Light Scattering by Plasmons and Landau Levels of Electron Gas in InAs

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We report the observation of inelastic scattering of 10.6 μ radiation by collective and single-particle cyclotron excitations of mobile carriers in InAs. The observed cross sections of $\sim 10^{-25}$ cm^2 sr^{-1} for both the plasmon and the Landau-Raman $\Delta l=1$ and $\Delta l=2$ scattering processes are in agreement with calculations. Raman scattering of light by optic phonons is found to be at least a factor of 100 weaker than the plasmon scattered light. Effects of magnetic field on plasmon scattering are also described.

INELASTIC scattering of 10.6 μ radiation from a CO_2 laser provides a powerful technique for studying properties of a mobile electron gas in narrow-band-gap semiconductors such as InAs, InSb, and PbTe. The frequency spectrum of such scattered light is a measure of the energies of both the collective and single-particle excitations. We report the observation of inelastic scattering by plasmons as well as by single-particle Landau level excitations[1] in InAs. In contrast to the earlier experiments by Mooradian and Wright[2] on III–V compounds with band gaps larger than $\sim$1 eV, Raman scattering of light by longitudinal and transverse optic phonons could not be observed. The intensity and polarization of the scattered light were independent of orientation of the crystal lattice with respect to the incident and scattered light directions. These observations indicate that the scattering from the collective modes described here is caused by the plasmons of the electron gas independent of the lattice.

In order to observe the Raman scattering from collective excitations of a degenerate electron gas in InAs, the momentum transfer q ($\mathbf{q}=\mathbf{k}_1-\mathbf{k}_2$, where subscripts 1 and 2 refer to incoming laser and outgoing scattered light, respectively) must be much smaller than the Fermi-Thomas screening wave vector q_{FT}.[3] The pure-plasmon differential scattering cross section is given by

$$d\sigma_p/d\Omega \cong r_0^2(q/q_{FT})^2(\boldsymbol{\varepsilon}_1\cdot\boldsymbol{\alpha}\cdot\boldsymbol{\varepsilon}_2). \tag{1}$$

[An additional factor of the order of $\hbar\omega_p/E_F$ has been omitted for the cross section of Eq. (1) since it is of the order of unity for InAs.]

Equation (1) has no enhancement factor,[4] since the laser photon energy $\hbar\omega_0$ is about one third the band-gap energy E_G. Here r_0 is the classical electron radius ($\sim 2.8\times10^{-13}$ cm) for free electrons, $\boldsymbol{\varepsilon}_{1,2}$ are the polarizations of the incoming and outgoing waves, respectively, and $\boldsymbol{\alpha}$ is the reciprocal effective mass tensor, which is diagonal with components of the order of 40 for mobile electrons in InAs. Using the 10.6 μ radiation, $q/q_{FT}\approx 1/30$ for InAs. The observed plasmon scattering cross section of $\sim 10^{-25}$ cm^2 sr^{-1} agrees with that calculated from Eq. (1).

For single-particle scattering from electrons in Landau levels, the cross sections are finite because of the nonparabolicity of the conduction band. For an incoming wave linearly polarized in the x direction, the cross sections are of the order of[1,4]

$$d\sigma_{\Delta l=2}/d\Omega \approx r_0^2\alpha^2(\hbar\omega_c/E_G)^2(\epsilon_{1x}^2)(\epsilon_{2x}^2+\epsilon_{2y}^2) \tag{2}$$

and

$$d\sigma_{\Delta l=1}/d\Omega \approx r_0^2\alpha^2(\hbar\omega_c E_F/E_G^2)(\epsilon_{1x}^2\epsilon_{2z}^2), \tag{3}$$

where l is the Landau-level quantum number, $\omega_c = eB_z/m^*c$ is the cyclotron frequency, B_z is the z-directed magnetic field, and E_F is the Fermi energy. For $B_z=55$ kOe, these cross sections are also about 10^{-25} cm^2 sr^{-1}.

The CO_2 laser produced peak power of about 25 kW in 200 nsec pulses at a repetition rate of 120 Hz. The experimental geometry used a parabolic reflector[1] to gather about 0.1 sr of light scattered at 90° to the incident beam. Scattered light was analyzed with a 0.25-m grating spectrometer, and a Ge:Cu photoconductor was used as detector. A gated integrator was used to average signals over times of the order of 300 sec in order to observe scattered light power as small as 10^{-9} W. Inelastically scattered light from the sample was of the order of 10^{-7} W, and after spectrometer analysis, a signal-to-noise ratio of 5:1 could be obtained for both the plasmons and the Landau level scattering processes. In addition, a multichannel analyzer was used to average several spectrometer sweeps to improve the signal-to-noise ratio by a factor of 2. InAs single crystals were in the form of polished bars 2×2×10 mm, maintained at a temperature of about 30°K. The magnetic field and the propagation vector of incident radiation were along the long dimension of the sample. Electron concentration was in the range of 10^{16} to 10^{17} cm^{-3}, and electron mobilities were of the order of 25 000 cm^2 V^{-1} sec^{-1} at 77°K. Free-carrier absorption at longer wavelengths prevented using higher electron concentration materials.

Figure 1(a) shows a typical spectrometer trace of the light scattered by plasmons in InAs with 5.5×10^{16} electrons cm^{-3}. It is interesting that no scattered light

[1] R. E. Slusher, C. K. N. Patel, and P. A. Fleury, Phys. Rev. Letters **18**, 77 (1967).
[2] A. Mooradian and G. B. Wright, Phys. Rev. Letters **16**, 999 (1966).
[3] P. M. Platzman, Phys. Rev. **139**, A379 (1965).
[4] P. A. Wolff, Phys. Rev. Letters **16**, 225 (1966).

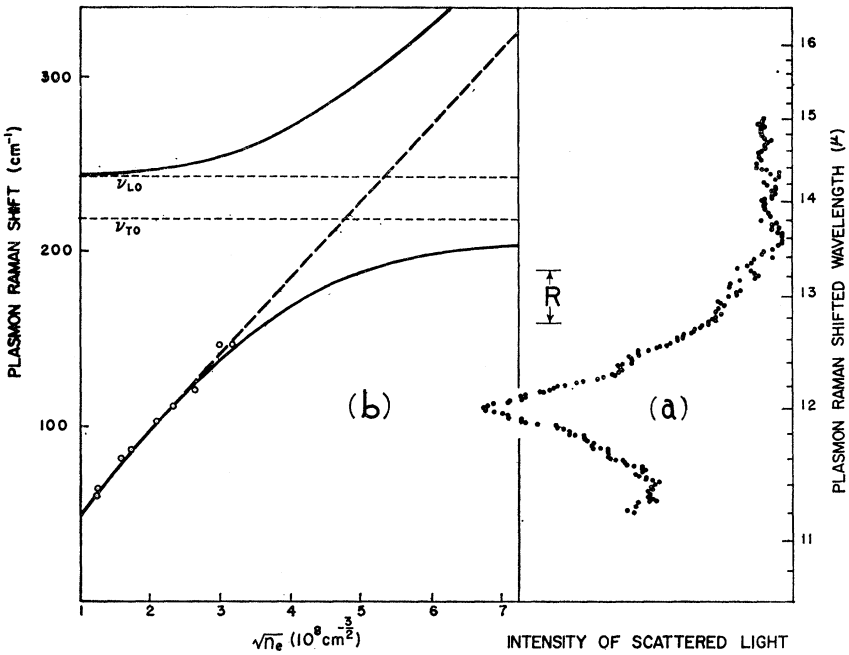

FIG. 1. (a) shows a typical spectrometer trace of light scattered by plasmons in n-InAs with $n_e \approx 5.5\times10^{16}$ cm^{-3}. Notice the absence of scattered light peaks at 13.8 and 14.2 μ, corresponding to Raman scattering by TO and LO phonons. Spectrometer resolution $\sim$30 cm^{-1}. (b) shows carrier concentration dependence of frequency shift of Raman scattered light. Solid line is the calculated resonance frequency at $\mathbf{q}=0$, including plasmon-phonon interaction. The change in effective mass caused by the nonparabolic conduction band produces the slight curvature for calculated plasma frequencies below 100 cm^{-1}.

was observed at 13.8 or 14.2 μ, corresponding to Raman scattering of light by the transverse and the longitudinal optic phonons, respectively. Even for the lowest carrier concentration InAs samples, where the absorption coefficient at 14 μ was <3 cm^{-1}, allowing us to observe Raman scattering efficiency[5] as small as 10^{-12} sr^{-1} cm^{-1}, we did not detect Raman scattering from optic phonons. We estimate our plasmon scattered signal to be $\gtrsim$100 times the phonon scattered signal. Mooradian and Wright's results[2] for GaAs indicate nearly equal intensities for both the plasmon and phonon scattered light when plasmons and phonons are separated so that nearly pure plasmon scattering is observed. Pure plasmon scattering cross section for GaAs, including the band-gap enhancement factor, is expected to be $\sim$10 times the pure plasmon scattering cross section for InAs. This difference of a factor of $>$1000 between the plasmon to phonon scattering ratios for InAs and GaAs may arise from the frequency and band-gap dependence of the phonon scattering efficiency.

The linewidth of 30 cm^{-1} seen in Fig. 1(a) is nearly the instrument linewidth. Measurement with narrower instrument width indicated a linewidth of about 5–10 cm^{-1} for the plasmon scattered light. The electron collision time τ_e is expected to broaden the plasmon scattered light by[6]

$$\Delta\nu_p \approx 1/4\pi\tau_e, \qquad (4)$$

[5] We define Raman scattering efficiency as the ratio of number of scattered photons per unit solid angle and unit crystal length to the number of incident photons measured during a unit time.

[6] H. Fröhlich and H. Pelzer, Proc. Phys. Soc. (London) **A68**, 525 (1955).

which is $\sim$7 cm^{-1}, using τ_e obtained from measured electron mobility. The electron collision time appears to be a major contribution to the observed plasmon linewidth.

Figure 1(b) shows the carrier concentration dependence of the frequency shift of the plasmon scattered light. The experimental points seem to follow the calculated resonance frequency at $\mathbf{q}\approx0$ with no plasmon-phonon interaction. Since, for our experimental conditions, $qv_F/\omega_p \ll 1$, there is no frequency shift caused by the plasmon dispersion (see, for example, Pines[7]). The plasmon scattering cross section is observed to be in the range of 10^{-25} cm^2 sr^{-1}, as expected for pure plasmon scattering from Eq. (1). The plasmon scattered light was polarized (better than 10:1) in the same direction as the incident 10.6 μ laser radiation, in agreement with the polarization dependence also described in Eq. (1). As the carrier concentration increased towards the region of plasmon-phonon interaction described by Varga,[8] and shown on Fig. 1(b), the observed signal strength decreased and finally vanished, indicating a decrease in the scattering cross section.

On application of a magnetic field B_z, the plasma mode is expected to mix with the cyclotron mode, giving rise to hybrid resonances.[9,10] For observation of light scattering at 90° to the incident beam, the momentum transfer $\mathbf{q}$ vector selects a component of the density

[7] D. Pines, *Elementary Excitations in Solids* (W. A. Benjamin, Inc., New York, 1963), p. 147.

[8] B. B. Varga, Phys. Rev. **137**, A1896 (1965).

[9] W. P. Allis, S. J. Buchsbaum, and A. Bers, *Waves in Anisotropic Plasmas* (MIT Press, Cambridge, Mass., 1963).

[10] N. D. Mermin and E. Canel, Ann. Phys. (N.Y.) **26**, 247 (1964).

fluctuations at 45° to B_z, with the resonance frequencies given by

$$\omega_{\pm}=\{\tfrac{1}{2}[\omega_p^2+\omega_c^2\pm(\omega_p^4+\omega_c^4)^{1/2}]\}^{1/2}, \qquad (5)$$

which are shown by dashed lines on Fig. 2. We can follow the high-frequency hybrid resonance (ω_+) up to ~10 kOe, where the change in frequency is small compared to linewidth. At magnetic fields where the frequency shift becomes sizeable, the observation of the upper hybrid mode is obscured by the $\Delta l=1$ and $\Delta l=2$ Landau-Raman scattering described below. At maximum field (55 kOe), the plasmon scattered light was absent from its zero-field position, indicating a shift of the scattered light towards longer wavelengths. Inelastic scattering from the low-frequency hybrid mode (ω_-) is not expected to be obscured by Landau-Raman scattering, but could not be observed.

Figure 2 shows the positions of $\Delta l=1$ and $\Delta l=2$ Landau-Raman scattered light as a function of the magnetic field for $n_e=5.5\times10^{16}$ cm^{-3} concentration InAs sample. From the average slopes, we obtain $m^*=0.028$ and 0.030 for the $\Delta l=1$ and $\Delta l=2$ transitions, respectively. These effective mass values agree with those obtained from interband magnetoabsorption measurements.[11] For all magnetic fields, linewidths of the $\Delta l=1$ and $\Delta l=2$ scattered light were ~20 cm^{-1}. The expected electron collision time broadening is given by

$$\Delta\nu_{\Delta l=1,2}\approx 1/2\pi\tau_e, \qquad (6)$$

which is ~15 cm^{-1}, indicating that the linewidth contribution from averaging over various Landau levels of the nonparabolic band is small in contrast with the results obtained in InSb.[1]

Raman scattered light involving an electron spin flip[12] was also observed for $B=55$ kOe at a frequency shift of 35–40 cm^{-1}. This frequency shift yields an effective g value of 14 ± 1 for the conduction electrons in InAs, which again compares favorably with that obtained from interband magnetoabsorption measurements.[11] Elastically scattered laser light could not be discriminated against for frequency shifts of less than 30 cm^{-1}. [*Note added in proof.* Recent measurements of Raman scattered light involving electron spin flip with B up to 100 kOe yield g values which vary from 14.5 at ~50 kOe to 13.5 at ~100 kOe as expected for the nonparabolic conduction band. Linewidths of the spin-flip scattered light were of the order of 1 cm^{-1}.]

The intensity of Landau-Raman scattered light was nearly the same as that for the plasmon scattered light, corresponding to scattering cross sections of about 10^{-25} cm^2 sr^{-1}, as expected from Eqs. (2) and (3). Intensities of $\Delta l=1$ and $\Delta l=2$ scattered radiation corrected for

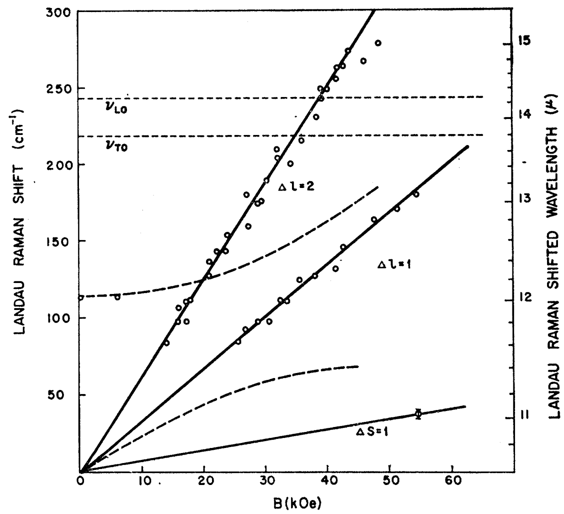

FIG. 2. Plot of frequency shift for $\Delta l=1$ and $\Delta l=2$ Landau-Raman scattered light observed for $n_e=5.5\times10^{16}$ cm^{-3} concentration InAs. Average slope for $\Delta l=1$ transition corresponds to $m^*=0.028$, and for $\Delta l=2$, the average slope gives $m^*=0.030$. Slope for the spin transition $\Delta s=1$ gives an effective g-value of 14 ± 1. Also shown is the position of low-field plasmon scattered light for the same sample. The dashed curves show hybrid plasma resonances given by Eq. (6).

absorption[13] in InAs remained nearly constant as the magnetic field changed from 0 to ~30 kOe, and the intensities decreased by a factor of 2 when the magnetic field increased from 30 to 55 kOe. As in InSb,[1] the position of Fermi level[14] with respect to various Landau levels could account for the absence of linear and quadratic field dependence of the $\Delta l=1$ and $\Delta l=2$ scattering cross sections respectively predicted for a single pair of Landau levels.

In conclusion, the small electron effective mass and low Raman scattering efficiency for the optic phonons make the electron gas in InAs an interesting subject for light scattering studies. These experiments provide a set of parameters which is complementary to those obtained by Mooradian and Wright[2] for the study of the light scattering from coupled plasmon-phonon systems. Single-particle scattering provides an independent measurement of m^* and effective g value. With improved detection capability, it whould be possible to study light scattering from hybrid plasma modes because the expected frequency shift is greater than the plasmon linewidth.

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[11] C. R. Pidgeon, D. L. Mitchell, and R. N. Brown, Phys. Rev. **154**, 737 (1967).

[12] Y. Yafet, Phys. Rev. **152**, 858 (1966).

[13] Absorption coefficient for InAs with $n_e\cong5.5\times10^{16}$ cm^{-3} was measured to be $\sim2\times10^{-2}\,\lambda^{2.8}$ cm^{-1} (λ in microns).

[14] Fermi level for InAs with $n_e\cong5.5\times10^{16}$ cm^{-3} is ~155 cm^{-1}; thus, at $B=30$ kOe, the $l=2$ level is just rising above the Fermi sea.