[5]P. O. Löwdin, J. Chem. Phys. 18, 365 (1950).
[6]E. Feenberg and C.-W. Woo, Phys. Rev. 137, A391 (1962); C.-W. Woo, Phys. Rev. 151, 138 (1966); H.-T. Tan and E. Feenberg, Phys. Rev. 176, 370 (1968).
[7]Strictly this is true only if one retains the leading order in the concentration.
[8]Same results can be obtained in the response function theory.
[9]T. Davison and E. Feenberg, to be published; T. Davison, thesis, Washington University, 1968 (unpublished).

RAMAN SCATTERING BY POLARITONS IN PRESENCE OF ELECTRON PLASMA IN GaAs

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The effects of electron plasma and magnetic field on the polaritons in GaAs are studied by Raman scattering at small angles.

A polariton is a combination state[1,2] comprised of a propagating photon and a transverse-optical (TO) phonon having nearly the same momenta and energies in a crystal. Raman scattering from semiconductors and insulators is a new and accurate probe of such mixed excitations.[3-5] In the past, crystals with essentially no free carriers have been investigated. The character of photon propagation is significantly modified when the effects of an electron (or hole) plasma are included.[6] Below the plasma frequency, ω_p, no photon propagation is possible since the propagation vector is purely imaginary. However, above ω_p, propagation of a mixed excitation consisting of photon, plasmon, and phonon is possible in the region where wave vectors, q, and frequencies, ν, or the elementary excitations are nearly equal. The dispersion relation for this excitation is similar to that of a polariton but is modified by the presence of the plasma. This paper reports a study of these mixed modes by small-angle Raman scattering in GaAs. Previously Raman scattering from a coupled plasmon-LO-phonon system[7] has been reported at large angles where the coupling between the photon and plasmon-phonon system cannot be observed because of large q. In addition, this Letter reports the effect of a magnetic field on these mixed excitations.

In absence of free carriers, the longitudinal dielectric constant is given by[8]

$$\epsilon(\omega) = \epsilon(\infty) + \frac{\epsilon(0)-\epsilon(\infty)}{1-(\omega/\omega_{\mathrm{TO}})^2}, \qquad (1)$$

where $\epsilon(\infty)$, $\epsilon(0)$ are the high-frequency and static dielectric constants and ω_{TO} is the TO-phonon frequency. In the presence of an electron plasma with a density n cm^{-3}, the dielectric constant can be modified by a simple Drude term[9] so that

$$\epsilon(\omega) = \epsilon(\infty) + \frac{\epsilon(0)-\epsilon(\infty)}{1-(\omega/\omega_{\mathrm{TO}})^2} - \frac{\epsilon(\infty)\omega_p^{\,2}}{\omega(\omega+i\omega_0)}, \qquad (2)$$

where $\omega_p = [4\pi ne^2/m^*\epsilon(\infty)]^{1/2}$ is the plasma frequency and ω_0 is the characteristic relaxation frequency of plasma given by $\omega_0 = 1/\tau_0$ (τ_0 is the average collisional relaxation time for the electrons). In the absence of a magnetic field, the longitudinal and the transverse modes are separable and are given by

$$\epsilon(\omega) = 0 \quad \text{for longitudinal modes} \qquad (3)$$

and

$$\omega^2 = q^2c^2/\epsilon(\omega) \quad \text{for transverse modes.} \qquad (4)$$

The peaks in Raman scattering occur at frequencies and wave vectors given by solutions of Eqs. (3) and (4) as shown in Fig. 1 for $n=0$, and 2.9×10^{17} cm^{-3} for GaAs with a dc mobility of 4500 cm^2 V^{-1} sec^{-1}. Previously measured values of $\epsilon(\infty)=10.719$, $\omega_{\mathrm{LO}}=291.9$ cm^{-1}, $\omega_{\mathrm{TO}} = 268.6$ cm^{-1}, and $m^*=0.073m_0$ were used.[10,11] Note that with no carriers, photon couples with TO phonon when their energies and momenta are nearly equal and give the simple polariton dispersion relation investigated by a number of authors. Addition of carriers is seen to result in a coupling between the longitudinal plasmon and phonon[7] and a modification of the polariton dispersion relation. Light scattering using a 1.06-μ source from these modes, described below, is restricted by energy-momentum conservation considerations to lie along the dotted lines shown for various scattering angles.

In the presence of a magnetic field such that

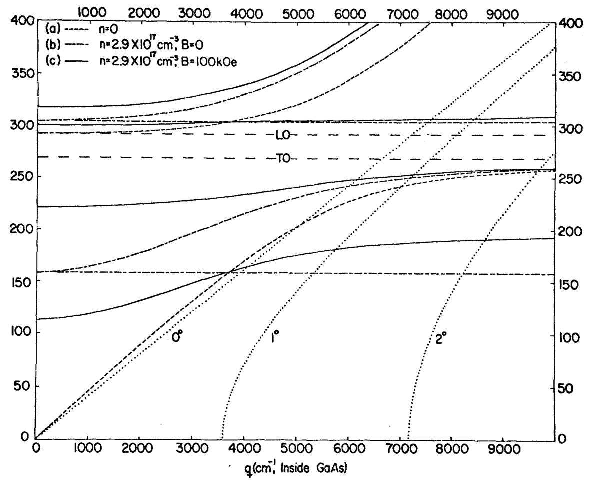

FIG. 1. Theoretical dispersion curves for polaritons in GaAs. (a) Semi-insulating, (b) $n=2.9\times10^{17}$ cm^{-3}, $B=0$, (c) $n=2.9\times10^{17}$ cm^{-3}, $B=100$ kOe. LO and TO phonons for large q and no carriers are shown as dashed lines.

$\vec{q}\perp\vec{B}$, the plasmon mode is no longer purely longitudinal because of $\vec{v}\times\vec{B}$ terms ($\vec{v}$ is the electron velocity) and the separation of the longitudinal and the transverse modes is not possible. The scattering in this case arises from density fluctuations as well as from phonon contributions. Light scattering from density fluctuations accompanying these mixed cyclotron-plasmon-phonon-photon system can be best described by[12,13]

$$\frac{d^2\sigma}{d\Omega d\omega}=\left(\frac{\omega_s}{\omega_0}\right)^2\left(\frac{\hbar}{\pi e^2}\right)[n(\omega)+1]\times\operatorname{Im}[\vec{q}\cdot\mathcal{E}^{-1}\cdot\vec{q}], \quad (5)$$

where ω_s, ω_0 are the scattered and incident photon frequencies, $n(\omega)$ is the Bose-Einstein occupation number, and

$$\mathcal{E}=\frac{q^2c^2}{\omega^2}\left(I-\frac{\vec{q}\vec{q}}{q^2}\right)-\epsilon \quad (6)$$

with I the unit tensor and ϵ the complete dielectric tensor. The free-carrier contribution to ϵ is given by simple Drude theory.[14] The lattice contribution is diagonal and is given by Eq. (1). The solution of Eq. (5) is shown in Fig. 1 for $B=100$ kOe for the same GaAs parameters as without B. Note that the partially transverse hybrid plasmon couples to the photon to give an additional splitting, the plasmariton,[13] which is independent of phonons. Equation (5) gives the correct positions of the scattering peaks, but to obtain the correct relative intensities along various branches of the dispersion curve one must include scattering mechanisms other than the density fluctuations such as the phonon-scattering mechanisms. Work towards this theoretical goal is in progress. The interaction of the hybrid plasmon with Bernstein modes is neglected since the interaction is proportional to[15] $q/q_D\approx10^{-3}$ for these experiments (q_D is the Debye wave vector for the plasma).

Experiments were carried out by Raman scattering at 1.06 μ using a 2-W neodymium-doped yttrium aluminum garnet laser operated in a low-order transverse mode of the cavity to obtain a beam divergence of less than 2 mrad. Near-forward Raman scattering was observed with apertures such that $\vec{q}\perp\vec{B}$ and was analyzed with a tandem spectrometer. ($\vec{q}=\vec{q}_{sc}-\vec{q}_{inc}$ where $\vec{q}_{sc}$ = wave vector of anti-Stokes scattered light and $\vec{q}_{inc}$ is the wave vector of the incident light.) Scattering angles from 0.3° to 1.2° (inside GaAs sample) were investigated. In order to obtain good signal-to-noise ratio the angular width of the aperture was ~0.05°. Spectrometer resolution was ~5 cm^{-1}. GaAs samples with carrier concentrations ~10^{18} cm^{-3} (semi-insulating sample), 6.7×10^{16}, 1.4×10^{17}, and 2.9×10^{17} cm^{-3} were investigated at room temperature. The dc mobility varied from 3000 to 4500 cm^2 V^{-1} sec^{-1}. Sample orientation in all cases was $\vec{q}_{inc}\parallel\langle111\rangle$ where $\vec{q}_{inc}\perp\vec{B}$.

The experimental results are shown in Fig. 2 for the four different carrier concentrations. It can be seen that the agreement between the theoretical curves calculated above and the experimental points is excellent. This verifies that the values of $\epsilon(\infty)$, ω_{LO}, ω_{TO}, and m^* used in calculations are correct. Figure 2 also shows the experimental points for the same four samples with $B=100$ kOe. As expected, the simple polariton dispersion curve for the semi-insulating sample remains unchanged while the results for the doped samples show the shifts in agreement with the calculations described earlier. This suggests that the Drude theory is adequate for the present experiments.

The cross section for scattering from density fluctuations is proportional to $(q/q_D)^2$. Scaling from previous large-angle plasmon-scattering results[7] shows that for near-forward scattering the cross section is approximately 10^{-5} times the q-independent phonon contributions. Thus the density-fluctuation contribution to the polariton scattering strength (in the magnetic field case) is too small to be observed, and the observed scattering arises primarily from the pho-

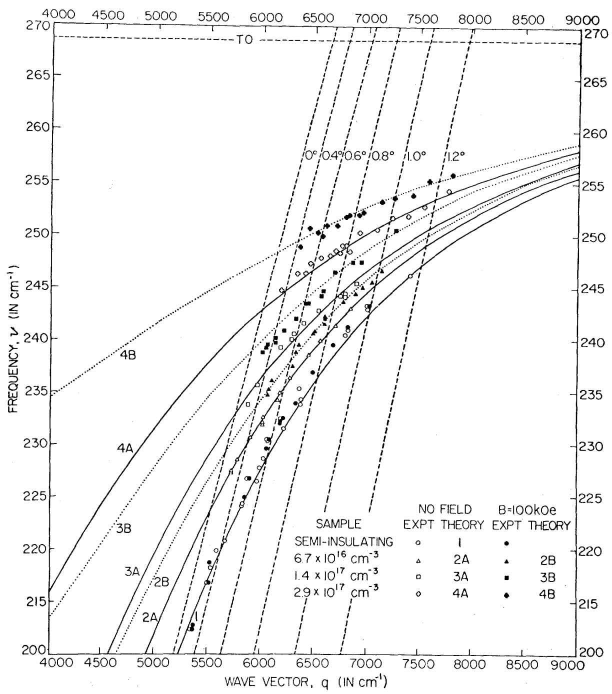

FIG. 2. Experimental and theoretical dispersion curves for peaks of light scattering in GaAs with and without carriers and magnetic field.

non contributions which are expected to decrease as the frequency of the excitation departs from the phonon frequency. The 50-cm^{-1} plasmariton splitting shown in Fig. 1 is larger than the observed plasmon linewidth of ~30 cm^{-1} and should be easily resolved. Thus it is not entirely clear why the lower mode was not observed.

In conclusion, we have investigated the influence of an electron gas and a magnetic field on the polaritons in semiconductors. Our experimental dispersion relation is in agreement with the present calculations and is significantly different from the previously considered interaction including only the phonons and the electromagnetic wave. Thus we have for the first time investigated the total polariton in semiconductors.

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[1]U. Fano, Phys. Rev. 103, 1202 (1956).

[2]J. J. Hopfield, Phys. Rev. 112, 1555 (1958).

[3]C. H. Henry and J. J. Hopfield, Phys. Rev. Letters 15, 964 (1965).

[4]S. P. S. Porto, B. Tell, and T. C. Damen, Phys. Rev. Letters 16, 450 (1966).

[5]J. F. Scott, L. E. Cheesman, and S. P. S. Porto, Phys. Rev. 162, 834 (1967).

[6]See, for example, O. P. Madelung, Physics of III-V Compounds (John Wiley & Sons, Inc., New York, 1964), pp. 79-83.

[7]A. Mooradian and G. B. Wright, Phys. Rev. Letters 16, 999 (1966), and Solid State Commun. 4, 431 (1966).

[8]See, for example, M. Born and C. Huang, Dynamical Theory of Crystal Lattices (Clarendon Press, Oxford, England, 1954).

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

[10]K. G. Hambleton et al., Proc. Phys. Soc. (London) 77, 1147 (1961); C. K. N. Patel, Phys. Rev. Letters 16, 613 (1966).

[11]H. Piller, J. Phys. Soc. Japan Suppl. 21, 206 (1966).

[12]A. L. McWhorter, in Physics of Quantum Electronics, edited by P. L. Kelley, B. Lax, and P. E. Tannenwald (McGraw-Hill Book Company, Inc., New York, 1966), pp. 111-118.

[13]P. A. Wolff, to be published.

[14]W. P. Allis, S. J. Buchsbaum, and A. Bers, Waves in Anisotropic Plasmas (Massachusetts Institute of Technology Press, Cambridge, Mass., 1963).

[15]P. M. Platzman, P. A. Wolff, and N. Tzoar, Phys. Rev. 174, 489 (1968).