hollow cylinder the situation does not correspond simply to that of a persistent surface current on the inside surface. There must be additional currents flowing either in the bulk or on the outer surface even in the absence of an external field. An interesting consequence of this model shows that it is possible to have a persistent current on the inside surface of the cylinder and no field in the void. This can be seen from Eq. (3) by letting $A_0 \rightarrow 0$. In this case there would be no field in the wall except for an infinitesimal layer just outside the surface current.

The observation that this technique allows one to generate a single sharp superconducting interface which can be moved easily and reversibly without intermediate-state complications suggests the possible application to NMR experiments and others in which time-dependent effects are important. In this work we have not determined the velocity of interface motion.

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Light Scattering from Plasmas and Single-Particle Excitations in Cadmium Sulfide near Resonance

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Coupled phonon-plasmon excitations and single-particle excitations in CdS have been observed by means of inelastic light-scattering experiments using argon and krypton ion lasers with emission frequencies near that of the CdS band gap. The coupled-mode frequencies, linewidths, and cross sections have been measured as functions of temperature, laser frequency, and carrier concentrations and have been compared with theoretical predictions. The simultaneous observation of coupled one-LO-phonon excitations and uncoupled two-LO-phonon excitations is explained in terms of plasmon dispersion and damping which uncouple the plasmon and LO phonon at wave vectors near q_{FT}, the Fermi-Thomas wave vector.

Raman scattering from electrons in semiconductors has previously been observed in experiments using infrared lasers.[1–3] The reason semiconductors such as CdS and ZnTe, which have band gaps in the visible region of the spectrum, have not been subject to such experimentation is that they have characteristically larger effective masses and lower mobilities than the III-V's. This results in low-frequency, highly damped plasmons, and, therefore, difficulties in detection. In the present study of high-carrier-concentration CdS, we have found that these anticipated drawbacks are compensated experimentally by resonant enhancement and by the excellent properties of the S-20 phototube; we have been able to obtain very good Raman data on coupled LO phonon-plasmon excitations, using low-power argon and krypton ion lasers.

Data were obtained from right-angle scattering experiments on CdS samples (Harshaw) having nominal room-temperature carrier concentrations (due to deviation from stoichiometry) of 2×10^{19} and 4×10^{18} cm^{-3}. Low-power (< 1W) Ar II (5145 Å) and Kr II (5208 and 5682 Å) cw lasers were used, and detection was by means of a Spex 1400 double monochromator, a cooled S-20 (ITT FW-130) photomultiplier, and a Keithley 610B electrometer. A conventional helium Dewar was used for measurements between 6 and 190°K. Figure 1 presents traces of the spectrum of each sample. The data shown are unpolarized. Features labled L_+ and L_- are the high- and low-frequency components of the coupled LO phonon-plasmon excitation, TO is the transverse (A_1 or E_1 symmetry) optical phonon, E_2 is a nonpolar optical mode, 2LO and 3LO are resonant 2 and 3 LO phonon processes consisting of near zone center LO phonons.[4] The data in Fig. 1 were obtained with 5145-Å excitation and are identical to those obtained at 5208 Å.

The theory of light scattering from coupled phonon-plasmon modes has been examined previously in extensive detail; discussion and references to earlier work may be found in Refs. 1–3 and 5. The dielectric function is expressed in terms of phonon oscillators plus an additive Drude term containing a phenomenological lifetime:

$$\epsilon(\omega)=\epsilon_\infty+\sum_j\frac{S_j\omega_j^2}{(\omega_j^2-\omega^2+i\Gamma_j\omega)}-\frac{\omega_p^2\epsilon_\infty}{\omega^2+i\omega/\tau} \quad . \qquad (1)$$

In Eq. (1), ϵ_∞ is the high-frequency dielectric constant; ω_j is the frequency of the jth TO phonon with linewidth Γ_j and oscillator strength S_j; τ is a phenomenological relaxation time which we shall equate with the electron collision time (since Landau damping is insignificant for the low-mobility samples we employed) $\tau_c=\mu m^*/e$, where μ is the mobility, m^* is the effective mass, and e is the electron charge, ω_p is the uncoupled plasmon frequency given by $\omega_p=(1/2\pi c)[4\pi n e^2/\epsilon_\infty m^*]^{1/2}$, where n is the carrier concentration and the factor $1/2\pi c$ yields frequencies in cm^{-1}.

The frequencies of the longitudinal phonon-plasmon modes are given by the zeros of the dielectric function $\epsilon(\omega)$ written in Eq. (1). Since CdS has the uniaxial wurtzite structure, there exist two independent equations of the form given in (1): one for the dielectric response $\epsilon^{\perp}(\omega)$ to phonon polarization fields perpendicular to the hexagonal axis and one for response $\epsilon^{\parallel}(\omega)$ to phonons polarized [0001]. Since CdS is nearly isotropic optically [the poles and zeros of $\epsilon(\omega, k=0)$ are known to vary by less than 8 cm^{-1} as a function of the angle between phonon polarization field and optic axis], we have treated CdS as cubic in this preliminary report and have consequently not discriminated between A_1 and E_1 spectra in Figs. 1 and 2. Figure 2 shows the coupled plasmon-phonon frequency versus ω_p as given by Eq. (2) with $m^*=0.16m_e$ and $\epsilon_\infty=5.2$,[6] and with damping neglected.

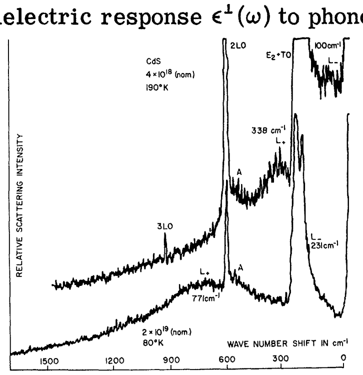

FIG. 1. Unpolarized Raman spectra of CdS samples having nominal room-temperature carrier concentrations of 2×10^{19} and 4×10^{18} cm^{-3}, using 5145-Å excitation. The weak feature at ~ 560 cm^{-1} labeled A is a nonresonant second-order scattering process. [See B. Tell, T. C. Damen, and S. P. S. Porto, Phys. Rev. 144, 771 (1966).] An additional anomaly occurs near the 250-cm^{-1} region in which E_2, E_1 (TO), and L_- are found, and will be discussed in a subsequent paper.

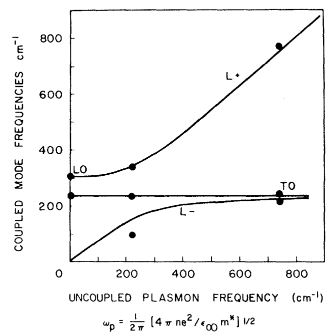

FIG. 2. Plasmon-phonon frequencies calculated from Hall measurement of carrier concentration. Carrier concentrations determined from Hall and Raman data agree perfectly but are 1.9 times the nominal values quoted by the supplier for the 2×10^{19} sample. The mobility inferred from measured plasmon (L_+) linewidth for the 2×10^{19}-cm^{-3} (nom.) concentration sample is 80 cm^2/V sec and agrees fairly well with the measured Hall mobility (which involves several geometrical approximations) of 55 cm^2/V sec. Mobilities for the 4×10^{18}-cm^{-3} sample are 25% higher.

It is to be noted in Fig. 1 that the linewidths for the L_+ excitation are substantially larger than those displayed[1,2] in GaAs. Observe that the linewidth of L_- in the 4×10^{18} sample is much greater than that of L_- in the 2×10^{19} sample. This is due to the fact that L_- in the latter is almost entirely phononlike, while in the former it is more plasmonlike.

The cross section for inelastic scattering of light by electrons in a semiconductor has been predicted to exhibit a divergence given[7] by the expression $I\sim(E_g^2/E_g^2-\hbar^2\omega_L^2)^2$. Under near resonant conditions (laser $\hbar\omega_L\approx$ band gap E_g), this becomes, to a good approximation, $I\sim(E_g-\hbar\omega_L)^{-2}$ since $(E_g+\hbar\omega_L)^{-2}$ is a slowly varying function of ω_L or $E_g(T)$. The resulting factor is algebraically similar to, but of lower power than, Birman and Ganguly's[8] phonon cross-section divergence for $E_g-\hbar\omega_L\gtrsim 3R$, where R is the exciton binding energy. The L_+ excitation in our 2×10^{19}-cm^{-3} sample is essentially pure plasmon. Its cross section has been measured relative to those of the two-LO and one-TO excitations, since the resonant behavior of each of those is known from our earlier work. In the present study, we have varied $E_g(T)$ from 2.56 to 2.58 eV and $\hbar\omega_L$ from 2.20 to 2.44 eV (in discrete steps). The cross section observed over this region $[(E_g - \hbar\omega_L)$ from 0.12 to 0.38 eV$]$ increases by a factor of 10, in agreement with that of 10.0 predicted. A more detailed comparison will be given in a subsequent paper.

Figure 3 illustrates the scattering observed from single-particle electrons[3,5,7] in CdS at different temperatures and laser frequencies. At low temperatures, this scattering is characterized by a linear cross-section dependence upon frequency shift, as indicated on the lower two traces. At higher temperatures, the scattering peak moves away from the phonon features in toward the laser line. At still higher temperatures, a Maxwellian distribution, characteristic of a nondegenerate plasma, would be expected. The single-particle electron scattering was of about the same intensity for polarized α_{xx} and depolarized α_{xy} (or $\parallel$, $\perp$ in Mooradian's notation) scattering. The integrated single-particle scattering is of the order of 10%

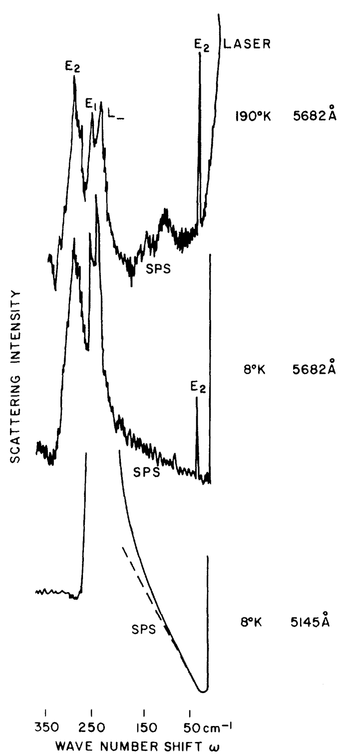

FIG. 3. Single-particle electron scattering in CdS. Note that cross section is proportional to ω at low temperatures (up to a cutoff obscured by phonon scattering). The E_2 mode at 44 cm^{-1} is not seen with 5145-Å excitation. This is probably a resonance phenomenon and is also observed in insulating CdS.

of the collective excitation scattering. Charge-density fluctuation scattering should be negligible compared to spin-density fluctuation scattering under the highly screened plasma conditions in our samples. Hence $\alpha_{xx}(\perp,\perp)$ and $\alpha_{xz}(\perp,\parallel)$ scattering intensities observed for the single-electron feature remain to be explained since spin-density scattering involves only nondiagonal (α_{ij}) terms. This selection-rule breakdown was also observed in heavily doped GaAs by Mooradian.[5]

Note that the two-LO and three-LO processes in Fig. 1 remain sharp and distinct. Since we have previously demonstrated[4] that the two-LO excitation consists of two near-zone-center LO phonons, there is an obvious need to explain why the constituent LO's and the resulting two-phonon line are not broadened enormously by the collision-damped plasmon interaction at $K=0$. Since plasmons become highly damped near the Bohm-Pines plasma cutoff, the requirement for a strong phonon-plasmon interaction is that the wave number of the phonon must be much less than q_C, the Bohm-Pines cutoff. For the temperatures and carrier concentrations considered in this note, q_C is approximately equal to q_{FT}, the reciprocal of the Fermi-Thomas screening length in a degenerate plasma, or, characteristically, $10^7\,\mathrm{cm}^{-1}$. Thus, LO phonons having wave number between 10^6 and $10^7\,\mathrm{cm}^{-1}$ do not couple well to plasmons in CdS under the conditions of our experiments. The plasmon-phonon uncoupling in this region is in part due to the strong plasmon dispersion $\omega_p(q) \approx \omega_p(q=0) \times [1+0.9(q/q_{\mathrm{FT}})^2]$ which separates the uncoupled phonon and plasmon frequencies, but more directly attributable to the increased plasmon damping near $q=q_{\mathrm{FT}}$. In the most general form of Eq. (1), the dielectric function is written $\epsilon(\omega,k)$, and the relaxation time τ in the Drude term is explicitly wave number dependent. $\tau(q)\to 0$ as $q\to q_{\mathrm{FT}}$ and the Drude term $\epsilon_\infty\omega_p^2(\omega^2+i\omega/\tau)^{-1}$ asymptotically approaches $[\epsilon_\infty\omega_p^2\tau^2 - i(\epsilon_\infty\omega_p^2\tau/\omega)]$; the resulting zeros in ϵ are then easily seen to be essentially those given by the uncoupled LO frequency in the absence of carriers. This analysis is an additional confirmation of our identification of phonons comprising two-LO scattering as having $q\sim 10^6\,\mathrm{cm}^{-1}$; while in the past we could show that q(phonon) was not *greater* than $\sim 10^7\,\mathrm{cm}^{-1}$, by reference to the two-LO frequency, we can show that q(phonon) is not *less* than $\sim 10^6\,\mathrm{cm}^{-1}$, by reference to the two-LO linewidth, and the phonon-plasmon decoupling scheme which it implies. We note that the observation of resonant two-LO and three-LO scattering in a sample of 2×20^{19}-cm^{-3} carrier density and 10-Å screening length is not compatible with discrete exciton intermediate states.

In conclusion, the measurement of plasmon frequencies, linewidths, and cross sections in CdS have been shown to be in good agreement with theoretical predictions; single-particle electron scattering has been examined; new information has been obtained concerning the wave number of phonons participating in resonant multiphonon scattering. The general implication of the work is that plasmons in II - VI's can be studied with the most commonly available low-power lasers and that extensive work in this area should be undertaken in view of the interest in II - VI's and the ease of data acquisition.

We would like to thank J. P. Gordon and C. K. N. Patel for their interest in this project and J. J. Hopfield and P. A. Wolff for helpful discussions.

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