = 6.6\times 10^{16}\ cm^{-3}$.

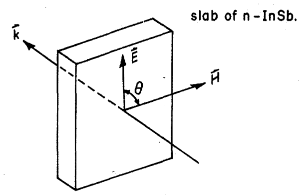

FIG. 1. Experimental configuration.

The samples were 0.5 and 2.0 mm thick and were polished on both faces. They were slightly wedged to reduce the effects of the standing-wave pattern. The incident focused intensity was kept as low as 10^5 W/cm^2 to alleviate the danger of sample heating. Since the nonlinear polarizability is due to conduction electrons in a spherical energy band, the crystallographic orientation was not significant. The coherence length[5] may be regarded as constant in the Voigt configuration[7] (direction of propagation perpendicular to the magnetic field) for this range of carrier concentration, magnetic field, and sample thickness. Our measurements were made in the Voigt configuration (see Fig. 1), and therefore any changes in the output power can be attributed to changes in $\chi^{(3)}(-\omega_3, \omega_1, \omega_1, -\omega_2)$.

III. RESULTS AND INTERPRETATION

Owing to the Brewster angle windows, the laser radiation was linearly polarized with the same polarization for all wavelengths. Under these conditions $\underline{\chi}_{nr}^{(3)}$ is real and isotropic leading to a nonlinear polarization which is parallel to the electric field vectors of the incident radiation. However, $\underline{\chi}_{res}^{(3)}$ is neither isotropic nor real. We must write the power generated at ω_3 as

$$I(H) = \text{const}\times \left| (\underline{\chi}_{nr}^{(3)} + \underline{\chi}_{res}^{(3)}) \vdots \vec{E}(\omega_1)\vec{E}(\omega_1)\vec{E}^*(\omega_2)\right|^2$$

$$= \text{const}\times\left[\left| (\underline{\chi}_{nr}^{(3)} + \underline{\chi}'^{(3)}_{res}) \vdots \vec{E}(\omega_1)\vec{E}(\omega_1)\vec{E}^*(\omega_2)\right|^2 + \left|\underline{\chi}''^{(3)}_{res} \vdots \vec{E}(\omega_1)\vec{E}(\omega_1)\vec{E}^*(\omega_2)\right|^2\right] . \quad (1)$$

If, in addition $\underline{\chi}_{res}^{(3)} \ll \underline{\chi}_{nr}^{(3)}$, the normalized power may be written as

$$\frac{I(H)}{I(0)} \cong 1 + 2\frac{[\underline{\chi}_{nr}^{(3)} \vdots \vec{E}(\omega_1)\vec{E}(\omega_1)\vec{E}^*(\omega_2)][\underline{\chi}'^{(3)}_{res} \vdots \vec{E}(\omega_1)\vec{E}(\omega_1)\vec{E}^*(\omega_2)]}{|\underline{\chi}_{nr}^{(3)} \vdots \vec{E}(\omega_1)\vec{E}(\omega_1)\vec{E}^*(\omega_2)|^2}$$

$$\equiv 1 + 2(\underline{\chi}'^{(3)}_{res}/\underline{\chi}_{nr}^{(3)})_{eff} . \quad (2)$$

Figures 2 and 3 show the results at $T = 15\,^\circ$K. The steplike S shape of the curves is typical of an inhomogeneously broadened resonant dispersion such as might be described by Eq. (2). Figure 1(b) shows a resonance at 9.5 kG corresponding to $\omega_2 - \omega_1 = 2\omega_c$ and at 19 kG corresponding to $\omega_2 - \omega_1 = \omega_c$, where ω_c is the cyclotron frequency. For this choice of difference frequency it was not possible to reach sufficiently high magnetic fields to observe the spin-flip transition.[4] The polarization dependence is the same as one would predict from the results of spontaneous Raman scattering.[4] In Fig. 2, the resonance for $2\omega_c$ appears at 13 kG, an increase from the previous case due to the higher difference frequency. At $T = 77\,^\circ$K the structure had a slightly more "washed-out" appearance but was otherwise unchanged. Each curve is the average of several sweeps taken to reduce drift errors.

Owing to the nonparabolicity[8] of the conduction band in InSb, each electron has a different effective mass depending upon its average momentum.

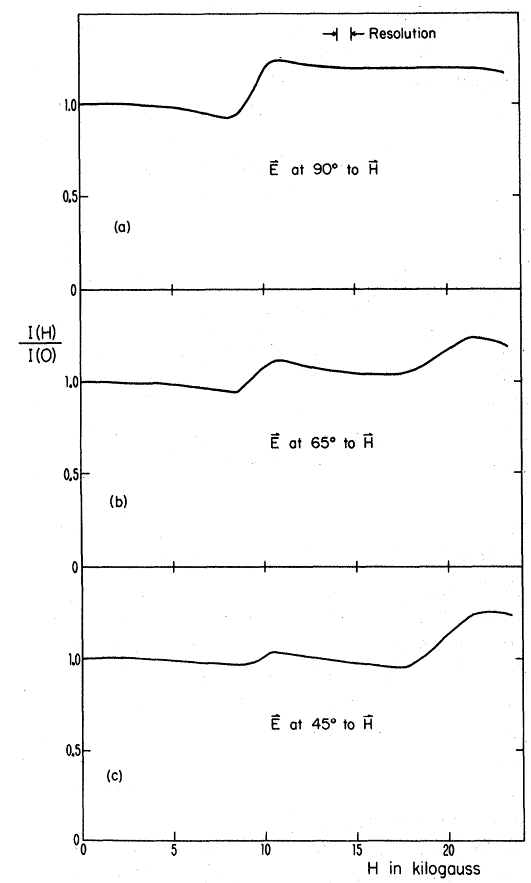

FIG. 2. Output power at $2\times(944\text{ cm}^{-1}) - 1047\text{ cm}^{-1}$ as a function of magnetic field for three different polarizations. E is the electric field vector and H the magnetic field vector. The $\omega_c = \omega_2 - \omega_1$ resonance at 19 kG vanishes when E is perpendicular to H. Both resonances are absent when E is parallel to H. The curves are normalized at $H = 0$ and the accuracy in the experimental power data at the high-field end is about 4%.

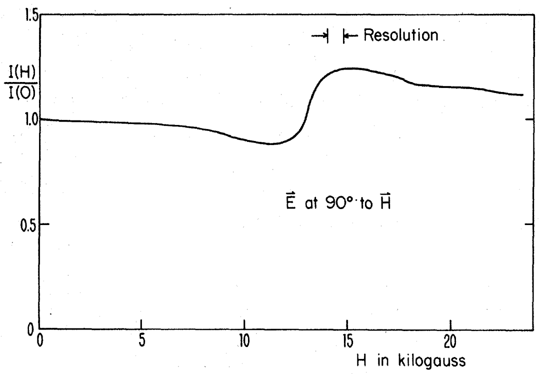

FIG. 3. Output power at $2\times(944\ \mathrm{cm}^{-1}) - 1082\ \mathrm{cm}^{-1}$ as a function of magnetic field for E perpendicular to H. The position of the $2\omega_c=\omega_2-\omega_1$ resonance is different from Fig. 1 owing to the higher difference frequency.

This is the source of the inhomogeneous broadening of the cyclotron resonance. By comparison, the linewidth caused by the finite lifetime of electron states is expected to be smaller.[7, 7a]

IV. THEORY

We are able to obtain qualitative agreement with our data by using a simple semiclassical treatment for the nonparabolic conduction band in InSb. This model was first used by Wolff and Pearson[9] to explain the large observed three-wave mixing in InSb and InAs,[2] in the absence of a magnetic field. Lax, Zawadski, and Weiler[10] extended it to include the magnetic field, but they ignored terms in the nonlinear velocity which were smaller than the terms they considered by a factor of p^2/m^*E_g, where p is the conduction-electron momentum in the absence of an applied optical electric field. We have repeated Lax *et al.*'s calculation but have included these terms and taken careful account of the components of the electron momentum which are zeroth, first, and second order in the electric fields. The resulting expression simplifies considerably under the following additional assumptions: (a) $\omega_c \ll \omega_1, \omega_2, \omega_3$, (b) $E_1 \| E_2$, and (c) the beams propagate normal to the magnetic field. Then we find that

$$\left(\frac{\chi'^{(3)}_{\mathrm{res}}}{\chi^{(3)}_{\mathrm{nr}}}\right)_{\mathrm{eff}} = \frac{16}{3}\,\omega_c^2\sin^2\theta\left\langle \frac{\sin^2\theta}{4(\omega_c-\Delta\omega')^2-(\omega^2-\omega^1)^2}\left(\frac{p_x^2+p_y^2}{2m^*E_g}\right) + \frac{2\cos^2\theta}{(\omega_c-\Delta\omega'')^2-(\omega_2-\omega_1)^2}\left(\frac{p_z^2}{2m^*E_g}\right)\right\rangle_{\mathrm{FS}} \qquad (3)$$

where $\langle\,\rangle_{\mathrm{FS}}$ means average over the Fermi sphere. The inhomogeneous broadening is described by the terms in the denominators:

$$\Delta\omega' = \omega_c\left(\frac{\frac{3}{2}(p_x^2+p_y^2)+p_z^2}{m^*E_g}\right), \qquad \Delta\omega'' = \omega_c\left(\frac{2(p_x^2+p_y^2)+p_z^2}{m^*E_g}\right). \qquad (4)$$

The cyclotron frequency is given by $\omega_c = eH/m^*c$, where m^* is the effective mass at the band edge and H, the magnetic field, is taken along the $\hat{z}$ direction. The polarization dependence is described by θ, the angle between the electric field vectors and the magnetic field. E_g is the band gap. The parameters for this sample of n-InSb give a value of about $\frac{1}{6}$ for $p_F^2/2m^*E_g$, where p_F is the electron momentum at the Fermi level.

This would predict an inhomogeneous broadening $\Delta\omega/\omega_c$ approximately equal to $\frac{1}{3}$ in Eq. (4). Substitution of these values in Eq. (3) then yields good agreement with the observed curves in Figs. 1 and 2. In particular, the asymptotic change for large magnetic fields well above resonance is in agreement with theory.

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[7a] *Footnote added in proof.* Recent computer calculations show that the smaller homogeneous broadening still has an appreciable effect on the shape of the resonances. These computer calculations are in good agreement with the experimental curves.

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