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INELASTIC LIGHT SCATTERING BY CHARGE CARRIER EXCITATIONS IN TWO-DIMENSIONAL PLASMAS: THEORETICAL CONSIDERATIONS

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We discuss the single particle and collective charge carrier excitations of the two-dimensional plasmas that occur in non-polar semiconductors and the microscopic mechanisms for the resonant inelastic light scattering by the single particle and collective inter-subband excitations. Two limiting cases are analyzed, the "flat-band" model which is a rough approximation to the configuration used by Pinczuk et al., and the "bent-band" model in which Franz–Keldysh effects play an important role, that is an approximation to the configuration used by Abstreiter and Ploog. Unlike Raman scattering by optical phonons, the dominant contributions to the light scattering by the single particle and collective charge carrier excitations come from scattering processes at optical energy gaps, such as the $E_0 + \Delta_0$ gap, that directly involve the charge carriers, particularly in non-polar semiconductors. In III–V compound semiconductors sizeable contributions to the inelastic scattering by coupled LO phonon–collective inter-subband excitation modes may be expected to come from electro-optic, deformation potential and Fröhlich scattering processes at the E_1 and $E_1 + \Delta_1$ gaps.

1. Introduction

Although the inter-subband excitations, of two-dimensional (2D) plasmas have been studied extensively in silicon and in III–V compound semiconductors using infrared absorption [1,2], emission [3] and photoconductivity [4], efforts to investigate them by means of inelastic light scattering have, until very recently,

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been unsuccessful. Inelastic light scattering by the single particle and collective (e.g. plasmon) excitations of three-dimensional (3D) plasmas has been observed in GaAs, InP, and CdTe by Mooradian and co-workers [5–7] and in InAs by Slusher and Patel [8], using photon energies below the E_0 gap. Pinczuk et al. [9] have extended such studies to resonant inelastic scattering by the spin-flip and non-spin-flip single particle electron excitation in GaAs using frequencies in the vicinity of the $E_0 + \Delta_0$ gap. The theoretical cross-section for inelastic light scattering by spin-flip, as well as non-spin-flip, single particle excitations of bulk plasmas in semiconductors has been given by Hamilton and McWhorter [10]. It was recently pointed out by Burstein et al. [11] that the cross-section for light scattering by single particle excitations is strongly enhanced at energy gaps, such as the $E_0 + \Delta_0$ gap, where the direct optical interband transitions involves carrier occupied states in the conduction (or valence) band. In fact, the experimental data at the $E_0 + \Delta_0$ gap for bulk n-GaAs [9,12] showed that the resonant inelastic scattering by single particle excitations can be observed for surface carrier densities, as low as $5 \times 10^{11}/\mathrm{cm}^2$, which are typical of the carrier densities in surface inversion and accumulation layers. They therefore suggested that it should be possible to observe the resonance enhanced inelastic scattering by carriers in 2D plasmas and to obtain, thereby, information about the energies of the single particle and collective inter-subband excitations and, in the case of polar semiconductors, about the coupling of the inter-subband excitations with LO phonons.

During the past year Abstreiter and Ploog [13], using a $GaAs/Ga_xAl_{1-x}As$ heterojunction, have observed resonant inelastic light scattering, at the $E_0 + \Delta_0$ gap, by spin-flip single particle inter-subband excitations of electrons in GaAs accumulation layer. In addition, Pinczuk et al. [14] have observed resonant scattering at the $E_0 + \Delta_0$ gap by spin-flip single particle and by collective inter-subband excitations of the electron plasma in the GaAs layers of a multilayer GaAs/AlGaAs heterojunction supperlattice. The data obtained in these investigations reinforce our expectation that resonant inelastic light scattering can be used as a meaningful probe of the electronic excitations of 2D plasmas in semiconductors.

We present in this paper a discussion of the theoretical considerations which underly the observation of inelastic light scattering by the inter-subband excitations of 2D plasmas in semiconductors. We will, among other matters, discuss: (i) the single particle and collective excitations of a 2D plasma, (ii) the coupling of the inter-subband excitations with LO phonons in polar materials, (iii) the key mechanisms for the inelastic light scattering by the single particle and collective inter-subband excitations of 2D plasmas in non-polar semiconductors, and (iv) the scattering matrix elements for two limiting cases, a "bent-band" model which corresponds to the confined electrons in GaAs at a GaAs/GaAlAs heterojunction, and a "flat-band" model which is a rough approximation to the confined electrons in the GaAs layers of the multilayer GaAs/GaAlAs heterojunction superlattice.

2. The single particle and collective excitations of 2D plasmas

As is well known, the motion of the electrons in an n-channel accumulation (or inversion) layer at the surface of a semiconductor is quantized in the direction perpendicular to the plane of the layer, but remains free in the plane. As a consequence, the energy levels of the electrons form a set of two-dimensional subbands. The single particle (sp) and collective excitations of the 2D electron plasma in the semiconductor (which we assume for present purposes has spherical energy bands and is at low temperature) fall into two categories: *intra*-subband excitations and *inter*-subband excitations (fig. 1).

The sp intra-subband excitations are "uncorrelated" electron–hole pair excitations in which electrons with wavevector $\boldsymbol{\kappa}_{/\!/}$ in a given subband σ are excited from states $\mathcal{E}_\sigma(\boldsymbol{\kappa}_{/\!/})$ below the Fermi surface to empty states $\mathcal{E}_\sigma(\boldsymbol{\kappa}_{/\!/}+\boldsymbol{q}_{/\!/})$ above the Fermi surface with wavevector $(\boldsymbol{\kappa}_{/\!/}+\boldsymbol{q}_{/\!/})$ within the same subband. For both spin-flip and non-spin-flip excitations, the excitations energies are given by:

$$\hbar\omega_{\sigma\sigma}(\boldsymbol{q}_{/\!/}) = \mathcal{E}_\sigma(\boldsymbol{\kappa}'_{/\!/}) - \mathcal{E}_\sigma(\boldsymbol{\kappa}_{/\!/}) = \frac{\hbar^2}{2m^*}\,[(\boldsymbol{\kappa}_{/\!/}+\boldsymbol{q}_{/\!/})^2 - \boldsymbol{\kappa}_{/\!/}^2]\ , \tag{1}$$

where m^* is the effective mass of the electrons.

The collective intra-subband excitations are correlated 2D plasma oscillations of the electrons parallel to the surface e.g. two-dimensional plasmons, whose frequency (neglecting retardation) is given by [15,16]

$$\omega_{\mathrm{P}}^2(q_{/\!/}) = \frac{4\pi n_{\mathrm{s}} e^2}{m^*(\epsilon_1+\epsilon_2)}\ \text{(cgs)}\ , \tag{2}$$

where n_{s} is the surface charge density: ϵ_1 and ϵ_2 are the dielectric constants of the semiconductor and adjacent medium, respectively.

The sp inter-subband excitations are uncorrelated e–h pair excitations in which

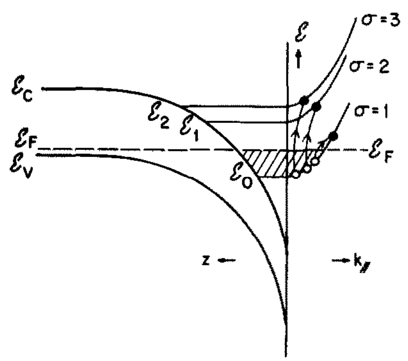

Fig. 1. Schematic diagram of the subbands of the 2D electron plasma in an inversion layer showing intra-subband ($\Delta\sigma = 0$) and inter-subband ($\Delta\sigma = 1$ and $\Delta\sigma = 2$) excitations.

electrons make transitions from states $\mathcal{E}_\sigma(\mathbf{\kappa}_\parallel)$ in a given subband σ below the Fermi surface into empty states $\mathcal{E}'_\sigma(\mathbf{\kappa}'_\parallel)$ in a different subband σ'. For the case in which the Fermi level $\mathcal{E}_F$ lies below the $\sigma = 2$ subband, i.e. the electrons only occupy the $\sigma = 1$ subband, the energies of the spin-flip and non-spin-flip inter-subband excitations are given by,

$$\hbar\omega_{\sigma\sigma'}(\mathbf{q}_\parallel) = \Delta_{\sigma\sigma'} + \frac{\hbar^2}{2m^*}\,[(\mathbf{\kappa}_\parallel + \mathbf{q}_\parallel)^2 - \mathbf{\kappa}_\parallel^2] \tag{3}$$

where $\Delta_{\sigma\sigma'}$ is the sp inter-subband excitation energy for $\mathbf{q}_\parallel = 0$ which differs from $\mathcal{E}_{\sigma'}(\mathbf{\kappa}_\parallel + \mathbf{q}_\parallel) - \mathcal{E}_\sigma(\mathbf{\kappa}_\parallel)$ due to exciton and other effects.

The collective non-spin-flip inter-subband excitations have a macroscopic polarization electric field perpendicular to the 2D layer and, therefore, have energies which are greater than those of the corresponding sp inter-subband excitations [16, 18,19]. The frequencies of the collective modes in the limit $\mathbf{q}_\parallel \to 0$ correspond to the zeroes of $\epsilon_\perp(\omega)$, the dielectric constant of the 2D plasma layer for electric fields normal to the layer. With electrons only occupying the lowest ($\sigma = 1$) subband, $\epsilon_\perp(\omega)$ is given by [16],

$$\epsilon_\perp(\omega) = \epsilon_m(\omega) + \epsilon_{\perp ex} = \epsilon_m(\omega) + \sum_n \frac{4\pi n_s e^2 f_{1n}}{z_{1n} m^*(\omega_{\perp 1n}^2 - \omega^2 - i\omega\gamma_{1n})}\,, \tag{4}$$

where $\epsilon_m(\omega)$ is the dielectric constant of the semiconductor medium: $\epsilon_{\perp ex}$ is the contribution to $\epsilon_\perp(\omega)$ from the non-spin-flip inter-subband excitations; f_{1n} is the oscillator strength of the $\sigma = 1$ to $\sigma = n$ inter-subband transition; z_{1n} is the corresponding effective width of the 2D plasma; and $\hbar\omega_{\perp 1n}$ and γ_{1n} are the energy and damping constant of the $\sigma = 1$ to $\sigma = n$ inter-subband excitation. Thus, on this basis, one finds that the frequency of the $\sigma = 1$ to $\sigma = 2$ collective inter-subband excitation, in the limit $\mathbf{q}_\parallel \to 0$ is given by

$$\omega_{\perp 12}^{*2} = \omega_{\perp 12}^2 + \frac{4\pi n_s e^2 f_{12}}{z_{12} m^* \epsilon_\perp^0(\omega_{\perp 12}^*)} = \omega_{\perp 12}^2 + \frac{\Omega_{\perp 12}^2}{\epsilon_\perp^0(\omega_{\perp 12}^*)}\,, \tag{5}$$

where $\epsilon_\perp^0(\omega_{\perp 12}^*)$ is the contribution to $\epsilon_\perp(\omega)$ at $\omega = \omega_{\perp 12}^*$ from all other electronic transitions (including the electron transitions to higher subbands) and, in polar materials, from optical phonons; and $\Omega_{\perp 12}$ is the unscreened "plasmon" frequency of the $\sigma = 1$ to $\sigma = 2$ inter-subband excitation whose magnitude is determined by the macroscopic polarization electric field of the collective inter-subband excitations of polar medium.

The ω versus $q_{\parallel x}$ dispersion curves of the $1 \to 2$ and $1 \to 3$ plasmon in a non-polar medium (e.g. Si) at low temperatures are shown in fig. 2. We note that the dispersion curves for the sp inter-subband excitations are very similar in character to the dispersion curves for the corresponding sp inter-Landau-subband (e.g. cyclotron resonance) excitations of a bulk plasma in an externally applied magnetic field [20].

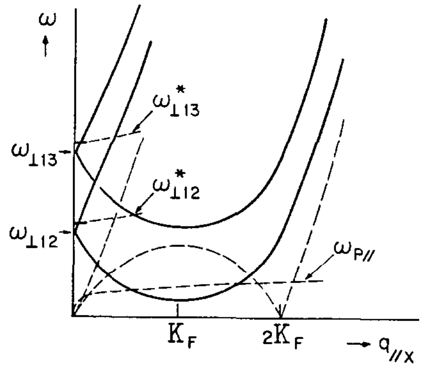

Fig. 2. Schematic diagram showing the ω versus $q_{/\!/x}$ dispersion curves for the single particle and collective *inter*-subband ($\Delta\sigma = 1$ and 2) excitations of a 2D plasma, and the corresponding dispersion curve (dashed line) for the single particle and collective *intra*-subband excitations. We note that the collective inter-subband excitations are Landau damped in the region of the sp excitations.

3. Coupling of charge carrier excitations with LO phonons

In polar materials, the collective excitations of a bulk electron plasma form coupled modes with LO phonons which are admixtures of electron and lattice oscillations [22]. A similar coupling also occurs between LO phonons and the collective inter-subband excitations of the 2D plasma.

Consider, for example, electrons that are confined in a thin inversion layer at a polar semiconductor–vacuum interface. Following Chen et al. [18] we may regard this as a three layer (e.g. vacuum–inversion layer–bulk medium) configuration. The bulk semiconductor has a dielectric constant which, neglecting spatial dispersion and damping, is given by

$$\epsilon_m(\omega) = \epsilon_\infty + \Omega_p^2/(\omega_{TO}^2 - \omega^2)\,,$$

where $\Omega_p^2/\epsilon_\infty = \omega_{LO}^2 - \omega_{TO}^2$. The surface layer is regarded as an anisotropic dielectric slab of thickness L, whose response to an electric field perpendicular to the interface is described by,

$$\epsilon_\perp^s(\omega) = \epsilon_m(\omega) + \epsilon_{\perp 12}(\omega) = \epsilon_m(\omega) + \Omega_{\perp 12}^2/(\omega_{\perp 12}^2 - \omega^2)\,, \tag{6}$$

where $\Omega_{12}^2/\epsilon_m(\omega) = \omega_{\perp 12}^{*2} - \omega_{\perp 12}^2$, and whose response to a field parallel to the surface is described by,

$$\epsilon_{/\!/}^s(\omega) = \epsilon_m(\omega) - \Omega_{/\!/ex}^2/\omega^2\,. \tag{7}$$

(We are neglecting the contributions to $\epsilon_\perp^s(\omega)$ from inter-subband transitions to subbands with $\sigma' > 2$.)

The dispersion relation of the normal modes that are localized at the thin inversion layer, whose wavevector component parallel to the interface is such that $q_{/\!/}L \ll 1$, is (neglecting retardation) readily shown to be given by,

$$\epsilon_{\perp}^{s}(\omega)\,[\epsilon_{m}(\omega)+1] + q_{/\!/}L\{\,[\epsilon_{/\!/}^{s}(\omega)\,\epsilon_{\perp}^{s}(\omega)]^{1/2}\}\{\,[\epsilon_{/\!/}^{s}(\omega)\,\epsilon_{\perp}^{s}(\omega)]^{1/2} - \epsilon_{m}(\omega)\} = 0\,. \quad (8)$$

There are four distinct modes. The first, which corresponds to $\epsilon_m(\omega) + 1 = 0$, is the surface optical phonon mode of the substrate. As $q_{/\!/} \to 0$, its field penetrates deeply into the substrate and its frequency is unaffected by the thin inversion layer. There are in addition two modes, $S_+(\omega)$ and $S_-(\omega)$, namely coupled LO phonon-collective inter-subband excitations, that are the analogs of the L^+ and L^- coupled plasmon–LO phonon modes in a bulk semiconductor. In the limit $q_{/\!/} \to 0$ they have fields that are entirely confined to the plasma layer and directed normal to the interfaces. Finally, one also has the two-dimensional plasma oscillations whose field is parallel to the interface and whose frequency vanishes in the limit $q_{/\!/} \to 0$.

Our interest is in the S_+ and S_- coupled modes. Considering only the $\sigma = 1$ to $\sigma = 2$ inter-subband excitation, the condition $\epsilon_{\perp}(\omega) = 0$ yields the frequencies

$$\omega_{s\pm}^2 = \tfrac{1}{2}(\omega_{\perp 12}^{*2} + \omega_{LO}^2) \pm [(\omega_{\perp 12}^{*2} - \omega_{LO}^2)^2 + 4\Delta^4]^{1/2}\,, \quad (9)$$

where

$$\Delta^4 = \Omega_{\perp 12}^2\,\Omega_p^2/\epsilon_\infty^2 = (\omega_{LO}^2 - \omega_{TO}^2)(\omega_{\perp 12}^{*2} - \omega_{\perp 12}^2)\,.$$

When $\omega_{\perp 12}^* \approx \omega_{LO}$, one has $\omega_{s\pm}^2 = \omega_{LO}^2 \pm \Delta^2$. In the limit $\omega_{\perp 12}^* \ll \omega_{LO}$, one has $\omega_{s+}^2 = \omega_{LO}^2$ and $\omega_{s-}^2 = \omega_{\perp 12}^2 + \Omega_{\perp 12}^2/\epsilon_m(0)$, i.e. it is the static dielectric constant $\epsilon_m(0)$ of the semiconductor medium that provides the screening of the collective inter-subband excitation. In the opposite limit $\omega_{LO} \ll \omega_{\perp 12}^*$, one has $\omega_{s+}^2 = \omega_{\perp 12}^{*2}$

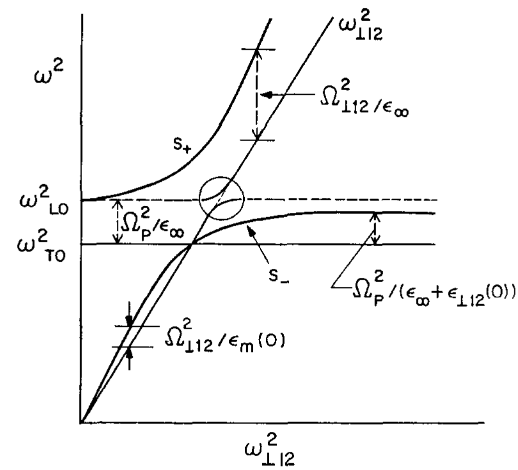

Fig. 3. Schematic diagram showing the S_+ and S_- coupled modes of a 2D plasma in a polar semiconductor. The effect of the polaron type coupling of the sp inter-subband excitations with LO phonons is shown in the circled frequency region.

and

$$\omega_{s-}^2 = \omega_{TO}^2 + \Omega_p^2/(\epsilon_\infty + \epsilon_{\perp 12}(0))$$

which lies intermediate between ω_{TO}^2 and ω_{LO}^2.

In addition to the coupling between the collective excitation and the LO phonons, there will also be a polaron-type coupling of the non-spin-flip sp inter-subband excitations with LO phonons. This polaron-type coupling, which is similar in nature to the polaron-type coupling of sp cyclotron resonance excitations with LO phonons [21], will be particularly pronounced when $\omega_{\perp 12} \approx \omega_{LO}$.

A plot of ω_{s+}^2 and ω_{s-}^2 versus $\omega_{\perp 12}^2$, the square of the sp inter-subband excitation frequency, is given in fig. 3. In practice, one can sweep $\omega_{\perp 12}$ through ω_{LO} by varying the bias voltage across the inversion layer. The effect of the polaron-type coupling of the non-spin-flip sp inter-subband excitation with LO phonons is also shown in the figure.

4. Inelastic light scattering by a 2D plasma

We now discuss the inelastic light scattering by the charge carrier excitations of an n-channel inversion (or accumulation) layer, in which an incident photon (ω_i, $\boldsymbol{k}_i$) is annihilated, a scattered photon ($\omega_s, \boldsymbol{k}_s$) is created and a charge carrier excitation (ω_{ex}, $\boldsymbol{q}_{ex}$) is either created (Stokes scattering) or annihilated (anti-Stokes scattering). Energy is conserved in the light scattering process, so that $\omega_s = \omega_i \mp \omega_{ex}$ where the $-$ and $+$ signs correspond to Stokes and anti-Stokes scattering, respectively. However, because of the quantization of the carriers perpendicular to the surface, and also because L is very much smaller than the wavelengths of the incident and scattered radiation in the inversion layer, only $\boldsymbol{k}_{s\parallel} = \boldsymbol{k}_{i\parallel} \mp \boldsymbol{q}_\parallel$, the component of the scattering wavevector parallel to the surface is conserved, i.e. $\boldsymbol{q}_{ex\parallel} = \boldsymbol{q}_\parallel$.

The inelastic light scattering by the sp and collective intra-subband electronic excitations in a 2D semiconductor plasma is very similar to the inelastic light scattering by the sp and collective excitation of a bulk plasma. An excellent review of the latter has been given by Mooradian [23]. We will therefore only focus our attention on the light scattering by inter-subband excitations. We will, in fact, consider a backward scattering geometry in which $\boldsymbol{q}_\parallel \approx 0$, namely a geometry in which one does not observe inelastic light scattering by the intra-subband excitations. We consider, in particular, resonant inelastic scattering at the $E_0 + \Delta_0$ direct gap of a diamond or zincblende type semiconductor, where both the conduction band and the valence band are, apart from spin, non-degenerate. The scattering processes, i.e. the sequences of electronic transitions, which contribute to the Stokes scattering by the inter-subband excitations of an n-channel inversion layer in which only the lowest (e.g., $\sigma = 1$) conduction subband has carriers are shown in figs. 4, 5 and 6. The spread in energy and wavevector of the valence band states that is indicated in

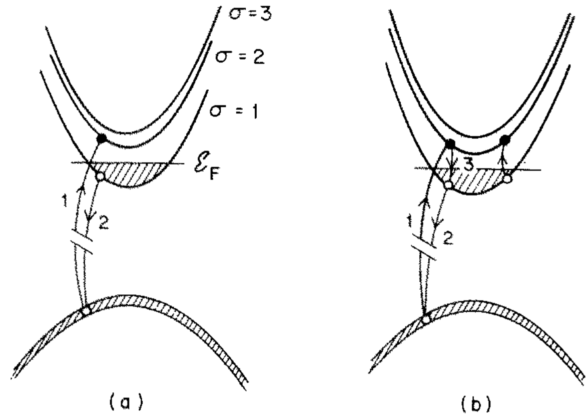

Fig. 4. Schematic diagram showing the transitions involved in (a) the two-step scattering mechanism and (b) in the three-step scattering mechanism.

the figures corresponds to the range of energies and wavevectors of the states of the "bent" valence band that take part in Franz–Keldysh (tunneling-assisted) optical interband transitions.

The key mechanisms for resonant light scattering by the inter-subband excitations of 2D plasmas in the non-polar semiconductors Si and Ge are the two-step and three-step "carrier density" scattering mechanisms, so called because they require a non-zero carrier density in either the conduction band or the valence band [9],

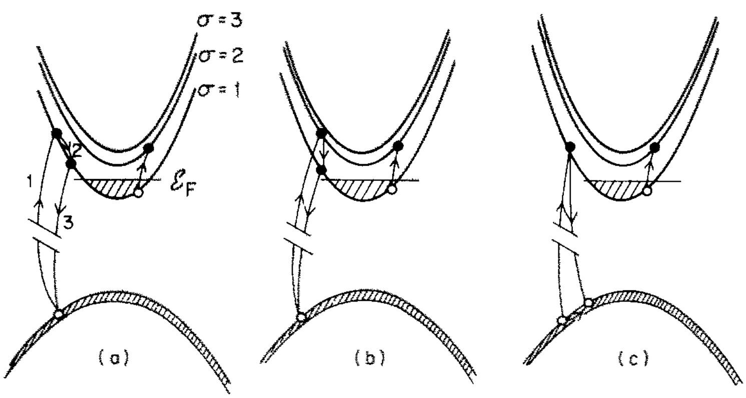

Fig. 5. Schematic diagrams showing the transitions involved in the Fröhlich mechanism for light scattering by collective inter-subband excitations. In (a) the excited electron is scattered into the same subband. In (b) the excited electron is scattered into another subband. In (c) the excited hole is scattered into another valence band state.

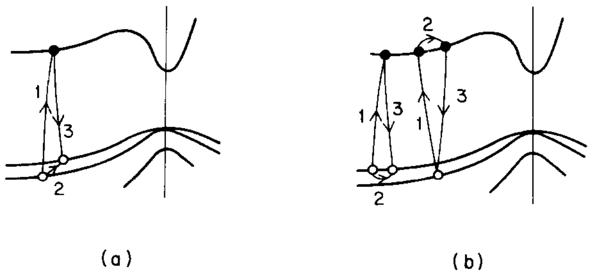

Fig. 6. Schematic diagrams showing (a) the transitions involved in the electro-optic scattering mechanism and in the 3-band deformation potential scattering mechanism, and (b) the transitions involved in the 2-band deformation potential and Fröhlich scattering mechanisms.

and the Fröhlich scattering mechanism which exhibits a strong resonance enhancement at all optical gaps, particularly when there is an appreciable bending of the energy bands. (Optical gaps at which the two-step and three-step "carrier density" mechanisms are operative are listed in table 1.)

In the case of polar semiconductors which lack a center of inversion, such as the III–V compounds, one must also include the deformation potential and electro-optic scattering mechanisms which exhibit resonant enhancement at all optical gaps and particularly at the E_1 and $E_1 + \Delta_1$ gaps.

We note that the two-step and three-step carrier density scattering mechanisms and the Fröhlich scattering mechanism involve two energy bands; the deformation potential scattering mechanism involves either two or three energy bands; and the electro-optic scattering mechanism involves three energy bands.

4.1. Scattering by single particle excitations

The two-step scattering process which is the only one that can lead to resonant light scattering by sp spin-flip excitations is an "electronic Raman scattering" process [24]. In the first step an electron is optically excited from the valence band

Table 1
Optical energy gaps at which resonant inelastic light scattering can occur via the two-step and three-step carrier density mechanisms

Semiconductor	Optical gap	E_g (eV)
n-Si	E_2	4.35
p-Si	E_0'	3.35
n-Ge	E_1	2.20
n-Ge	$E_1 + \Delta_1$	2.40
n-GaAs, p-GaAs	E_0	1.51
n-GaAs	$E_0 + \Delta_0$	1.85

to a virtual vertical state associated with the $\sigma = 2$ conduction subband. In the second step, an electron in the $\sigma = 1$ conduction subband, with either the same or the opposite spin as that of the "excited" electron, annihilates the hole in the valence band. The interband electronic transitions that are involved in the two steps, are shown in fig. 4a. The net effect of the two interband transitions is to generate either a spin-flip or a non-spin-flip sp inter-subband ($\Delta\sigma = 1$) excitation. In the III–V compounds, where there is a strong spin–orbit coupling, the cross-sections for light scattering by the spin-flip and non-spin-flip excitations via the two-step mechanism are comparable in magnitude [10].

In the three-step carrier-density scattering process which only contributes to scattering the non-spin-flip charge carrier excitations, the first and second steps involve the same interband transitions between the valence band and the $\sigma = 2$ and $\sigma = 1$ conduction subband states that take part in the two-step scattering process (fig. 4b). The third step involves the transfer of excitation, via coulomb interaction, from the non-spin-flip interband excitation generated by the two interband transitions, to another non-spin-flip sp excitation. (The third step can be viewed as representing the screening of the sp non-spin-flip inter-subband excitation by the other sp non-spin-flip inter-subband excitations.)

The polarization selection rules for the inelastic light scattering by sp excitation in 2D plasmas via the two-step and three-step mechanisms are the same as those in bulk plasmas. Namely, the backward scattering of light by spin-flip sp inter-subband excitation appears in $\hat{e}_i \perp \hat{e}_s$ spectra, whereas the scattering by non-spin-flip excitations appears in $\hat{e}_i /\!/ \hat{e}_s$ spectra, where $\hat{e}_i$ and $\hat{e}_s$ are the polarization vectors of the incident and scattered light.

On applying the formulation of Hamilton and McWhorter [10] to the scattering by the sp inter-subband excitations of the 2D plasma, one obtains the following expressions for the contributions to the scattering cross-sections of the sp spin-flip and non-spin-flip $\sigma = 1$ to $\sigma = 2$ inter-subband excitations from the two-step and three-step carrier density scattering mechanisms in the limit $q_{/\!/} \to 0$:

$$\frac{\mathrm{d}^2 R_{\mathrm{sf}}}{\mathrm{d}\Omega\,\mathrm{d}\omega} \propto (\hat{e}_i \cdot \hat{e}_s)^2 \sum_{\kappa_{/\!/}} |M(\kappa_{/\!/})|^2_{\mathrm{sf}}\, \mathrm{Im}[\epsilon_{\perp 12}(\omega)]\,, \tag{10}$$

$$\frac{\mathrm{d}^2 R_{\mathrm{nsf}}}{\mathrm{d}\Omega\,\mathrm{d}\omega} \propto (\hat{e}_i \cdot \hat{e}_s)^2 \sum_{\kappa_{/\!/}} |M(\kappa_{/\!/})|^2_{\mathrm{nsf}}\, |\epsilon_{\mathrm{m}}|^2\, \mathrm{Im}[1/\epsilon_{\perp}(\omega)]\,, \tag{11}$$

where the sum is taken over electrons with wavevector $\kappa_{/\!/}$, and over the valence band states that participate in the Franz–Keldysh optical interband transitions, and we have made use of the relation,

$$\left[1 - \frac{\epsilon_{\perp 12}(\omega)}{\epsilon_{\perp}(\omega)}\right]^2 \mathrm{Im}[\epsilon_{\perp 12}(\omega)] = \frac{|\epsilon_{\mathrm{m}}(\omega)|^2\, \mathrm{Im}[\epsilon_{\perp 12}(\omega)]}{|\epsilon_{\perp}(\omega)|^2} = |\epsilon_{\mathrm{m}}(\omega)|^2\, \mathrm{Im}\left[\frac{1}{\epsilon_{\perp}(\omega)}\right]. \tag{12}$$

$|M(\kappa_{/\!/})|$ is the scattering matrix element which contains the interband momentum matrix elements $|p_{\mathrm{cv}}|_i$ and $|p_{\mathrm{cv}}|_s$, and the energy denominator for the optical interband transition that is involved in the first step of the two-step and three-step carrier density mechanisms. (A discussion of $|M(\mathbf{\kappa}_{/\!/})|$ is given in section 5.)

The scattering cross-section for the sp spin-flip inter-subband excitations is proportional to $\mathrm{Im}[\epsilon_{\perp 12}(\omega)]$. The $\hat{e}_i \perp \hat{e}_s$ spectrum will therefore exhibit a peak at the frequency $\omega_{\perp 12}$ of the sp inter-subband excitations, whose width is $2\gamma_{12}$ and whose strength is proportional to the total number of electrons in the scattering volume. For a given beam width of the incident radiation, the scattering intensity will be proportional to n_s, the number of carriers per unit area in the 2D plasma layer.

The two terms on the left side of eq. (12) correspond to the contributions from the two-step and three-step mechanisms, respectively, to the scattering by sp non-spin-flip excitations. Since $\epsilon_{\perp}(\omega) = \epsilon_m(\omega) + \epsilon_{\perp 12}(\omega) \approx \epsilon_{\perp 12}(\omega)$ at $\omega = \omega_{\perp 12}$ and, therefore, $\epsilon_{\perp 12}(\omega)/\epsilon_{\perp}(\omega) \approx 1$, the contribution from the two-step mechanism is largely canceled ("screened") by the contribution from the three-step mechanism. As in the case of sp non-spin-flip excitations in 3D plasmas, the net contribution to the scattering cross-section is proportional to $\mathrm{Im}[1/\epsilon_{\perp}(\omega)]$. The resonant light scattering by sp non-spin-flip inter-subband excitations via the two-step and three-step charge density mechanism is equivalent to light scattering by charge density fluctuations [28]. The "screening" of the sp non-spin-flip inter-subband excitations by other sp non-spin-flip excitations will also greatly decrease the contribution to the scattering cross-section from the Fröhlich scattering mechanism. We will therefore only consider the role of the Fröhlich scattering mechanism in light scattering by the collective inter-subband excitations and their coupled modes with LO phonons.

4.2. Scattering by collective excitations

In the three-step carrier density mechanism for Stokes scattering by collective inter-subband excitations of the 2D plasma, the third step involves the transfer of excitation from the sp non-spin-flip inter-subband excitation to the collective inter-subband excitation (i.e., an inter-subband electron–hole pair is annihilated and a collective excitation is created) via coulomb interaction with the macroscopic field $E_{\perp ex}$ of the collective excitation.

In the Fröhlich scattering mechanism for resonant light scattering by collective excitations, which is applicable at all optical gaps (fig. 5), the first step involves the creation of an electron–hole pair by an optical interband transition. The second step involves the scattering of the electron (hole) in the intermediate state via the Fröhlich interaction with the macroscopic field. In the light scattering at the $E_0 + \Delta_0$ gap, the electron may be scattered into a state within the same subband or into another subband. The electron contribution (figs. 5a and 5b) and the hole contribution (fig. 5c) have opposite signs. However, unlike the case of scattering in a bulk semiconductor with flat energy bands where the electron and hole contributions cancel in the limit $q \to 0$, the electron and hole contributions to the scattering by the collective excitations of the 2D plasma in an inversion layer do not cancel. The non-cancellation is due, on the one hand, to be sizeable perpendicular component of the wavevector of the macroscopic field of the collective inter-subband excitation, e.g. $q_{\perp ex} \approx 2\pi/L \approx 10^6$ cm^{-1}, and on the other hand, to the spatial separation of the excited electron and hole in the intermediate state that results from Franz–Keldysh optical interband transitions when the energy bands inolved are bent [25–28].

As in the case of the inelastic light scattering by the collective excitations (e.g. plasmons) of bulk plasmas, the contributions to the scattering cross-sections of the collective inter-subband excitations from the three-step charge density scattering mechanism and the Fröhlich scattering mechanism are both proportional to $E_{\perp ex}^2$.

In the III–V compounds, the electro-optic and deformation potential scattering mechanisms also play roles in resonant light scattering by the collective inter-subband excitations and their coupled modes with LO phonons. They are particularly important at the E_1 and $E_1 + \Delta_1$ gap where in the case of bulk plasmas they yield a strong resonance enhancement of the light scattering by LO phonons and their coupled modes with plasmons [29,30].

The electro-optic (three-band) and deformation potential (two- and three-band) scattering processes at the E_1 (and $E_1 + \Delta_1$) gap are shown in figs. 6a and 6b. Since the conduction band does not have a minimum at this gap, the states in the conduction band do not form well-defined quantized subbands.

In the electro-optic (three-band) scattering mechanism (fig. 6a) the first step involves the creation of an electron–hole pair. The second step involves the *interband* scattering of the electron (also hole) into another band by the macroscopic electric field of the coupled LO phonon–collective inter-subband excitation mode. The third step involves the recombination of the electron and hole. In this mechanism which requires the absence of a center of inversion, the electron and hole contributions to the scattering matrix elements do not cancel. It should be noted that, apart from the fact that it involves three bands, the electro-optic mechanism differs from the three-step carrier density and the Fröhlich mechanism in that the "scattering" of the electron (hole) in the electro-optic mechanism involves an interband matrix element in which the macroscopic field acts on the orbital (periodic) part of the electron wavefunction; whereas the scattering of the electron (hole) in the three-step and Fröhlich mechanism involves an intraband (Fröhlich) matrix element in which the macroscopic field acts on the envelope part of the electron wavefunction.

In the deformation potential (two-band and three-band) scattering mechanism (fig. 6b) the first step involves the creation of an electron–hole pair. The second step involves the scattering of the electron (hole) into another state, in either the same energy band or another energy band, by the coupled LO phonon–collective inter-subband excitation mode, via the deformation potential interaction with the optical phonon content of the coupled mode which acts on the orbital part of the electron (hole) wavefunction. The third step involves the recombination of the electron–hole pair. Since the deformation potentials of the valence and conduction bands (i.e. the modulation of the electronic energy states by the periodic deformation of the lattice) are in general different, the electron and hole contributions to the scattering cross-section do not cancel.

Scattering via the wavevector and electric field dependent Fröhlich mechanism is also quite strong at the E_1 and $E_1 + \Delta_1$ gaps, particularly when there are sizeable surface space charge fields. In this connection it should be emphasized that the electro-optic and deformation potential mechanisms for scattering by coupled LO phonon–collective inter-subband excitation modes are "allowed" scattering mechanisms, and their polarization selection rules are different from those for the Fröhlich scattering mechanism which is a "forbidden" i.e. wavevector dependent scattering mechanism. Moreover, in contrast to the Fröhlich mechanism, the electro-optic and deformation potential mechanism are relatively insensitive to band bending effects.

5. The scattering matrix elements

We turn next to a discussion of the interband optical transitions and the associated momentum matrix elements that are involved in the resonant inelastic light scattering at the $E_0 + \Delta_0$ optical gap by the sp and collective inter-subband excitation of a 2D electron plasma. For this purpose we consider two limiting models for the confinement of the 2D electron plasma. In the first, which we call the "flat-band" limit the electrons are confined within a slab of thickness L by potential walls of essentially infinite height. This is a rough approximation to the confined electronic plasma in the GaAs layers of the multilayer GaAs/GaAlAs heterojunction superlattice which was used by Pinczuk et al. [14]. In the second limit which we will call the "bent-band" limit, the electrons are trapped at a surface by means of a uniform electronic field that sets up a triangular potential well. This limit is a rough approximation to the confinement of electrons in an n-channel accumulation layer in GaAs at the interface of the GaAs/$Ga_xAl_{1-x}As$ heterojunction which was used by Abstreiter and Ploog [13]. We note that in the "flat-band" limit, the motion of both electrons and holes perpendicular to the slab is "quantized", whereas, in the "bent-band" limit, only that of the electrons is "quantized".

At Γ the wavefunctions for the conduction and valence band states in both models can be written, within the effective mass approximation, in the form

$$\psi(\boldsymbol{x}) = \exp(\mathrm{i}\boldsymbol{\kappa}_{/\!/} \cdot \boldsymbol{x})\, \Phi_{n\sigma}(z)\, \psi_{n\sigma}(\boldsymbol{x}) .$$

Here n stands for conduction (c) or valence (v) band; $\psi_{n\sigma}(\boldsymbol{x})$ is the relevant Bloch function at $\boldsymbol{\kappa} = 0$; and $\Phi_{n\sigma}(z)$ is the envelope function associated with subband σ in the conduction or valence band. For both models the wavevector $\boldsymbol{\kappa}_{/\!/}$ of the electrons parallel to the plane of the 2D plasma is a good quantum-quantum number.

The scattering cross-sections $|M(\boldsymbol{\kappa}_{/\!/})|$ for the two-step and three-step carrier density mechanisms, in the limit $\boldsymbol{q} \to 0$, has the form,

$$|M(\boldsymbol{\kappa}_{/\!/})| = \frac{\langle \mathrm{v}|p_{\mathrm{s}}|\mathrm{c}_\beta\rangle\langle \mathrm{c}_\alpha|p_{\mathrm{i}}|\mathrm{v}\rangle\langle\Phi_{\mathrm{v}}|\exp(\mathrm{i}\boldsymbol{k}_{\mathrm{s}}\cdot\boldsymbol{z})|\Phi_{\mathrm{c}\sigma}\rangle\langle\Phi_{\mathrm{c}\sigma'}|\exp(-\mathrm{i}\boldsymbol{k}_{\mathrm{i}}\cdot\boldsymbol{z})|\Phi_{\mathrm{v}}\rangle}{[\mathcal{E}_{\mathrm{g}} + \mathcal{E}_{\mathrm{v,c}\sigma'}(\ _{/\!/}) - \hbar\omega_{\mathrm{i}}]}\ , \tag{13}$$

where $\langle \mathrm{v}_0|p|\mathrm{c}_0\rangle$ is the momentum matrix element for interband transitions at the $E_0 + \Delta_0$ gap of bulk GaAs; α and β designate the spins of the excitated electrons in the conduction band, which are different in light scattering by spin-flip excitations, but are the same in light scattering by non-spin-flip excitations; $\phi_{\mathrm{c}\sigma}$ and $\phi_{\mathrm{c}\sigma'}$ are the envelope functions for the electrons in the σ and σ' conduction subbands, which are different in the two-step and three-step carrier density mechanisms; ϕ_{v} is the envelope function for the valence band hole, which involves quantized levels only in the flat-band model; $\mathcal{E}_{\mathrm{g}}$ is the band gap at $\boldsymbol{\kappa}_{/\!/} = 0$; $\mathcal{E}_{\mathrm{g}} + \mathcal{E}_{\mathrm{v,c}\sigma'}(\boldsymbol{\kappa})$ is the energy separation of the excited electron in the σ' conduction subband and the hole in the valence band; and in the "bent-band" model, the sum is taken over the valence band states E_{v} which participate in the Franz–Keldysh tunneling-assisted optical transitions. In the dipole approximation, i.e. $\exp(\mathrm{i}\boldsymbol{k}_{\mathrm{s}}\cdot\boldsymbol{z}) \approx 1$, the matrix elements for the envelope functions reduce to overlap integrals; specifically

$$\langle\Phi_{\mathrm{v}}|\exp(\mathrm{i}\boldsymbol{k}_{\mathrm{s}}\cdot\boldsymbol{z})|\Phi_{\mathrm{c}\sigma}|\ \exp(-\mathrm{i}\boldsymbol{k}_{\mathrm{i}}\cdot\boldsymbol{z})|\Phi_{\mathrm{v}}\rangle \cong \langle\Phi_{\mathrm{v}}|\Phi_{\mathrm{c}\sigma}\rangle\langle\Phi_{\mathrm{c}\sigma'}|\Phi_{\mathrm{v}}\rangle\ . \tag{14}$$

In the "flat-band" model, the envelope functions for the quantized levels of both the conduction and valence band are given by

$$\Phi_{n\sigma}(z) = (z/L)^{1/2}\sin(\pi\sigma z/L)\ .$$

The corresponding energies of the quantized levels are given by

$$\mathcal{E}_{\mathrm{c}\sigma}(\boldsymbol{\kappa}_{/\!/}) = \mathcal{E}_{\mathrm{g}} + \frac{\pi^2\hbar^2\sigma^2}{2m_{\mathrm{c}}L^2} + \frac{\hbar^2\kappa_{/\!/}^2}{2m_{\mathrm{c}}}\ , \tag{15}$$

$$\mathcal{E}_{\mathrm{v}\sigma}(\boldsymbol{\kappa}_{/\!/}) = -\frac{\pi^2\hbar^2\sigma^2}{2m_{\mathrm{v}}L^2} - \frac{\hbar^2\kappa_{/\!/}^2}{2m_{\mathrm{v}}}\ . \tag{16}$$

The wavevector components $\kappa_\perp^{\mathrm{v}}$, $\kappa_\perp^{\mathrm{c}}$ normal to the interfaces are confined to the value $\pi\sigma/L$ for both bands, independent of the effective mass. Thus $\langle\Phi_{\mathrm{c}\sigma'}|\Phi_{\mathrm{v}\sigma}\rangle \equiv 0$ unless $\sigma' = \sigma$. The selection rule is analogous to the selection rule $\Delta l = 0$ that applies to the Landau level quantum number for optical interband transitions in an externally applied magnetic field. Since one of the two interband transitions in the two-step and three-step carrier density mechanisms involves $\Delta\sigma = 1$, the contributions from these mechanisms to the scattering by spin-flip and non-spin-flip sp inter-subband excitations are forbidden at this level of approximation. However, there are several ways by which the $\Delta\sigma = 1$ selection rule for the interband transitions may be broken. For one thing the potential barriers that confine the electrons are actually of the order of 1 eV or less. Thus $\kappa_\perp^{\mathrm{c,v}}$ is equal to $\sigma(\pi + \delta_\sigma^{\mathrm{c,v}}\surd L$, where the phase $\delta_\sigma^{\mathrm{c,v}}$ will depend on the effective mass of the carriers which controls the penetration of the wavefunction beyond the barrier and will, therefore, be different for the conduction and valence bands. As a consequence, $\langle\Phi_{c\sigma'}|\Phi_{v\sigma}\rangle$ is non-zero when $\sigma \neq \sigma'$. The electric quadrupole or magnetic dipole contribution to the matrix elements of the envelope function, which depend on the effective wavevector of the EM radiation within the slab, i.e. $k_\perp \approx 2\pi/L$, is also non-zero when $\sigma \neq \sigma'$.

In the Fröhlich mechanism for scattering by the collective excitation at the $E_0 + \Delta_0$ gap, both of the optical interband transitions involve $\Delta\sigma = 0$, specifically, when the electron and hole in the intermediate state both undergo intra-subband scattering. The individual electron and hole contributions to the scattering matrix elements will therefore be non-zero. However, in this case one must still invoke the effect of the finite perpendicular component of the wavevector of the collective excitation to achieve a breakdown in the cancelation of the electron and hole contributions.

We turn next to the "bent-band" model in which the electrons are trapped at the surface by an applied electric field E_0. (The surface space charge field in an accumulation, or inversion layer, is actually highly non-uniform.) The energies of the subband is given by

$$\mathcal{E}_{c\sigma}(\boldsymbol{\kappa}_{/\!/}) = \Delta_\sigma + \frac{\hbar^2\kappa_{/\!/}^2}{2m_c} = \mathcal{E}_g + \left(\frac{e^2E_0^2\hbar^2}{2m_c}\right)^{1/3} x_\sigma + \frac{\hbar^2\kappa_{/\!/}^2}{2m_c} \tag{17}$$

where Δ_σ is the energy of the subband σ at $_{/\!/} = 0$; x_σ is the σth zero of the Airy function $A_i(z)$, and z is the direction of the normal to the surface. The envelope functions $\Phi_{c\sigma}(z)$ for the electrons constitute a set of orthogonal Airy functions. The envelope function for the $\sigma = 1$ (ground state) subband is nodeless. The $\sigma = 2$ (first excited) subband has a single node and each higher subband has an additional node. Holes, on the other hand, are accelerated into the bulk by the electric field; their motion normal to the surface is not quantized, but rather has a continuous distribution of energies. The energy of a hole measured with respect to the valence band edge is given by $\mathcal{E}_v(\ _{/\!/}) = \mathcal{E}_v + \hbar^2\kappa_{/\!/}^2/2m_v$, where $\mathcal{E}_v$ is the total energy associated with the motion perpendicular to the surface, and the classical turning point of the hole orbit is $z_0 = \mathcal{E}_v/eE_0$. The envelope function $\Phi_v(z)$ for the hole is a linear combination of

$$J_{\pm 1/3}\{[2/3(2eE_0m_v)^{1/2}]\,[(z+z_0)^{3/2}]\}$$

which depends on the boundary condition that one imposes on the envelope function at the interface $z = 0$.

When one evaluates the overlap function $\langle\Phi_v|\Phi_{c\sigma}\rangle\,\langle\Phi_{c\sigma'}|\Phi_v\rangle$ using the envelope functions for the "bent-band" model, one finds that, unlike the corresponding overlap function for the "flat-band" model, it is non-zero. This is essentially due to the sizeable perpendicular component of the electron wavevector that is introduced by the localization of the electron wavefunction at the surface. The magnitude of the electron wavevector is typically $\kappa_\perp \simeq (2m_c\Delta_\sigma)^{1/2}/\hbar$. That the overlap function is non-zero can also be viewed as a breakdown of the selection rules by the electric field-induced spatial separation of the electron–hole pairs in the intermediate state, i.e. Franz–Keldysh interband transition between the bent valence band and the conduction subbands.

We note one further difference in the character of the light scattering between the "flat-band" model and the "bent-band" model, namely a quantitative difference in the widths of the resonance excitation (i.e., intensity versus ω) curves. In the case of the "flat-band" model, the width of the resonance excitation curve for $\boldsymbol{q}_{/\!/} = 0$ is $(\hbar^2 k_{F/\!/}^2/2)\,(m_i^{-1} + m_v^{-1}) + 2\gamma_{12}$ [14], which is comparable in magnitude to the width of the resonance excitation curve for scattering by sp charge carrier excitations in bulk semiconductors [12]. On the other hand, in the case of the "bent-band" model, the width of the resonance excitation curve is appreciably greater due to Franz–Keldysh effects.

A brief comment is in order regarding the resonant inelastic light scattering at the E_1 and $E_1 + \Delta_1$ gaps of III–V compound semiconductors. Since, as noted earlier, the conduction band states that participate in the inelastic light scattering processes at the E_1 and $E_1 + \Delta_1$ gaps are not quantized into clearly defined subbands, the matrix elements for inelastic light scattering by the coupled LO phonon–collective inter-subband excitations of 2D plasmas, via the Fröhlich, electro-optic and deformation potential mechanisms, are similar in character to the corresponding matrix elements for the inelastic light scattering by the collective excitations of bulk semiconductors. In the "flat-band" limit, the major difference between the 2D plasma and the 3D plasma resides in the different magnitudes of $q_{\perp ex}$, the perpendicular component of the wavevector of the excitation that participates in the light scattering. In the 2D case, $q_{\perp ex} \simeq 2\pi/L$ and in the 3D case $q_{\perp ex} \simeq q_\perp = k_{i\perp} + k_{s\perp}$. In the "bent-band" limit the difference is due to differences in the magnitude of the band bending that occurs in the 2D plasma and in the surface space charge region of the bulk semiconductor.

6. Concluding remarks

It is evident that resonant inelastic light scattering is a useful spectroscopic probe for investigating the charge carrier excitations of 2D plasmas in semiconductors, whose full potential has not yet been exploited. The use of resonant light scattering should be particularly advantageous in studying the interaction of the sp and collective excitations with LO phonons in polar semiconductors. This would be much more difficult to do using infrared absorption, particularly in the region of the restrahlen frequencies where the dielectric constant of a polar semiconductor is highly dispersive.

In the analysis of the resonant light scattering by inter-subband excitations in 2D plasmas, which we have carried out thus far, we have for simplicity only treated the scattering by charge carrier excitations when $\boldsymbol{q}_{/\!/} = 0$. We are now looking into the scattering by the charge carrier excitations when $\boldsymbol{q}_{/\!/} \neq 0$. On the basis of the dispersion curves for the charge carrier excitations (fig. 2), it is obvious that the widths of the sp inter-subband scattering peaks for $q_{/\!/} \neq 0$ will be appreciably larger than those for $q_{/\!/} = 0$. Also, at the magnitudes of $q_{/\!/}$ which are readily achievable, namely 2×10^5 cm^{-1}, we may expect that the collective inter-subband excitations will be appreciably Landau damped. Moreover, the coupling of the collective inter-subband excitations with the 2D plasma oscillations will also manifest itself. It may even be possible to observe the light scattering by the *intra*-subband excitations.

Of even greater interest is the effect of an externally applied magnetic field on the nature of the sp and collective excitations, and on the character of the light scattering spectra. In the simplest situation, namely that in which the magnetic field is directed normal to the surface, the magnetic field will quantize the motion of the carriers parallel to the surface, so that the two-dimensional subbands will coalesce into Landau levels which have a zero-dimensional density of states. The spin degeneracy will also be lifted. The charge carrier excitations will involve changes in the Landau (orbital) and spin quantum numbers, as well as σ. Furthermore, there will be selection rules regarding the changes in the Landau and spin quantum numbers [31] that are permitted in optical interband transitions, which apply equally to the "bent-band" and the "flat-band" models.

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