high carrier concentrations due to deviations from stoichiometry.

For the $Pb_{1-x}Sn_xTe$ alloys, the change in band structure with composition has been attributed to the influence of relativistic effects on the energies of the conduction and valence bands.[1] It is probable that the changes in $Pb_{1-x}Sn_xSe$ band structure are also due principally to the difference between the relativistic energy shifts of the Pb and Sn atomic valence states. Approximate calculations[14] indicate that the rate of decrease in E_g from this source would be 0.7 eV/100 mole % SnSe, compared with the observed rate of about 0.9 eV/100 mole % SnSe.

In Fig. 1, negative values of E_g have been assigned to those sample compositions and temperatures for which the energy bands are inverted from those in PbSe. Adopting this convention makes it possible to represent the data by straight lines satisfying the equation $E_g=0.13+(4.5\times10^{-4}\ \text{deg}^{-1})\ T-0.89x$ eV. According to this equation, band inversion occurs at 77°, 195°, and 300°K for $x=0.19$, 0.25, and 0.30, respectively. The deviations from the straight line at 77°K for samples with $x=0.21$, 0.23, and 0.25 may be due to the presence of hole concentrations large enough to place the Fermi level inside the valence band and therefore to shift the absorption edge to shorter wavelengths (Burstein effect[15]). The relatively large deviations at 77°K for 2 samples with $x=0.32$ have the wrong sign to be explained in this way.

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Raman Scattering by Coupled Plasmon–Longitudinal-Optical-Phonon Modes in Zincblende-Type Crystals*

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A phenomenological theory of Raman scattering by the $q\approx0$ coupled plasmon–(LO) longitudinal-optical-phonon modes, which takes into account the electro-optic effect of the longitudinal electric fields associated with the coupled modes, is presented. Estimates of the scattering efficiency of the plasmon-like mode at high carrier densities, for which the major contribution comes from the electro-optic effect, are obtained using values of the electro-optic coefficient derived from second-harmonic light-generation data. For *n*-type GaAs with a carrier density of $3\times10^{18}/\text{cm}^3$, the scattering efficiency of the plasmon-like mode is found to be 1×10^{-5}. The theoretical treatment is also applicable to Raman scattering by the collective cyclotron excitation modes of free carriers in a magnetic field and by their coupled modes with LO phonons.

THERE has been considerable interest in recent years in the scattering of electromagnetic (EM) radiation by plasmas in semiconductors, and theoretical expressions have been derived for the contributions to the scattering cross section arising from intraband processes[1,2] and from interband processes.[3] In the case of polar semiconductors which lack a center of symmetry, such as those with ZnS-type structures, it was suggested[4] that the two $q\approx0$ coupled plasmon–(LO) longitudinal-optical-phonon collective modes would participate in first-order Raman scattering via mechanisms analogous to those involved in scattering by the $q\approx0$ LO phonons in the absence of free carriers. The first-order Raman scattering of EM radiation by the $q\approx0$ coupled plasmon–LO-phonon modes in GaAs was recently reported by Mooradian and Wright.[5] Their spectra, which were obtained for samples with different carrier densities, clearly exhibit two Raman lines due to the coupled plasmon–LO-phonon modes as well as the Raman line due to the $q\approx0$ transverse-optical (TO) phonons. In their discussion of the relative intensities of

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the lines due to the coupled modes, Mooradian and Wright noted that the scattering cross section of a given line should be proportional to the phonon or plasmon content of the mode at that frequency. However, there is an important additional contribution to the scattering cross section, which they and previous investigators did not consider, namely, that arising from the electro-optic effect of the longitudinal electric fields associated with the coupled modes.[4] In fact, at high carrier densities, the Raman scattering by the "high-frequency" predominantly plasmon-like mode will be due largely to the electro-optic effect. We present here a phenomenological theory of the Raman scattering by the coupled plasmon–LO-phonon modes in ZnS-type semiconductors, which includes the change in the electronic polarizability induced by the longitudinal electric fields of the coupled modes.[6–8] We also present estimates of the scattering efficiency of the predominatly plasmon-like modes at high carrier densities based on values for the electro-optic coefficients derived from second-harmonic light-generation data. In the case of n-type GaAs with a free-carrier density of $3\times10^{18}/\mathrm{cm}^3$, the Raman-scattering efficiency (per unit length of crystal per unit solid angle) of the plasmon-like coupled mode (plasma strength ≈0.92, phonon strength ≈0.03) is found to be 1×10^{-5}.

The Stokes-Raman scattering efficiency per unit length of crystal per unit solid angle for Raman-active optical phonons is given by[9]

$$S=(\omega_s^4V/2c^4)[\hat{e}_0(\omega_0,\mathbf{k}_0)\chi_j^{(1)}\hat{e}_s(\omega_s,\mathbf{k}_s)]^2(\bar{n}_j+1), \quad (1)$$

where $\hat{e}_0(\omega_0,\mathbf{k}_0)$ and $\hat{e}_s(\omega_s,\mathbf{k}_s)$ are, respectively, the polarization vectors of the incident and scattered radiation whose frequencies and wave vectors are ω_0, $\mathbf{k}_0$ and ω_s, $\mathbf{k}_j$, respectively; $\chi_j^{(1)}$ is the first-order change in the electric susceptibility (electronic polarizability per unit volume) resulting from the creation of a phonon of type j having frequency ω_j and wave vector $\mathbf{q}_j$; V is the scattering volume of the crystal; and $\bar{n}_j$ is the phonon occupation number. The requirements of energy and momentum conservation take the form $\omega_0=\omega_s+\omega_j$ and $\mathbf{k}_0=\mathbf{k}_s+\mathbf{q}_j$, where $|k_0|\approx|k_s|\approx0$ and therefore $|q_j|\approx0$.

In a polar semiconductor, the LO phonons and the plasmons (i.e., the longitudinal collective excitations of the carriers) are coupled by the macroscopic longitudinal electric field which is set up by the relative longitudinal displacement of the positive and negative ions, $\mathbf{u}_L(\omega)$, and by the longitudinal displacements of the free carriers relative to the static charges of opposite sign, $\mathbf{x}_L(\omega)$.[10,11] The longitudinal electric field is given by

$$\mathbf{E}_L(\omega)=-(4\pi Ne^*/\epsilon_\infty)\mathbf{u}_L(\omega)-(4\pi ne/\epsilon_\infty)\mathbf{x}_L(\omega), \quad (2)$$

where N is the number of unit cells per unit volume, e^* is the absolute value of the effective charge of the ions, ϵ_∞ is the high-frequency dielectric constant, n is the density of free carriers, and e is the charge of the carriers.

The $q\approx0$ frequencies of the two coupled plasmon–LO-phonon collective modes are given by[10]

$$2\omega_\pm^2=\omega_L^2+\omega_P^2\pm[(\omega_L^2+\omega_P^2)^2-4\omega_P^2\omega_T^2]^{1/2}, \quad (3)$$

where $\omega_P^2=4\pi ne/m^*\epsilon_\infty$, m^* is the effective mass of the carriers, ω_L is the frequency of the $q\approx0$ LO phonons, and ω_T is the frequency of the $q\approx0$ TO phonons. In the high-frequency ($\omega=\omega_+$) mode, the contributions to the longitudinal field from the displacements of the ions and from the displacements of the carriers have the *same* sign, whereas in the low-frequency ($\omega=\omega_-$) mode the two contributions have *opposite* signs.

The first-order change in electronic polarizability per unit volume induced by the $q\approx0$ coupled modes, $\chi^{(1)}{}_\pm$, is given in component form by

$$\chi^{(1)}{}_{\pm\lambda\nu}=\sum_\sigma\left[\left(\frac{\partial\chi_{\lambda\nu}(\omega_0)}{\partial u_{L\sigma}(\omega_\pm)}\right)_{\mathbf{x},\mathbf{E}^*}du_{L\sigma}(\omega_\pm)+\left(\frac{\partial\chi_{\lambda\nu}(\omega_0)}{\partial x_{L\sigma}(\omega_\pm)}\right)_{\mathbf{u},\mathbf{E}^*}dx_{L\sigma}(\omega_\pm)+\left(\frac{\partial\chi_{\lambda\nu}(\omega_0)}{\partial E_{L\sigma}^*(\omega_\pm)}\right)_{\mathbf{x},\mathbf{u}}dE^*_{L\sigma}(\omega_\pm)\right], \quad (4)$$

where $\chi_{\lambda\nu}(\omega_0)=N\alpha_{\lambda\nu}(\omega_0)$ is the $\lambda\nu$ component of the electric susceptibility at $\omega=\omega_0$; $\alpha_{\lambda\nu}(\omega_0)$ is the $\lambda\nu$ component of the electronic polarizability per unit cell; $du_{L\sigma}(\omega_\pm)$ and $dx_{L\sigma}(\omega_\pm)$ are the σ components of the longitudinal displacements of the ions and of the carriers, respectively, produced by the creation of one coupled-mode quantum; and $dE^*_{L\sigma}(\omega_\pm)$ is the σ component of the associated change in the effective longitudinal electric field. The second term in the expression for $\chi^{(1)}{}_{\pm\lambda\nu}$ represents the change in the electronic polarizability per unit volume induced by the displacements of the carriers at constant $\mathbf{u}_L$ and $\mathbf{E}^*_L$. It will, in general, be very small compared to the other two terms, particularly in ZnS-type semiconductors where the carriers are characterized by nearly-free-electron-type wave functions. We will therefore neglect it for present purposes. The coefficients $(\partial\chi_{\lambda\nu}/\partial u_{L\sigma})_{\mathbf{E}^*}=a_{\lambda\nu\sigma}$ and $(\partial\chi_{\lambda\nu}/\partial E^*_{L\sigma})_{\mathbf{u}}=b_{\lambda\nu\sigma}$ are third-rank tensors which, in the case of ZnS-type crystals, are equal to $|e_{\lambda\nu\sigma}|a$ and $|e_{\lambda\nu\sigma}|b$, respectively, where a and b are the single independent components of the corresponding tensors,

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and $e_{\lambda\nu\sigma}$ is the Levi-Civita function.[9] In the two-subscript notation, $|e_{\lambda\nu\sigma}|a=a_{41}$ and $|e_{\lambda\nu\sigma}|b=b_{41}$.

Since the electronic wave functions in III–V and II–VI ZnS-type semiconductors are nonlocalized, the effective longitudinal electric field can, to a good approximation, be assumed to be equal to the macroscopic longitudinal electric field.[12,13] Thus,

$$d\mathbf{E}^*_L \approx d\mathbf{E}_L(\omega) = \left(\frac{\partial E_L(\omega)}{\partial u_L(\omega)}\right)_{\mathbf{x}_L} d\mathbf{u}_L(\omega) + \left(\frac{\partial E_L(\omega)}{\partial x_L(\omega)}\right)_{\mathbf{u}_L} d\mathbf{x}_L(\omega) = -\frac{4\pi N e^*}{\epsilon_\infty} d\mathbf{u}_L(\omega) - \frac{4\pi n e}{\epsilon_\infty} d\mathbf{x}_L(\omega). \quad (5)$$

Following Varga,[10] the quantities $d\mathbf{u}_L(\omega_\pm)$ and $d\mathbf{x}_L(\omega_\pm)$ can be expressed in terms of the phonon strength $\mathcal{S}_u(\omega_\pm)$ and the plasmon strength $\mathcal{S}_x(\omega_\pm)$, respectively, of the coupled plasmon–LO-phonon modes. These are defined by

$$d\mathbf{u}_L(\omega_\pm)^2 = \mathcal{S}_u(\omega_\pm) d\mathbf{u}_L{}^0(\omega_L{}^2) = \mathcal{S}_u(\omega_\pm)(\hbar/2\bar{M}NV\omega_L), \quad d\mathbf{x}_L(\omega_\pm)^2 = \mathcal{S}_x(\omega_\pm) d\mathbf{x}_L{}^0(\omega_P)^2 = \mathcal{S}_x(\omega_\pm)(\hbar/2m^*nV\omega_P), \quad (6)$$

where $\bar{M}$ is the reduced mass of the two ions in the unit cell; $d\mathbf{u}_L{}^0(\omega_L)$ is the relative displacement of the ions resulting from the excitation of a $q\approx 0$ LO phonon; and $d\mathbf{x}_L{}^0(\omega_L)$ is the displacement of the carriers resulting from the excitation of a $q\approx 0$ plasmon.[7,14]

Upon introducing the expressions for $d\mathbf{u}_L(\omega_\pm)$ and $d\mathbf{x}(\omega_\pm)$ into Eq. (5) for $d\mathbf{E}_L(\omega_\pm)$, we obtain

$$d\mathbf{E}_L(\omega_\pm) = -(2\pi\hbar/\epsilon_\infty V)^{1/2}\left[(\mathcal{S}_u(\omega_\pm)\Omega_L{}^2/\omega_L)^{1/2} \pm (\mathcal{S}_x(\omega_\pm)\omega_P)^{1/2}\right]\hat{d}_\pm(\omega_\pm), \quad (7)$$

where $\hat{d}_\pm(\omega_\pm)$ is the polarization vector of the coupled modes, and $\Omega_L{}^2=\omega_L{}^2-\omega_T{}^2=4\pi Ne^{*2}/\bar{M}\epsilon_\infty$. The $\pm$ sign of the second term within the brackets arises from the fact that $e^*d\mathbf{u}_L(\omega_\pm)$ and $ed\mathbf{x}_L(\omega_\pm)$ have the same sign in the high-frequency ($\omega=\omega_+$) mode and opposite signs in the low-frequency ($\omega=\omega_-$) mode. The corresponding expression for $\chi^{(1)}{}_\pm$ is given in component form by

$$\chi^{(1)}{}_{\pm\lambda\nu} = \sum_\sigma \left\{a|e_{\lambda\nu\sigma}|\left(\frac{\hbar\mathcal{S}_u(\omega_\pm)}{2\bar{M}NV\omega_L}\right)^{1/2} - b|e_{\lambda\nu\sigma}|\left(\frac{2\pi\hbar}{\epsilon_\infty V}\right)^{1/2} \times\left[\left(\mathcal{S}_u(\omega_\pm)\frac{\Omega_L{}^2}{\omega_L}\right) \pm (\mathcal{S}_x(\omega_\pm)\omega_P)^{1/2}\right]\right\}\hat{d}_{\pm\sigma}(\omega_\pm). \quad (8)$$

The expression for the first-order change in the $\lambda\nu$ component of the electric susceptibility induced by $q\approx 0$ LO phonons in the absence of free carriers is the same as that for $\chi^{(1)}{}_{\pm\lambda\nu}$ with $\mathcal{S}_u(\omega_\pm)=1$ and $\mathcal{S}_x(\omega_\pm)=0$.

Since there is no macroscopic electric field associated with TO phonons, the Raman scattering by $q\approx 0$ TO phonons arises only from the first-order change in the electric susceptibility produced by the relative displacement of the positive and negative ions. The expression for $\chi^{(1)}{}_T$, assuming $\mathbf{E}_T{}^*\approx\mathbf{E}_T=0$, is given by

$$\chi^{(1)}{}_{T\lambda\nu} = \sum_\sigma \left(\frac{\partial\chi_{\lambda\nu}(\omega_0)}{\partial u_{T\sigma}(\omega_T)}\right)_{\mathbf{E}^*} \hat{d}u_{T\sigma}(\omega_T) = \sum_\sigma a|e_{\lambda\nu\sigma}|\left(\frac{\hbar}{2\bar{M}NV\omega_T}\right)^{1/2}\hat{d}_{T\sigma}(\omega_T), \quad (9)$$

where we have set $(\partial\chi_{\lambda\nu}(\omega_0)/du_{T\sigma}(\omega_T))=a|e_{\lambda\nu\sigma}|$, since the dependence of $\chi^{(1)}{}_j$ on phonon frequency is negligible over the frequency range ω_T to ω_L.

As shown by Varga,[10] the phonon strength of the coupled modes is given by

$$\mathcal{S}_u(\omega_\pm)^{-1} = \frac{\omega_\pm}{\omega_L}\left[1-\frac{\omega_P{}^2}{\omega_L{}^2}\left(\frac{\omega_T{}^2}{\omega_\pm{}^2}-1\right)^2\right]. \quad (10)$$

The corresponding expression for the plasmon strength of the coupled modes, which is derived by a similar procedure to that used by Varga for the phonon strength, is given by

$$\mathcal{S}_x(\omega_\pm)^{-1} = \frac{\omega_\pm}{\omega_P}\left[1+\frac{\omega_P{}^2\Omega_L{}^2}{(\omega_\pm{}^2-\omega_L{}^2)^2}\right]. \quad (11)$$

Curves of $\mathcal{S}_u(\omega_\pm)$ and $\mathcal{S}_x(\omega_\pm)$ versus carrier density for GaAs, based on $\omega_L=291$ cm^{-1}, $\omega_T=268$ cm^{-1}, $m^*=0.07$ m, $\epsilon_\infty=11.3$, and $\epsilon_0=13.3$, are shown in Fig. 1. The corresponding curves of $d\mathbf{E}_L(\omega_\pm)$ versus carrier

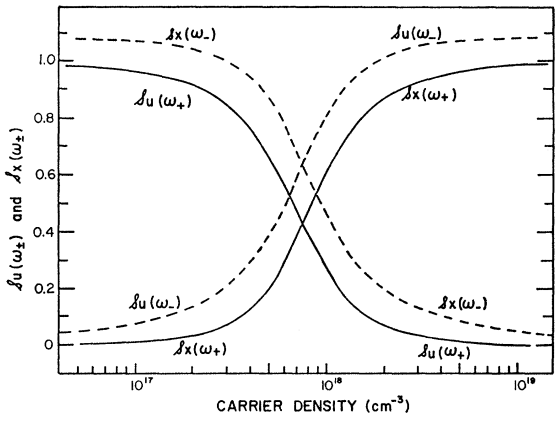

FIG. 1. The phonon strength $\mathcal{S}_u(\omega_\pm)$ and the plasmon strength $\mathcal{S}_x(\omega_\pm)$ of the coupled modes in GaAs. The full curves are for the high-frequency (ω_+) mode and the dashed curves are for the low-frequency (ω_-) mode.

[12] E. Burstein, J. Phys. Chem. Solids Suppl. **26**, 315 (1965).

[13] The effective electric field which determines the displacement force on the ions does contain a dipolar (Lorentz) contribution of $+4\pi/3\mathbf{P}_i$, where $\mathbf{P}_i$ is the contribution to the polarization from the relative displacement of the ions. The dipolar field leads to a modification of the frequencies of the TO and LO phonons. For present purposes we consider these frequencies to be phenomenological parameters.

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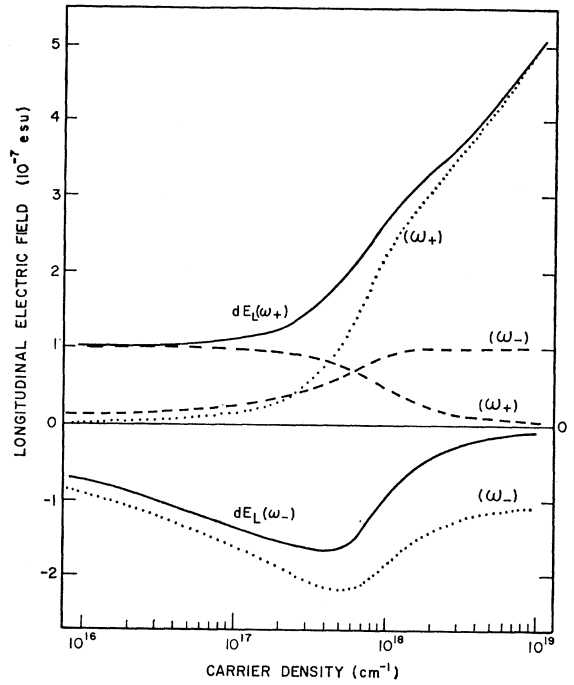

FIG. 2. The longitudinal electric fields in GaAs accompanying the excitation of one coupled-mode quantum: The ionic contributions $d\mathbf{E}_{Lu}(\omega_\pm)$ are indicated by the dashed line, the electronic contributions $d\mathbf{E}_{Lx}(\omega_\pm)$ by the dotted line, and the total field $d\mathbf{E}_L(\omega_\pm)$ by the straight line. The sign of the field is specified relative to the positive-ion displacements. The field set up by the excitation of a $q\approx 0$ LO phonon in a unit volume is 0.97×10^{-7} esu.

density are given in Fig. 2. We note that for $\omega_P \ll \omega_L$, the low-frequency mode is a plasmon-like mode with $\omega_-^2 \approx \omega_p^2(\epsilon_\infty/\epsilon_0)$, in which the displacements of the carriers is partly screened by ion displacements; and that for $\omega_p \gg \omega_L$, the low-frequency mode is a phonon-like mode, with $\omega_-^2 \approx \omega_T^2 = \omega_L^2(\epsilon_\infty/\epsilon_0)$, in which the displacements of the ions are *completely* screened by the electron displacements, i.e., $d\mathbf{E}_L(\omega_-)\approx 0$. On the other hand, for $\omega_P \ll \omega_L$, the high-frequency mode is essentially pure phonon in character with $\omega_+ \approx \omega_L$, and for $\omega_P \gg \omega_L$, the high-frequency mode is essentially pure plasmon in character with $\omega_+ \approx \omega_P$.

Estimates of the magnitudes of the electro-optic contributions to $\chi_L^{(1)}$ and $\chi_\pm^{(1)}$ can be obtained by using values for the electro-optic coefficient $|b_{41}|$ derived from data on the second-harmonic generation (SHG) of light. The SHG of light is determined by the nonlinear susceptibility $d_{\lambda\nu\sigma}$, which can be expressed phenomenologically in the form[15,16]

$$2d_{\lambda\nu\sigma}(2\omega)=\left(\frac{\partial\chi_{\lambda\nu}(\omega)}{\partial E_{T\sigma}(\omega)}\right)_{\mathbf{u}}+\left(\frac{\partial\chi_{\lambda\nu}(\omega)}{\partial u_{T\sigma}(\omega)}\right)_{\mathbf{E}}\frac{du_{T\sigma}(\omega)}{dE_{T\sigma}(\omega)}$$
$$=2|e_{\lambda\nu\sigma}|d(2\omega), \quad (12)$$

where $\mathbf{E}_T$ is the macroscopic transverse electric field and $\mathbf{u}_T$ is the transverse relative displacement of the ions induced by $\mathbf{E}_T$,

$$\mathbf{u}_T(\omega)=\frac{(e^*/\bar{M})\mathbf{E}_T(\omega)}{\omega^2[(\omega_T/\omega)^2-1]}. \quad (13)$$

For $\omega>\omega_T$, the contribution to $|d_{14}|$ from the relative displacement of the ions can be neglected. Thus, to a good approximation,

$$2|e_{\lambda\nu\sigma}|d(2\omega)=2d_{41}(2\omega)\approx(\partial\chi_{\lambda\nu}(\omega)/\partial E_{T\sigma}(\omega))_{\mathbf{u}}. \quad (14)$$

We note further that, under these circumstances, $2|d_{41}|$ and $|b_{41}|$ (the electro-optic coefficient which is involved in Raman scattering) will not differ appreciably from one another except at frequencies close to electronic-resonance transitions. We may therefore set $|b_{41}|\approx 2|d_{41}|$ and use the values of $2|d_{41}|$ obtained from SHG measurements, (at frequencies such that $\omega_T<\omega<\omega_G$) to calculate the magnitude of the electro-optic contributions to the Raman scattering by LO phonons and by the coupled plasmon–LO-phonon modes. For an n-type GaAs sample with a carrier density of $3\times 10^{18}/\text{cm}^3$, the electro-optic contribution to the scattering efficiency of the high-frequency plasmon-like mode ($S_x(\omega_+)\approx 0.92$ and $S_u(\omega_+)\approx 0.03$), on the basis of the experimental value of 0.9×10^{-6} esu for $|d_{14}|$ at 943 cm^{-1},[17] is estimated to be 1×10^{-5}. The corresponding electro-optic contribution to the scattering efficiency of LO phonons in the absence of a free carrier is 1×10^{-6}.[18]

After one takes into account polarization effects, the data on the ratio of the scattering efficiencies of the coupled modes relative to one another and relative to the TO phonons, should in principle enable one to obtain the sign as well as the magnitudes of a_{41} and b_{41}. However, in reporting their Raman data for GaAs, Mooradian and Wright specified neither the polarization nor the directions of the incident and scattered radiation. It is therefore not possible to make even a qualitative interpretation of the relative intensities of the ω_+, ω_-, and ω_T lines in the spectra.

In conclusion, we wish to point out that the theoretical treatment presented here is also applicable to the Raman scattering by the collective cyclotron excitation of free carriers in a magnetic field, and by the coupled modes involving the collective cyclotron excitations and LO phonons.[11] As in the case of the collective modes in the absence of a magnetic field, there will be an important contribution to the scattering efficiency from the electro-optic effect of the macroscopic longitudinal electric fields which are associated with the collective modes. This will be treated in a future paper.

We wish to acknowledge valuable discussions with R. Amado, A. A. Maradudin, Y. Sawada, J. R. Schrieffer, and P. A. Wolf.

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