



Quantum theory of elementary excitations on semiconducting surfaces and in  
low-dimensional-electronic systems  
by Hong Yu

A thesis submitted in partial fulfillment of the requirements for the degree Doctor of Philosophy  
Physics  
Montana State University  
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Abstract:

We discuss plasmon and phonon excitations in the accumulation layer of a semiconductor with the aid of calculations based on a nonlocal description of dynamical density response in the random-phase-approximation. The first model considered is that of a polarizable jellium slab at finite temperature with surface charges that bend the conduction bands downwards; lattice vibrations are ignored. Choosing model parameters appropriate to lightly doped InAs(110) at room temperature, and to ZnO accumulation layers at low temperature, we obtain intersubband as well as intrasubband plasmons and discuss their dispersion relations, localizations, and line shapes. Evidence for two-dimensional and "acoustic" plasmon is presented. All plasmon modes are strongly damped when their dispersion curves enter the single-particle continua. When the dynamical response of the lattice is included in the model, we obtain coupled plasmon-phonon modes or "plasmaphons" and study their dispersion and line shapes.

Self-consistent calculations of the subband electronic structure as well as the single-particle and collective excitations of semiconductor quantum wires are, for the first time, presented. Calculated energy levels, wavefunctions, charge density and potential for the quantum wire in the GaAs/AlGaAs heterostructure differ from the predictions of the simple particle-in-a box model because of the influence of accumulation and depletion regions. The dynamical structure factor, evaluated in the random-phase-approximation, displays unique line shapes and dispersion relations which are in sharp contrast with the results expected for higher-dimensional electron gases. A well defined spectral feature is attributed to an acoustic plasmon. The calculated intersubband resonance frequency of the quantum wire, shifted upward by the depolarization field, agrees well with previous experiments.

QUANTUM THEORY OF ELEMENTARY EXCITATIONS ON SEMICONDUCTING  
SURFACES AND IN LOW-DIMENSIONAL-ELECTRONIC SYSTEMS

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Hong Yu

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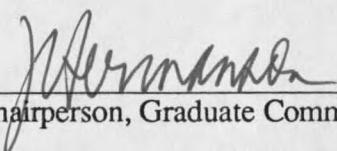
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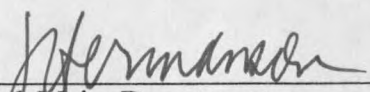
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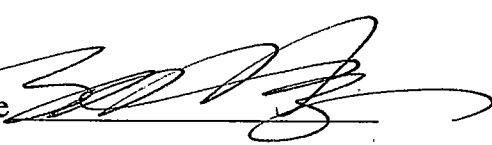
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## ABSTRACT

We discuss plasmon and phonon excitations in the accumulation layer of a semiconductor with the aid of calculations based on a nonlocal description of dynamical density response in the random-phase-approximation. The first model considered is that of a polarizable jellium slab at finite temperature with surface charges that bend the conduction bands downwards; lattice vibrations are ignored. Choosing model parameters appropriate to lightly doped InAs(110) at room temperature, and to ZnO accumulation layers at low temperature, we obtain intersubband as well as intrasubband plasmons and discuss their dispersion relations, localizations, and line shapes. Evidence for two-dimensional and "acoustic" plasmon is presented. All plasmon modes are strongly damped when their dispersion curves enter the single-particle continua. When the dynamical response of the lattice is included in the model, we obtain coupled plasmon-phonon modes or "plasmarons" and study their dispersion and line shapes.

Self-consistent calculations of the subband electronic structure as well as the single-particle and collective excitations of semiconductor quantum wires are, for the first time, presented. Calculated energy levels, wavefunctions, charge density and potential for the quantum wire in the GaAs/AlGaAs heterostructure differ from the predictions of the simple particle-in-a box model because of the influence of accumulation and depletion regions. The dynamical structure factor, evaluated in the random-phase-approximation, displays unique line shapes and dispersion relations which are in sharp contrast with the results expected for higher-dimensional electron gases. A well defined spectral feature is attributed to an acoustic plasmon. The calculated intersubband resonance frequency of the quantum wire, shifted upward by the depolarization field, agrees well with previous experiments.

## CHAPTER 1

## INTRODUCTION

During the past ten years the basic understanding of collective excitations near semiconductor surfaces and interfaces has been deepened through a variety of both experimental and theoretical studies. On the experimental side, several investigations of surface phonons and plasmons in doped semiconductor have been carried out by means of high-resolution-electron-energy-loss spectroscopy(HREELS)<sup>1-7</sup>. HREELS is a particularly useful probe of these excitations because its effective sampling depth is on the order of the width of the space-charge region, namely about 100 Å. Thus the energy-loss spectrum provides important information on the dynamics of an inhomogeneous electron gas.

Until recently, HREELS investigations of plasmon-phonon modes at semiconductor surfaces have been confined to depletion layers. Although collective excitations have been observed in the accumulation layer of the insulator ZnO<sup>8</sup>, studies of an accumulation layer formed on a free semiconductor surface were lacking until now. Mills and his co-workers have performed theoretical studies of surface plasmons on both depletion and accumulation layers on GaAs surface at room temperature<sup>9-12</sup>, using a nonlocal dielectric response formalism in the random-phase-approximation(RPA). Exploiting the RPA formalism, Ehlers<sup>13</sup> performed calculations of surface plasmons in GaAs accumulation layers at various temperatures. His results indicate that at low temperature two-dimensional plasmon behavior<sup>14</sup> becomes manifest in this material, owing to the factor that only the lowest few subbands have significant population. Chen *et al.*<sup>15</sup> have reported studies of collective excitations in InAs(110) accumulation layers. Finally, Yu and Hermanson<sup>16</sup> studied the InAs(110) accumulation layers at room temperature by the dynamical-linear-density-response theory of the conduction electrons

within the RPA, based on the knowledge of the electronic subband structure which was determined by a self-consistent-field method. We found evidence of 2D and acoustic plasmons in the InAs(110) accumulation layer. Our results included the dispersion relations, spatial localizations, and line shapes of intersubband as well as intrasubband plasmons. When the phonon excitations were included, we obtained coupled plasmon-phonon modes or "plasmarens". A more detailed calculation on a strong accumulation ZnO layer was also reported by us<sup>17</sup>. Because of the strong localization of the conduction electrons near the surface (a length scale of about  $10\text{\AA}$ ), microscopic details usually ignored in space-charge layers, such as quantum tunneling into the surface barrier, position dependence of the effective mass, and exchange and correlation effects<sup>18</sup> were taken into account. Some of the excitation modes we found have not yet been observed in experiments.

One-dimensional electron systems have attracted considerable interest following advances in high-resolution lithography which have made it possible to fabricate semiconductor structures that confine the motion of electrons in two directions. This confinement gives rise to quasi-one-dimensional electronic properties. The behavior of the quasi-one-dimensional electron gas is expected to be different dramatically from that of its higher-dimensional counterpart.

## CHAPTER 2

GENERAL THEORY OF ELEMENTARY EXCITATIONS OF AN INHOMOGENEOUS  
ELECTRON GASIntroduction

The interaction between an external electron-beam probing and an inhomogeneous electron gas such as that found in space-charge layers in semiconductors can be separated into two regimes. One is the so called dipole scattering regime, in which the electrons are scattered by the dipole field generated by charge density fluctuations in the electron gas into a very narrow cone with an axis along the specular direction; thus the momentum transfer parallel to the surface is very small. The modes excited in this regime are the highly surface localized phonons and collective excitations due to long range correlations in the medium<sup>19,20</sup>. The other regime is the impact scattering regime where the electrons are scattered by the local atomic or molecular field. The signature of such an interaction is a large momentum transfer parallel to the surface; thus the behavior of the excitation in the medium is single-particle like<sup>19,20</sup>. HREELS spectra are usually collected along the specular direction, and thus they reflect the physical properties within the dipole scattering regime. HREELS has been revealed to be a very powerful technique for the study of surface modes of semiconductors including collective excitations.

Theoretically, in the first Born approximation, the scattering efficiency is proportional to the loss function  $P(\mathbf{q}, \omega)$ <sup>21-23</sup> or, equivalently, the dynamical structure factor  $S(\mathbf{q}, \omega)$ <sup>24</sup>; these are the two most important functions in studying the scattering process due to the richness of the physical information contained in them. The energy-loss

function and the dynamical structure factor are closely related to the density response function  $\chi(\mathbf{r}, \mathbf{r}'; \omega)$ . To determine  $\chi(\mathbf{r}, \mathbf{r}'; \omega)$  in the random-phase-approximation, an integral equation has to be solved numerically. The main purpose of the following few sections is to address the details of the theoretical determination of the scattering efficiency which is directly comparable with the HREELS spectrum.

### Electron Energy Loss Function

In a high-resolution electron-energy-loss experiment, a highly monochromatic electron beam with energy  $E_I$  and momentum  $q_I (\hbar=1)$  impinges on the solid surface, and the scattered electrons of energy  $E_S$  and momentum  $q_S$  are collected by an analyzer. In the scattering process the electron energy loss  $\omega$  and momentum transfer  $q_{||}$  are conserved:  $E_S = E_I - \omega$  and  $q_S = q_I - q_{||}$  (the last subscript denotes the parallel to the surface).

The resulting near-specular scattering process can be described by the scattering efficiency per unit frequency,  $ds/d\omega$ , which is the probability that an electron will be scattered from its initial state into a final state with an energy loss between  $\omega$  and  $\omega+d\omega$ . The quantity may be written as <sup>10,12,25,26</sup>

$$\frac{ds}{d\omega} = \frac{2e^2 v_{\perp}^2}{\pi \hbar} \int d^2 \vec{q}_{||} \frac{P(\vec{q}_{||}, \omega)}{[v_{\perp}^2 q_{||}^2 + (\omega - \vec{v}_{||} \cdot \vec{q}_{||})^2]^2}, \quad (1)$$

where  $v_{\perp}$  and  $v_{||}$  are the components of the incident electron's velocity perpendicular and parallel to the surface. In the above equation  $P(q_{||}, \omega)$  is the so called frequency and wavevector-dependent energy-loss function.

Let us imagine the semiconductor medium with dielectric constant  $\epsilon$  is located in the half-space  $z > 0$  and describe the external charge's trajectory by

$$\vec{R}(t) = (v_{\parallel}t, v_{\perp}|t|), \quad (2)$$

where  $v_{\parallel}$  and  $v_{\perp}$  are defined above. The perturbing potential felt by the semiconductor is<sup>27</sup>

$$\phi_{\text{ext}}(\vec{r}, t) = \frac{-2e}{(1 + \epsilon)[\vec{r} - \vec{R}(t)]}, \quad (3)$$

or <sup>23,28,29</sup>, after Fourier transformation,

$$\phi_{\text{ext}}(q_{\parallel}, \omega; z) = \frac{8\pi e^2 v_{\perp} e^{-q_{\parallel} z}}{(1 + \epsilon)[q_{\parallel}^2 v_{\perp}^2 + (\omega - \vec{q}_{\parallel} \cdot \vec{v}_{\parallel})^2]}. \quad (4)$$

To return to the general discussion, we relate an essential quantity, the so-called induced charge density  $n_{\text{ind}}(\mathbf{r}, \omega)$ , to  $\chi^0(\mathbf{r}, \mathbf{r}; \omega)$  and  $\chi(\mathbf{r}, \mathbf{r}; \omega)$ , which are the non-interacting and interacting density response functions, respectively, by

$$\begin{aligned} n_{\text{ind}}(\vec{r}, \omega) &= \int d\vec{r}' \chi^0(\vec{r}, \vec{r}'; \omega) \phi_{\text{sc}}(\vec{r}'; \omega) \\ &= \int d\vec{r}' \chi(\vec{r}, \vec{r}'; \omega) \phi_{\text{ext}}(\vec{r}'; \omega), \end{aligned} \quad (5-a)$$

where the screened potential is given by

$$\phi_{\text{sc}}(\vec{r}; \omega) = \phi_{\text{ext}}(\vec{r}; \omega) + \phi_{\text{ind}}(\vec{r}; \omega). \quad (5-b)$$

Now, the induced potential  $\phi_{\text{ind}}(\mathbf{r};t)$  and the induced charge density are related by Coulomb's law<sup>27</sup>

$$\phi_{\text{ind}}(\mathbf{r};t) = \frac{e^2}{(1+\epsilon)} \int d\mathbf{r}' \frac{n_{\text{ind}}(\mathbf{r}';t)}{|\mathbf{r} - \mathbf{r}'|} \quad (6)$$

We next insert equations(4) and (5) into (6) and make a Fourier transformation, exploiting the translational invariance along the surface; thus we have the Fourier transformed induced potential,

$$\phi_{\text{ind}}(q_{\parallel}, \omega; z) = - \frac{32\pi^2 e^3 v_{\perp} e^{q_{\parallel} z}}{(1+\epsilon) q_{\parallel} [q_{\parallel}^2 v_{\perp}^2 + (\omega - \vec{q}_{\parallel} \cdot \vec{v}_{\parallel})^2]} \int_0^{\infty} dz' \int_0^{\infty} dz'' e^{-q_{\parallel}(z'+z'')} \text{Im} \chi(q_{\parallel}, \omega; z, z') \quad (7)$$

where  $\chi(q_{\parallel}, \omega; z, z')$  is the reduced density response function, defined as

$$\chi(\vec{r}, \vec{r}'; \omega) = \int \frac{d\vec{q}_{\parallel}}{(2\pi)^2} e^{i\vec{q}_{\parallel} \cdot (\vec{r}_{\parallel}' - \vec{r}_{\parallel})} \chi(q_{\parallel}, \omega; z, z') \quad (8)$$

To complete our formal development is now straightforward. The work done on the external electron by the induced charge is evaluated as

$$W = - \int_{-\infty}^{\infty} dt \vec{v}(t) \cdot \nabla \phi_{\text{ind}}(\vec{r};t) \quad (9)$$

with  $\vec{v}(t) = \vec{v}_{\parallel} - v_{\perp} |t| \vec{e}_z$ .

After some tedious algebra, we find the result

$$W = \int_0^\infty \omega d\omega \left\{ \int \frac{d\vec{q}_\parallel}{(2\pi)^2} \frac{64\pi e^4 v_\perp^2}{(1+\epsilon)^2 [q_\parallel^2 v_\perp^2 + (\omega - \vec{q}_\parallel \cdot \vec{v}_\perp)^2]^2} \int_0^\infty dz \int_0^\infty dz' e^{-q_\parallel(z+z')} \text{Im} \chi(q_\parallel, \omega; z, z') \right\} \quad (10)$$

The work  $W$  (total energy loss) on the electron can be reexpressed through the scattering efficiency,  $ds/d\omega$ , as

$$W = \int_0^\infty d\omega \hbar \omega \left( \frac{ds}{d\omega} \right) \quad (11)$$

By combining equations(10) and (11) one easily obtains the scattering efficiency for semiinfinite geometry at zero temperature,

$$\frac{ds}{d\omega} = \frac{16e^4 v_\perp^2}{\pi \hbar} \int d^2 \vec{q}_\parallel \frac{1}{[q_\parallel^2 v_\perp^2 + (\omega - \vec{q}_\parallel \cdot \vec{v}_\perp)^2]^2} \int_0^\infty dz \int_0^\infty dz' e^{-q_\parallel(z+z')} \text{Im} \chi(q_\parallel, \omega; z, z') \quad (12)$$

Finally, by considering the physical situation of a slab geometry at finite temperature, equation (12) can be generalized as

$$\frac{ds}{d\omega} = \frac{16e^4 v_\perp^2}{\pi \hbar} [1 + n(\omega)] \int d^2 \vec{q}_\parallel \frac{1}{[q_\parallel^2 v_\perp^2 + (\omega - \vec{q}_\parallel \cdot \vec{v}_\perp)^2]^2} \int_0^L dz \int_0^L dz' e^{-q_\parallel(z+z')} \text{Im} \chi(q_\parallel, \omega; z, z') \quad (13)$$

where  $n(\omega)$  is the Bose-Einstein function and  $L$  is the thickness of the slab. Comparing equations(13) and (1), we also find the electron energy loss function  $P(q_{||}, \omega)$

$$P(q_{||}, \omega) = \frac{8e^2}{(1 + \epsilon)^2} [1 + n(\omega)] \int_0^L dz \int_0^L dz' e^{-q_{||}(z+z')} \text{Im} \chi(z, z'; q_{||}, \omega) \quad (14)$$

### Dynamical Structure Factor

In this section, we shall briefly consider an alternate way of describing the inelastic scattering process in terms of the density response function  $\chi(r, r'; \omega)$ . More generally, in the first Born approximation, the weak interaction between the external electron beam and the medium can be modeled via the differential scattering cross section<sup>20,24,30,31</sup>,

$$\frac{d^2\sigma}{d\Omega dE} = \frac{m_p^2 |\vec{q}_s|}{(2\pi)^3 |\vec{q}_I|} v_{\text{ext}}(\vec{q}) S(\vec{q}, \omega) \quad (15)$$

where  $\vec{q}_s$  and  $\vec{q}_I$  are the scattered and incident momenta while  $\vec{q}$  and  $\omega$  are the momentum and energy transfer, respectively. The Fourier transform of the Coulomb interaction is denoted by  $v_{\text{ext}}(\vec{q})$  and  $m_p$  is the mass of the external charged particle. All the intrinsic many-body properties are included in the so called dynamical structure factor  $S(\vec{q}, \omega)$ . This factor was related to the density response function by Griffin and Harris<sup>32,33</sup>; at finite temperature we have

$$S(\vec{q}, \omega) = 2\hbar[1 + n(\omega)] \int d\vec{r} \int d\vec{r}' e^{-i\vec{q} \cdot (\vec{r} - \vec{r}')} \text{Im} \chi(\vec{r}, \vec{r}'; \omega), \quad (16)$$

$\chi(\vec{r}, \vec{r}'; \omega)$  is the density response function as defined in equation(5) and (8). The previous discussion has emphasized that the response function plays a basic role in a variety of physical problems.

### Density Response Function

A central quantity in the theory of the homogeneous electron gas is the density-density correlation function<sup>20,34</sup>,

$$\chi(\vec{r}, \vec{r}'; t) = \frac{i\theta(t)}{\hbar} < [\vec{n}(\vec{r}, t), \vec{n}(\vec{r}', 0)] >_T, \quad (17)$$

where  $n(\vec{r}, t)$  is the charge density operator, and the angular brackets denote a statistical average of the enclosed operator over a statistical ensemble in equilibrium at temperature  $T$ ;  $\theta(t)$  is a step function with respect to the time  $t$ . In the jellium model of a metal or semiconductor surface and quasi-one-dimensional system, the inhomogeneous electron gas can be studied through the same quantity although the problem becomes more complicated due to the lack of translational invariance. For example, the electron-energy-loss function and the inelastic electron scattering cross section which were discussed above, the image potential of a surface<sup>35</sup>, and the damping rate of collective excitations<sup>36</sup> can all be related to the density response function  $\chi(\vec{r}, \vec{r}'; \omega)$ , which is the Fourier transform of equation(17) in time. Zangwill and Soven<sup>34</sup> wrote the interacting density response function  $\chi(\vec{r}, \vec{r}'; \omega)$  explicitly in terms of many-particle quantum states. If  $|0\rangle$  and  $|f\rangle$  are the exact many-particle ground and final state, respectively, then their result is written as

$$\chi(\vec{r}, \vec{r}'; \omega) = \sum_f \left\{ \frac{\langle 0 | \vec{n}(\vec{r}) | f \rangle \langle f | \vec{n}(\vec{r}') | 0 \rangle}{\hbar\omega - (E_f - E_0) + i\delta} - \frac{\langle 0 | \vec{n}(\vec{r}') | f \rangle \langle f | \vec{n}(\vec{r}) | 0 \rangle}{\hbar\omega + (E_f - E_0) + i\delta} \right\}, \quad (18)$$

where the  $E_f$  are energy levels and  $\delta$  a positive infinitesimal. Unfortunately,  $\chi(\mathbf{r}, \mathbf{r}'; \omega)$  cannot be calculated exactly because of the complexity of many-particle states. Yet, we can make one more step without approximation: we can write the density response function of a non-interacting many-particle system  $\chi^0(\mathbf{r}, \mathbf{r}'; \omega)$  exactly since then the non-interacting many-particle states can be built up from single-particle states under the restriction of Fermi-Dirac statistics. So,  $\chi^0(\mathbf{r}, \mathbf{r}'; \omega)$ , the same quantity as defined in equation(5), reads<sup>24,34,37-39</sup>,

$$\chi^0(\vec{r}, \vec{r}'; \omega) = \sum_{i,j} (f_i - f_j) \frac{\phi_i^*(\vec{r}) \phi_j(\vec{r}) \phi_j^*(\vec{r}') \phi_i(\vec{r}')}{\hbar\omega - (\epsilon_j - \epsilon_i) + i\delta}, \quad (19)$$

where  $f_i$  and  $f_j$  are the Fermi distribution function and the single particle states  $i$  and  $j$  can be from the selfconsistent Kohn-Sham problem<sup>40,41</sup> of the many-particle ground state within the local-density formalism.  $\chi^0(\mathbf{r}, \mathbf{r}'; \omega)$  will be a very useful function in the approximation procedure used to derive  $\chi(\mathbf{r}, \mathbf{r}'; \omega)$ : the random phase approximation or the selfconsistent-field approximation which will be discussed in detail in the next section.

### Random-Phase Approximation

Hohenberg, Kohn and Sham<sup>40,41</sup> treated the particle density  $n(\mathbf{r})$  as the basic quantity in their theory of the ground-state properties of a many-particle system, the so-

called density-functional formalism. In this formalism, the wavefunctions  $\phi_i(\mathbf{r})$  satisfy the Schrodinger equation,

$$\left[-\frac{\hbar^2}{2m^*}\nabla^2 + v_{\text{eff}}(\vec{r})\right]\phi_i(\vec{r}) = \epsilon_i\phi_i(\vec{r}) \quad (20)$$

with

$$v_{\text{eff}}(\vec{r}) = v(\vec{r}) + e^2 \int d\vec{r}' \frac{n(\vec{r}')}{|\vec{r} - \vec{r}'|} + v_{\text{xc}}(\vec{r}) \quad (21)$$

where  $v(\mathbf{r})$  is the electron-nuclear potential, and  $v_{\text{xc}}(\mathbf{r})$  is the exchange and correlation potential. The particle density  $n(\mathbf{r})$  in the above equation is given by,

$$n(\vec{r}) = \sum_i f_i |\phi_i(\vec{r})|^2 \quad (22)$$

In general, the Kohn-Sham<sup>40</sup> equations defined by equations(20)-(22) must be solved selfconsistently since the unknown wavefunctions  $\phi_i(\mathbf{r})$  are included in the effective potential  $v_{\text{eff}}(\mathbf{r})$  if one inserts equation(22) into  $n(\mathbf{r})$  in equation (21).

Now, when a weak dynamical external potential  $\phi_{\text{ext}}(\mathbf{r}, \omega)$  is applied to a system of charged particles, whose ground state problem is described above, the induced charge density  $n_{\text{ind}}(\mathbf{r}, \omega)$  is given within linear response theory by

$$n_{\text{ind}}(\vec{r}; \omega) = \int d\vec{r}' \chi(\vec{r}, \vec{r}'; \omega) \phi_{\text{ext}}(\vec{r}'; \omega) \quad (23)$$

In a selfconsistent theory one also introduces the non-interacting response function  $\chi^0(\mathbf{r}, \mathbf{r}'; \omega)$ , defined in equation(19), which satisfies the equation

$$n_{\text{ind}}(\vec{r};\omega) = \int d\vec{r}' \chi^0(\vec{r}, \vec{r}';\omega) \phi_{\text{sc}}(\vec{r}';\omega) \quad (24)$$

Here the screened potential is given by

$$\phi_{\text{sc}}(\vec{r};\omega) = \phi_{\text{ext}}(\vec{r};\omega) + \phi_{\text{ind}}(\vec{r};\omega), \quad (25)$$

and  $\phi_{\text{ind}}(\vec{r};\omega)$  is the induced potential acting on an electron as a consequence of the screening response of the system of charged particles to the external field  $\phi_{\text{ext}}(\vec{r};\omega)$ . We can write  $\phi_{\text{ind}}(\vec{r};\omega)$  as a sum of Hartree and exchange-correlation potential:

$$\phi_{\text{ind}}(\vec{r};\omega) = \frac{e^2}{\epsilon} \int d\vec{r}' \frac{n_{\text{ind}}(\vec{r}',\omega)}{|\vec{r} - \vec{r}'|} + \frac{dv_{\text{xc}}(\vec{r})}{dn(\vec{r})} n_{\text{ind}}(\vec{r},\omega) \quad (26)$$

Because equations(23)-(26) have to be solved simultaneously(selfconsistently), we are led to a integral equation for  $\chi(\vec{r},\vec{r}';\omega)$ <sup>42-44</sup>,

$$\chi(\vec{r},\vec{r}';\omega) = \chi^0(\vec{r},\vec{r}';\omega) + \int d\vec{r}_1 \int d\vec{r}_2 \chi^0(\vec{r},\vec{r}_1;\omega) v(\vec{r}_1,\vec{r}_2) \chi(\vec{r}_2,\vec{r}';\omega), \quad (27)$$

where

$$v(\vec{r}_1,\vec{r}_2) = \frac{1}{\epsilon} \left[ \frac{e^2}{|\vec{r}_1 - \vec{r}_2|} + \frac{dv_{\text{xc}}(\vec{r})}{dn(\vec{r})} \delta(\vec{r}_1 - \vec{r}_2) \right] \quad (28)$$

is the interaction between charged particles. It is important to note that the selfconsistency of the dynamical properties(not the ground state properties) is built into the above integral equation. The induced potential  $\phi_{\text{ind}}(\vec{r};\omega)$  is fully specified once a choice has been made for

the exchange-correlation potential in the particle system. In the case that  $v_{\text{eff}}(\mathbf{r})$  (see equation(21)) and (28)) is approximated by averaging electrostatic interaction(the Coulomb interaction) between the electrons, the approximation is called the random-phase approximation(RPA). In the local-density-approximation(LDA), the potential is composed of not only the bare Coulomb but also the exchange-correlation potential as given in equation(28).

### Single-Particle and Collective(Spin-Density and Charge Density) Excitations

Single-particle excitations are uncorrelated excitations in which an electron is excited from an initial state below the Fermi level to an empty state above it. The collective excitations involve charge-density and spin-density fluctuations and therefore are dynamically screened by the electron-electron interactions. In linear response theory, the density response function  $\chi(\mathbf{r}, \mathbf{r}'; \omega)$  completely describes the dynamical response of the electron system to an external longitudinal field. The physics of the spin-density excitations (SDE) is best rendered by the response function  $\chi^{\text{spin}}(\mathbf{r}, \mathbf{r}'; \omega)$ <sup>53</sup> which includes only the spin-dependent exchange-correlation interaction(the direct Coulomb interaction cannot flip the spin), while the single-particle excitations(SPE) and the charge-density excitations(CDE) can be best studied through the noninteraction response function  $\chi^0(\mathbf{r}, \mathbf{r}'; \omega)$ , and  $\chi^{\text{chg}}(\mathbf{r}, \mathbf{r}'; \omega)$ , respectively. The latter includes both the direct Coulomb interaction and exchange-correlation.

The integral equation for the charge density response function  $\chi^{\text{chg}}(\mathbf{r}, \mathbf{r}'; \omega)$  of the surface problem is given by equation(27). The integral equation for the spin-density response function  $\chi^{\text{spin}}(\mathbf{r}, \mathbf{r}'; \omega)$  has the same as equation(27), but the electron-electron

interaction(equation(28)) is replaced by  $v(\vec{r}_1, \vec{r}_2) = \frac{1}{\epsilon} [\delta(\vec{r}_1 - \vec{r}_2) dv_{xc}(\vec{r}_1, \vec{r}_2, m) / dm]$

where  $m = n_+ - n_-$  is the local spin density. We used the Gunnarsson-Lundqvist exchange-correlation energy to evaluate the integral equation.<sup>53</sup>

To summarize our approach, the incorporation of different forms of the electron-electron interaction in the calculation of  $\chi(\mathbf{r}, \mathbf{r}'; \omega)$  leads to three different results:

- 1) When the energy-loss function  $P(q_{||}, \omega)$  (equation(14)) is evaluated using the noninteraction electron response function  $\chi^0(\mathbf{r}, \mathbf{r}'; \omega)$ , the single-particle excitation (SPE) is revealed;
- 2) The response function  $\chi^{\text{spin}}(\mathbf{r}, \mathbf{r}'; \omega)$ , which involved only the exchange-correlation potential, predicts the spectrum of spin-density excitations (SDE);
- 3) The full Coulomb interaction is included in the response function  $\chi^{\text{chg}}(\mathbf{r}, \mathbf{r}'; \omega)$ . The charge-density excitations (CDE, or plasmons) are found in the calculated spectrum in this case.

The lightscattering method is especially powerful because both spin-density and charge-density excitations can be measured.<sup>45</sup> At small wave vectors the energies of spin-density modes are shifted from single-particle transition energies by exchange-correlation interaction. Charge-density modes have energy shifts due to direct as well as exchange-correlation Coulomb interactions. However, since exchange interactions were expected to be small in GaAs,<sup>45</sup> spin-density excitations were interpreted as single-particle excitation.

Recently, Pinczuk *et al*<sup>45</sup> demonstrated that collective SDE and intersubband SPE as well as CDE of the quasi-two-dimensional electron gas could be measured with inelastic light scattering. The experiments show that exchange-correlation interactions are more important than previously appreciated. We calculated the spectrum of SDE in an accumulation layer of ZnO<sup>53</sup>, based on a formalism just described above. The dispersion relation for the SDE lies below the single-particle continuum at small wavevector, while

the CDE appears above the continuum. The dispersion relations of the spin-density and charge-density modes are very similar to those observed in light scattering experiments carried out on GaAs/AlGaAs quantum wells.<sup>45</sup>

## CHAPTER 3

ELECTRONIC SUBBAND STRUCTURE AND PLASMON EXCITATIONS OF  
QUASI-TWO-DIMENSIONAL ELECTRON GASIntroduction

By quasi-two-dimensional electron gas(Q2DEG) we mean a system in which the electrons are free to move in two spatial dimensions but have their motion constrained in the third dimension. If the range of the confining potential in that dimension is comparable with the de Broglie wavelength we observe new quantum phenomenon: the development of quantum-confined states, with energy levels  $E_0, E_1, \dots$  which are called electronic subbands. The quasi-two-dimensional electron system is reduced to the extreme quantum limit, which is called a two-dimensional electron system<sup>46</sup>, when the lowest subband contains all the electrons of the system. The great current interest in Q2DEG is in electron systems of reduced dimensionality, in semiconductors such as GaAs/AlGaAs heterojunctions<sup>47</sup>, multiquantum wells<sup>48</sup>, inversion layers and accumulation layers<sup>16,17</sup>. Our principal interest here is the systems of electrons in accumulation layers on InAs(110) and ZnO surfaces, and in GaAs/AlGaAs heterostructures.

HREELS and inelastic light scattering have been widely used to characterize the electronic subband properties and the free carrier excitations in semiconductors. Figure 1 shows an example of single-particle transitions in a degenerate quasi-2D electron system. Each confined state of the quantum well is associated with a subband in the two-dimensional wavevector space for electron motion in the plane. Figure 1(a) shows a vertical inter-subband transition between the two lowest subbands( $E_{01}=E_1-E_0$ ). Figure 1(b) represents an intra-subband transition with a wavevector transfer  $q_{||}$ . Such excitation within one subband is only possible if the wavevector transfer  $q_{||}$  is non-zero. Figure 1(c) shows

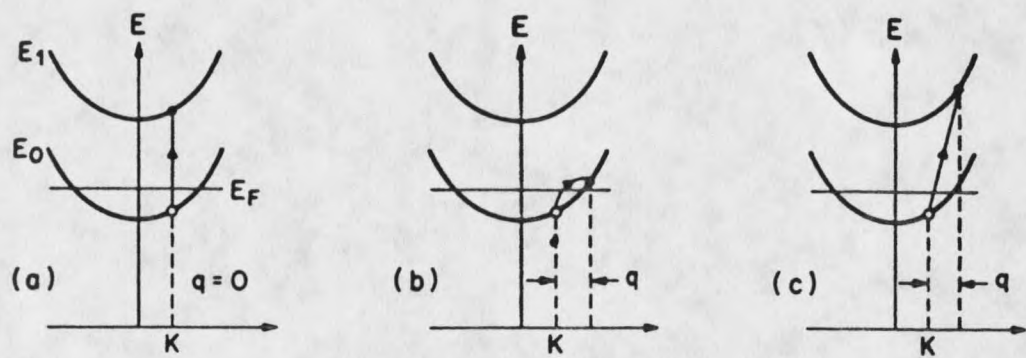


Fig.1 Electronic excitations in quasi-2D systems (a) vertical inter-subband excitation; (b) intrasubband excitation; (c) non-vertical inter-subband excitation.

a non-vertical inter-subband excitation. This excitation has a continuum of energies centered at  $E_{01}$ . The collective intra- and inter-subband excitations are two-dimensional plasmon oscillations of the electrons parallel and perpendicular to the surface, respectively. Although the collective and single-particle excitations have different energies, we can expect from Figure 1 that the frequencies for collective inter-subband and intra-subband transitions are finite and zero, respectively, when the wavevector transfer  $q_{||}$  vanishes. In the next two sections we present detailed calculations of (1) the self-consistent electronic structure of the subbands and (2) the dynamical response of the electron-phonon system, for accumulation layers on InAs<sup>50</sup> and ZnO<sup>51,52</sup>. These systems (especially the latter one) possess a strongly localized electron gas. One would anticipate that such systems exhibit 2D behavior<sup>46,53</sup> because a large majority of the conduction electrons occupy the lowest few subbands, whose spatial extension is much smaller than a typical wavelength involved in HREELS.

### Accumulation Layer on InAs (110) Surface

#### Model and RPA Formalism

A slab of semiconducting material with thickness  $L$  large enough to simulate a semiinfinite geometry is considered as our model. In the slab the conduction electrons are treated as a free-electron gas with effective mass  $m^*$ , situated in a uniformly charged jellium background with volume charge density  $n_D$  due to the donor ions. Auxiliary infinite potential barriers at  $z=0$  and  $z=L$  are introduced on both sides of the slab to enforce the boundary condition that the conduction-electron wavefunction vanishes there<sup>10,11</sup>. A positive surface charge density  $n_s$  is assumed on both surfaces of the slab. By considering the overall charge neutrality of the slab, if  $n(z)$  is the electron density at  $z$  and  $n_{tot}$  is the integral of  $n(z)$  over the slab, we have

$$n_{\text{tot}} = n_D L + 2n_s.$$

(29)

Taking into account the translational invariance in the surface parallel to the slab, the wave function of a conduction electron has the form

$$\varphi_{\vec{k}_{\parallel}, i}(\vec{x}_{\parallel}, z) = \frac{1}{A^{1/2}} e^{i\vec{k}_{\parallel} \cdot \vec{x}_{\parallel}} \phi_i(z), \quad (30)$$

where  $x_{\parallel}$  and  $k_{\parallel}$  are position and wave vector in the surface plane. Inserting equation(30) into the Schrodinger equation appropriate to our model one finds that the electron energy eigenvalues can be written as

$$\epsilon_i(\vec{k}_{\parallel}) = \frac{\hbar^2 \vec{k}_{\parallel}^2}{2m^*} + \epsilon_i \quad (31)$$

The wavefunctions  $\phi_i(z)$  and the energy levels  $\epsilon_i$  which correspond to the motion of the electron normal to the surface ( $z$ -direction) are related to the electrostatic potential  $v(z)$  by the one-dimensional Schrodinger equation

$$\left[ \frac{-\hbar^2}{2m^*} \frac{d^2}{dz^2} + v_{\text{eff}}(z) \right] \phi_i(z) = \epsilon_i \phi_i(z), \quad (32)$$

where  $v(z)$  is the Hartree potential given by

$$v_{\text{eff}}(z) = -\frac{2\pi e^2 L}{\epsilon_{\infty}} \int_0^L [n(z') - n_D] |z - z'| dz' \quad (33)$$

which is the solution of Poisson's equation for charge density  $-e[n(z)-n_D]$ . In equation(33)  $n(z)$  is the free-carrier density at depth  $z$ , given at finite temperature by<sup>10</sup>

$$n(z) = \frac{m^*}{\pi \hbar^2 \beta} \sum_i \ln[1 + e^{-\beta(\epsilon_i - \mu)}] |\phi_i(z)|^2 \quad (34)$$

where  $\beta = 1/k_B T$  and  $\mu$  is the chemical potential; the logarithmic factor in equation(34) is essentially the subband occupation number. The chemical potential is determined by the condition of charge neutrality for the slab:

$$n_{\text{tot}} = \frac{m^*}{\pi \hbar^2 \beta} \sum_i \ln[1 + e^{-\beta(\epsilon_i - \mu)}] \quad (35)$$

Equations(32)-(35) define our self-consistent problem to be solved.

It is convenient to work in Fourier space appropriate to the slab geometry. In order to satisfy the boundary conditions for free wavefunctions, namely  $\phi_i(0)=0$  and  $\phi_i(L)=0$ , we expand the wavefunctions  $\phi_i(z)$  in a Fourier sine series:

$$\phi_i(z) = \left[\frac{2}{L}\right]^{1/2} \sum_{j=1}^{\infty} b_{ij} \sin\left(\frac{j\pi z}{L}\right) \quad (36)$$

The Schrodinger equation can then be transformed into a matrix form<sup>11,36</sup>

$$\sum_{j=1}^{\infty} M_{ij} b_{ij} = \epsilon_i b_{ij} \quad i, j = 1, 2, 3, \dots, \quad (37)$$

where

$$M_{jj'} = \frac{\hbar^2}{2m^*} \left[ \frac{j\pi}{L} \right]^2 \delta_{jj'} + \frac{2}{L} \int_0^L dz \sin\left(\frac{j\pi z}{L}\right) \sin\left(\frac{j'\pi z}{L}\right) v(z) \quad (38)$$

In our numerical work a basis of 20 to 40 sine waves was found to be adequate to expand the wavefunction. We have performed calculations of the self-consistent potential and charge density for both depletion layers and accumulation layers in GaAs and InAs. The results from our calculations for GaAs are in excellent agreement with those presented previously.<sup>9-13</sup>

The dynamical response of the electron system is described by the RPA formalism developed earlier<sup>10</sup>. The full RPA density response function  $\chi(q_{||}, \omega; z, z')$  that describes the nonlocal density response of the slab is obtained by solving the integral equation

$$\chi(q_{||}, \omega; z, z') = \chi^0(q_{||}, \omega; z, z') - \int_0^L dz_1 \int_0^L dz_2 \chi^0(q_{||}, \omega; z, z_1) v_e(q_{||}; z_1, z_2) \chi(q_{||}, \omega; z_2, z') \quad (39)$$

where  $v_e(q_{||}; z, z')$  is the two-dimensional Fourier transform of the electron-electron interaction in the slab, not the Hartree potential of equation(33). We obtained the function  $v_e(q_{||}; z, z')$  by solving the Poisson equation for an external electron:

$$\left( \frac{d^2}{dz^2} - q_{||}^2 \right) v_e(q_{||}; z, z') = -\frac{4\pi e^2}{\epsilon_{\infty}} \delta(z - z') \quad (40)$$

together with the usual continuity conditions for  $v_e(q_{||}; z, z')$  and  $\epsilon(z)(d/dz)v_e(q_{||}; z, z')$  at  $z=0, L$ .

Using standard algebra for obtaining Green's function we have

$$\begin{aligned}
v_e(q_{\parallel}, z, z') = & \frac{2\pi e^2}{\epsilon q_{\parallel}} \{e^{-q_{\parallel}|z-z'|} \\
& + \frac{2}{[(\frac{\epsilon+1}{\epsilon-1})e^{2q_{\parallel}L} - 1]} [(\frac{\epsilon+1}{\epsilon-1})e^{-q_{\parallel}L} \cosh(q_{\parallel}(z+z'-L)) + \cosh(q_{\parallel}(z-z'))]\}
\end{aligned}
\tag{41}$$

In equation(39) the response function for non-interacting electrons is given by

$$\chi^0(q_{\parallel}, \omega; z, z') = \sum_{\alpha, \alpha'} S_{\alpha, \alpha'}(q_{\parallel}, \omega) \phi_{\alpha}(z) \phi_{\alpha}(z') \phi_{\alpha'}(z) \phi_{\alpha'}(z')
\tag{42}$$

where

$$S_{\alpha, \alpha'}(q_{\parallel}, \omega) = 2 \int \frac{d^2 \vec{k}_{\parallel}}{(2\pi)^2} \frac{f(E_{\vec{k}_{\parallel}, \alpha}) - f(E_{\vec{k}_{\parallel} + \vec{q}_{\parallel}, \alpha'})}{E_{\vec{k}_{\parallel} + \vec{q}_{\parallel}, \alpha'} - E_{\vec{k}_{\parallel}, \alpha} + \omega + i\eta},
\tag{43}$$

and

$$f(E_{\vec{k}_{\parallel}, \alpha}) = \frac{1}{e^{[\beta(\hbar^2 \vec{k}_{\parallel}^2 / 2m^* + \epsilon_{\alpha} - \mu)]} + 1}
\tag{44}$$

is the Fermi-Dirac distribution function. In our numerical work, the parameter  $\eta$  is chosen to be finite but very small and the double summation in equation(42) runs over all the energy levels obtained for the slab.

By using the double-cosine series representation for the response function<sup>36,38</sup>,

$$\chi(q_{\parallel}, \omega; z, z') = \sum_{m, n} \chi_{m, n}(q_{\parallel}, \omega) \cos\left(\frac{m\pi z}{L}\right) \cos\left(\frac{n\pi z}{L}\right),
\tag{45}$$

we transform the integral equation(39) for  $\chi(q_{||}, \omega; z, z')$  into the matrix equation

$$\chi_{m,n}(q_{||}, \omega) = \chi_{m,n}^0(q_{||}, \omega) - \sum_{m',n'} \chi_{m,m'}^0(q_{||}, \omega) v_{em'n'}(q_{||}) \chi_{n,n'}(q_{||}, \omega). \quad (46)$$

In the latter equation the coefficients of the double-cosine Fourier transformation of  $\chi^0(q_{||}, \omega; z, z')$  are given by

$$\chi_{m,n}^0(q_{||}, \omega) = \frac{\mu_m \mu_n}{L^2} \sum_{\alpha, \alpha'} S_{\alpha, \alpha'}(q_{||}, \omega) W_{\alpha, \alpha'}^m W_{\alpha, \alpha'}^n, \quad (47)$$

where the matrix  $S_{\alpha, \alpha'}(q_{||}, \omega)$  is given by equation(43) and

$$W_{\alpha, \alpha'}^m = \frac{1}{2} \sum_{\beta, \beta'} b_{\alpha\beta} b_{\alpha'\beta'} (\delta_{m, \beta-\beta'} + \delta_{m, \beta'+\beta} - \delta_{m, \beta+\beta'}) \quad (48)$$

The matrix element  $v_{emn}(q_{||}, \omega)$ , in equation(46), is the double-cosine Fourier transform of the electron-electron interaction given in equation(41), or

$$\begin{aligned} v_{emn}(q_{||}, \omega) = & \frac{2\pi e^2}{\epsilon} \frac{1}{q_{||}^2 + (\frac{n\pi}{L})^2} \left\{ \frac{2L}{(\mu_m \mu_n)^{1/2}} \delta_{mn} - [1 + (-1)^{m+n}] \frac{q_{||} [1 - (-1)^m e^{-q_{||}L}]}{q_{||}^2 + (\frac{n\pi}{L})^2} \right. \\ & + \left( \frac{\epsilon+1}{\epsilon-1} \right) e^{q_{||}L} \frac{q_{||}}{[(\frac{\epsilon+1}{\epsilon-1})^2 e^{2q_{||}L} - 1]} \frac{1}{[q_{||}^2 + (\frac{n\pi}{L})^2]} [4 \cosh q_{||}L - 2((-1)^m + (-1)^n)] \\ & \left. + \frac{q_{||}}{[(\frac{\epsilon+1}{\epsilon-1})^2 e^{2q_{||}L} - 1]} \frac{1}{[q_{||}^2 + (\frac{n\pi}{L})^2]} [-4 - 2((-1)^m + (-1)^n) \cosh q_{||}L] \right\} \quad (49) \end{aligned}$$

In our numerical calculations of the electron-energy-loss spectrum, most of the CPU time is required for the calculation of the matrix elements  $S_{\alpha, \alpha'}(q_{||}, \omega)$ . Without loss of generality, we choose  $q_{||}$  parallel to the y-axis. With this choice  $k_{||x}$  appears only in the

Fermi-Dirac function in  $S_{\alpha,\alpha'}(q_{||},\omega)$ ; the integral with respect to  $k_{||x}$  in equation(43) is simply the Fermi-Dirac integral. With reducing the 2-D integral to a 1-D integral, about 45 seconds of CPU time on a VAX 8550 are required to calculate one point on the surface  $P(q_{||},\omega)$ .

### Results and Discussion

It is convenient to adopt thermal units of length and energy<sup>10,11</sup>. In the figures that follow the energy will be scaled in units of  $k_B T$ , and the length in units of the thermal de Broglie wavelength  $\lambda = (\hbar^2 / 2m^*k_B T)^{1/2}$  or 81.8 Å for InAs at room temperature,  $T=300K$ . The units of wavevector are  $\lambda^{-1}$ . Thus  $\hbar\omega = k_B T$  corresponds to energy 25.9 meV and  $q_{||}\lambda=1$  to wavevector  $1.22 \times 10^6 \text{ cm}^{-1}$ . For InAs, the model parameters we used are  $m^*=0.022 m_0$  when  $m_0$  is the free-electron mass, static dielectric constant  $\epsilon_0=14.9$ ,  $\epsilon_{\infty}=12.3$ , and  $L=20 \lambda$ . The latter choice corresponds to a slab thickness of 1636 Å, which is large compared with the characteristic width of the accumulation layer, about 100 Å. We emphasize that typical values for  $q_{||}\lambda$  in HREELS in the range 0.1 to 0.3. In this range  $q_{||}L$  is about 4, so we expect that our slab model is thick enough to simulate a semi-infinite solid; indeed, we find no evidence for a splitting of plasmon modes into even- and odd-parity partners which would signal an interaction between the modes of the opposite sides of the slab.

Figure 2 shows the self-consistent electron density profile computed for a donor density  $n_D=1.3 \times 10^{16} \text{ cm}^{-3}$  and a surface charge density  $n_s=1.5 \times 10^{12} \text{ cm}^{-2}$  on each side of the slab. These parameters were chosen because the free carrier density for the sample studied by HREELS was about  $1.3 \times 10^{16} \text{ cm}^{-3}$  and because surface charge densities in the range of  $10^{11}$  to  $10^{12} \text{ cm}^{-2}$  are required to fit the experimental data<sup>15</sup>. We also show the self-consistent potential and the subband energy level diagram at  $k=0$  in Fig.2. We

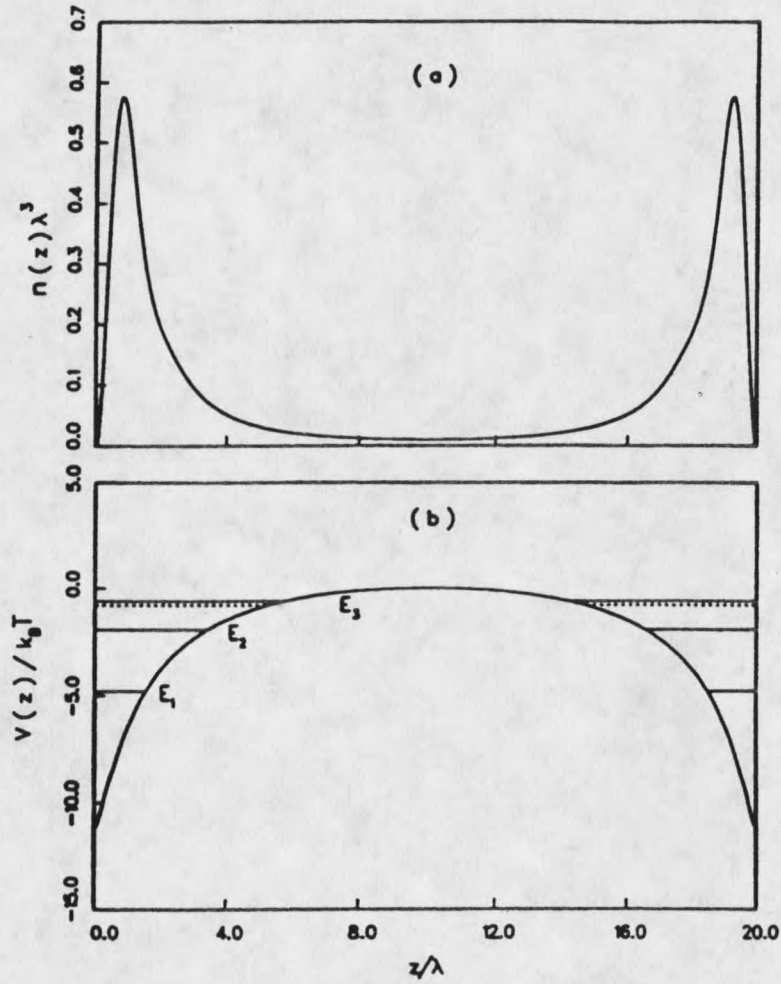


Fig. 2 (a) Self-consistent charge density and (b) potential for InAs accumulation layer;  $n_D = 1.3 \times 10^{16} \text{ cm}^{-3}$ ,  $n_s = 1.5 \times 10^{12} \text{ cm}^{-2}$ , and  $L = 20 \lambda$ . The subband energy levels  $E_i$  at  $k=0$  are also shown in (b), as well as the chemical potential  $\mu$  (dashed line). Dimensionless units are used for the axes.

emphasize that the three lowest subbands are all twofold degenerate; the degeneracy disappears only for bound states very close to the bottom of the bulk conduction band. This behavior indicates that our slab is thick enough to prevent significant overlap between electronic wavefunctions on different sides of the slab. We find that only two subbands are below the Fermi level ( $E_F = -0.8k_B T$ ), with 70% of the electrons occupying the lowest subband. Also, the maximum value of the electron density for the accumulation layer exceeds that in the bulk by a factor of about 80. Thus we anticipate 2-D character for our computed plasmon modes<sup>53</sup>. The band bending corresponding to the accumulation layer shown in Fig.2(a) is 0.29 eV. This agrees reasonably well with the experimental value of 0.22 eV obtained for InAs (110) with a donor concentration of  $1.3 \times 10^{16} \text{ cm}^{-3}$  and a saturation hydrogen exposure<sup>15</sup>.

To obtain an explicit expression for the cross-section of inelastic electron scattering we compute the loss function  $P(q_{||}, \omega)$  corresponding to energy and wavevector transfers  $\hbar\omega$  and  $q_{||}$ , respectively. For the slab model the loss function, within the dipole approximation, is given by<sup>10,11</sup>

$$P(q_{||}, \omega) = \frac{8e^2}{|1 + \epsilon|^2} [n(\omega) + 1] \int_0^L dz \int_0^L dz' e^{-q_{||}(z+z')} \text{Im}\{\chi(q_{||}, \omega; z, z')\} \quad , \quad (50)$$

where  $n(\omega)$  is the Bose-Einstein distribution function. Figure 3 shows our calculated loss function for different values of the surface wavevector  $q_{||}$  in the range pertinent to HREELS; these results were derived using the charge density shown in Fig.2. At long wavelength,  $q_{||}\lambda \leq 0.1$ , the loss function contains four narrow peaks, at 0.32, 0.88, 1.54 and 3.14 in units of  $k_B T$ . The first and second peaks are identified as intrasubband plasmons associated with the second and first subbands, respectively; in particular, the dominant peak in Fig.3 belongs mainly to the lowest subband. The frequency of the latter

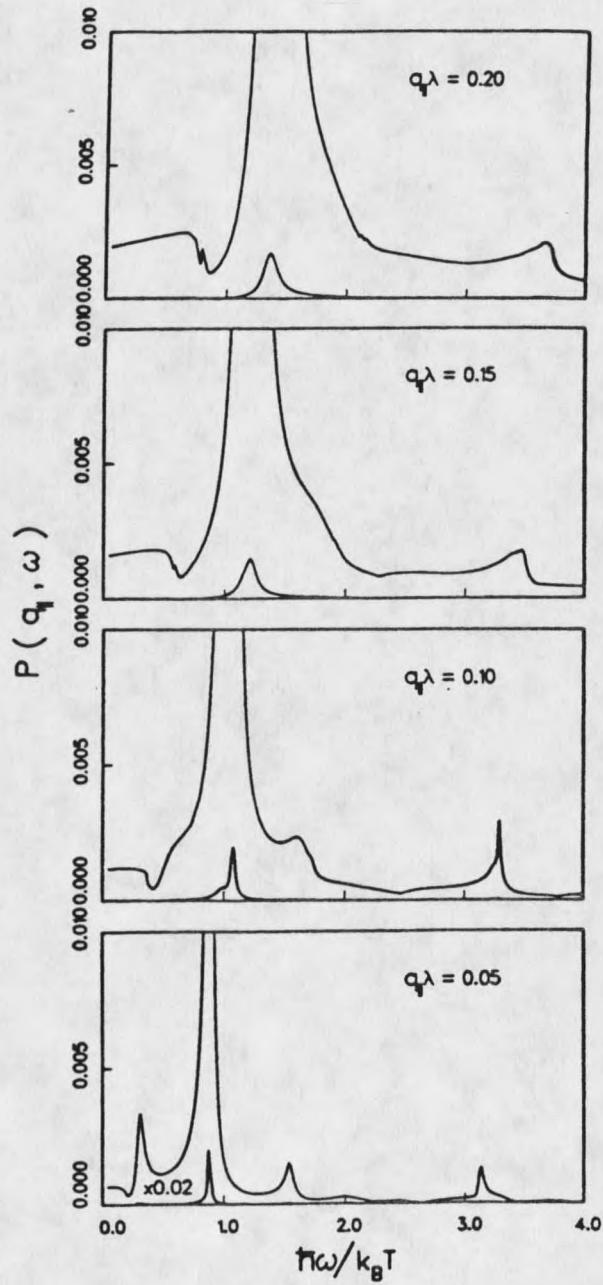


Fig. 3 Electron energy loss function  $P(q_{\parallel}, \omega)$  for four different values of the wavevector  $q_{\parallel}$ . Model parameters are the same as in Fig.2. The frequency is expressed in dimensionless units;  $P(q_{\parallel}, \omega)$  is given in units of  $2m^*e^2/\hbar^2$ .

peak increases roughly as the square root of the wavevector, as expected for a two-dimensional electron gas<sup>53</sup>. The lower-energy peak appears to disperse linearly with the wavevector at long wavelengths, so we tentatively assign it to an acoustic plasmon. An acoustic plasmon can arise in two-component plasmons in which the two electron gases have different spatial extent; it may be described as a plasmon belonging to the second subband, whose polarization field is screened by the charges in the first subband.<sup>54-57</sup>

The third peak and fourth peaks seen in Fig.3 at long wavelengths can be interpreted by noting that the energy differences between the first and second subbands in Fig.2, and between the second and third, are  $E_{12}=2.88k_B T$  and  $E_{23}=1.34k_B T$ , respectively. The high-energy loss peaks in Fig.3 are very close to these thresholds at long wavelengths, so it is natural to associate the peaks with "intersubband" plasmons.<sup>13</sup> The energy and dispersion of these modes will be discussed later.

Electron-hole pair excitations also contribute to the loss functions shown in Fig.3. Note the pronounced dip on the low-energy side of the main peak, which moves to higher energy as the wavevector is increased. This feature marks the high-energy edge of the pair-excitation spectrum belonging to the first subband; the associated continuum extends smoothly to zero energy in Fig.3. The acoustic plasmon appears near this dip at small wavevectors. Intersubband pair excitations are also present in Fig.3, providing a smooth background which is strongest for larger wavevectors.

A convenient way to investigate the spatial localization of plasmon modes is to plot the induced charge density at the appropriate frequency and wavevector. The potential and the charge density induced by the external field  $\phi_{\text{ext}}(q_{\parallel}, \omega; z)$  are given by<sup>13</sup>

$$n_{\text{ind}}(q_{\parallel}, \omega; z) = \int_0^L dz' \chi(q_{\parallel}, \omega; z, z') \phi_{\text{ext}}(q_{\parallel}, \omega; z') \quad (51)$$

$$\phi_{\text{ind}}(q_{\parallel}, \omega; z) = \int_0^L dz' v_e(q_{\parallel}, \omega; z, z') n_{\text{ind}}(q_{\parallel}, \omega; z') \quad (52)$$

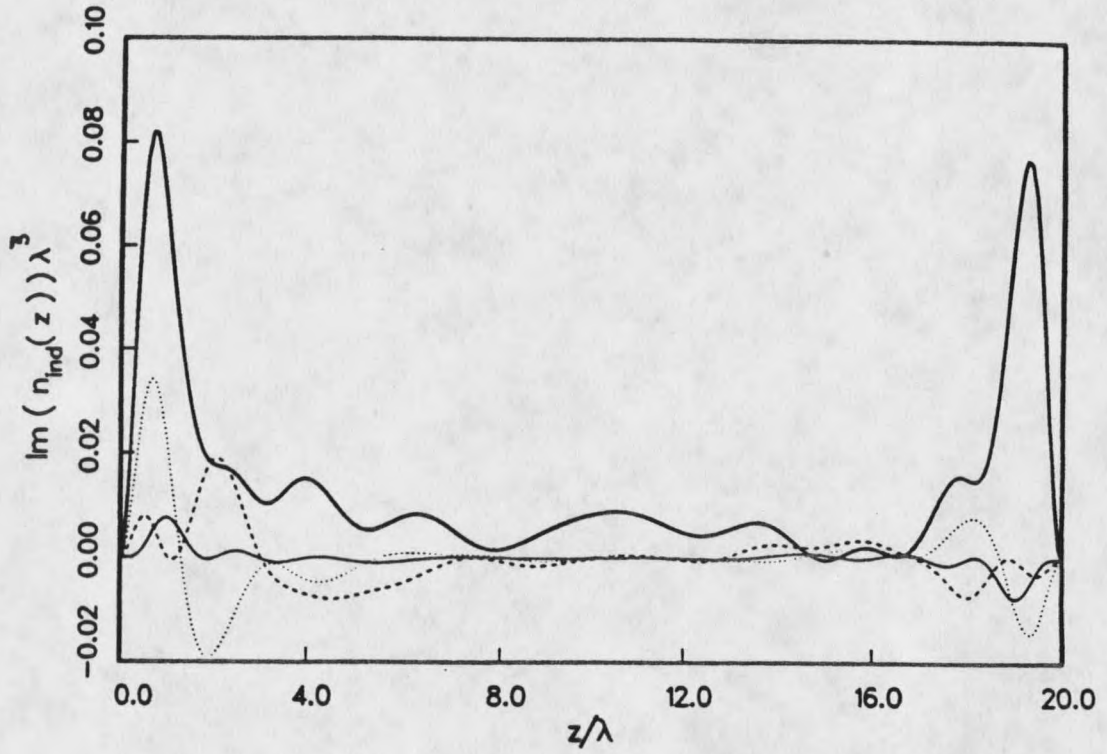


Fig. 4 Imaginary part of the induced charge density for  $q\parallel\lambda=0.05$ . The frequencies were chosen at the maxima of the loss function in Fig.3. The thin and thick solid lines give the results for intrasubband plasmons with  $\hbar\omega/k_B T=0.32$  and  $0.88$ , respectively; the dashed and dotted lines correspond to intersubband modes with  $\hbar\omega/k_B T=1.54$  and  $3.14$ .

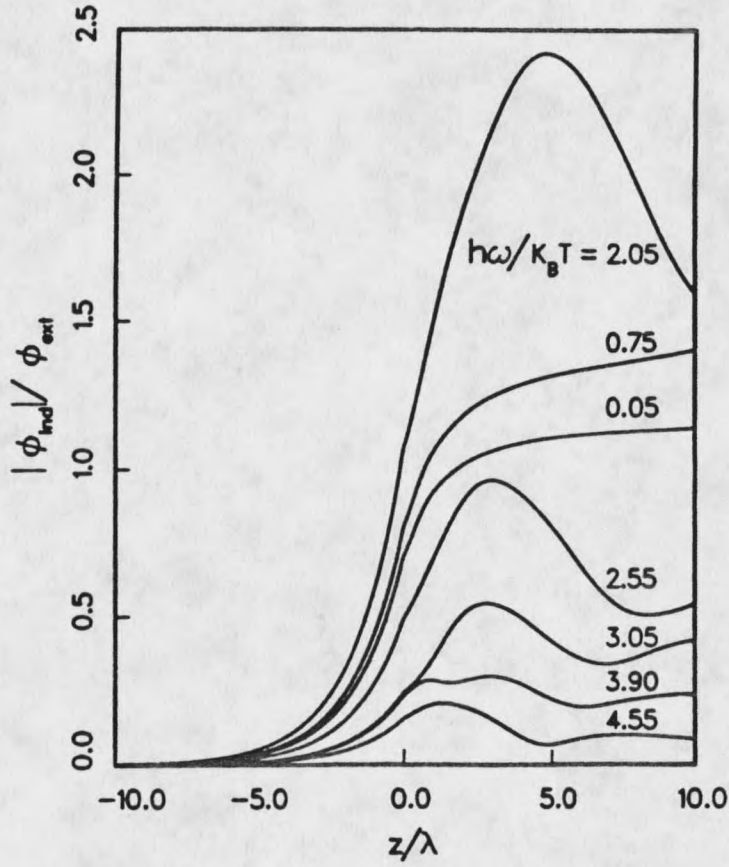


Fig. 5 Absolute value of the induced potential divided by the external field  $\phi_{\text{ext}}$ , for various frequencies as shown in the figure.  $q_{\parallel}\lambda=0.3$ ; other parameters are given in Fig.2.

with  $\phi_{\text{ext}}(q_{\parallel}, \omega; z)$  the external field. We choose the external field  $\phi_{\text{ext}}$  as the Coulomb potential associated with an electron approaching the surface. Its Fourier transform has the form of a decaying exponential inside the slab:  $\phi_{\text{ext}} \propto \exp(-q_{\parallel} z)$ . The induced charge densities calculated by equation(51) are shown in Fig.4 for  $q_{\parallel}\lambda=0.05$  and dimensionless energies 0.32, 0.88, 1.54, 3.14 corresponding to the four plasmon modes exhibited in the first panel of Fig.3. The distance between successive maxima in Fig.4 is determined by the screening length and is similar for all the modes. Note that all four modes are localized within a few hundred angstroms of the surface; thus, little effect of the finite slab thickness is expected. The induced potential (see equation(52)) shown in Fig.5 is in qualitative agreement with Ehlers' results for GaAs<sup>13</sup>. At the intersubband resonance frequency, the induced potential has its maximum very close to the surface. The maxima are slightly deeper inside the surface for frequencies off resonance. This behavior is closely related to that of the induced charge density. For better understanding of the behavior of the elementary excitations in the accumulation layer, we plot their dispersion curves in Fig.6. The solid lines are obtained from the peak positions of the loss function  $P(q_{\parallel}, \omega)$ . For comparison we also show the pair continua and the dispersion relations of ideal 2-D plasmons belonging to the first and second subbands. The 2-D frequency is computed according to the zero-temperature RPA formula<sup>53,58</sup>

$$\omega_{2D}^i = \left\{ \frac{2\pi n_i e^2}{m^*} \left[ \frac{2}{1 + \epsilon_{\infty}} \right] \right\}^{1/2} q_{\parallel}^2 \quad (53)$$

where  $n_i$  is the number of electrons per unit area in subband  $i$ . Except for large  $q_{\parallel}$  the second mode in Fig.6 matches the 2-D dispersion relation closely. We also see that the intersubband modes follow the upper edge of their respective continua. The lowest-energy dispersion curve in Fig.6 is essentially linear at small wavevectors (this was checked by computing the loss function for  $q_{\parallel}\lambda=0-0.06$  in increments of 0.01), as expected for an

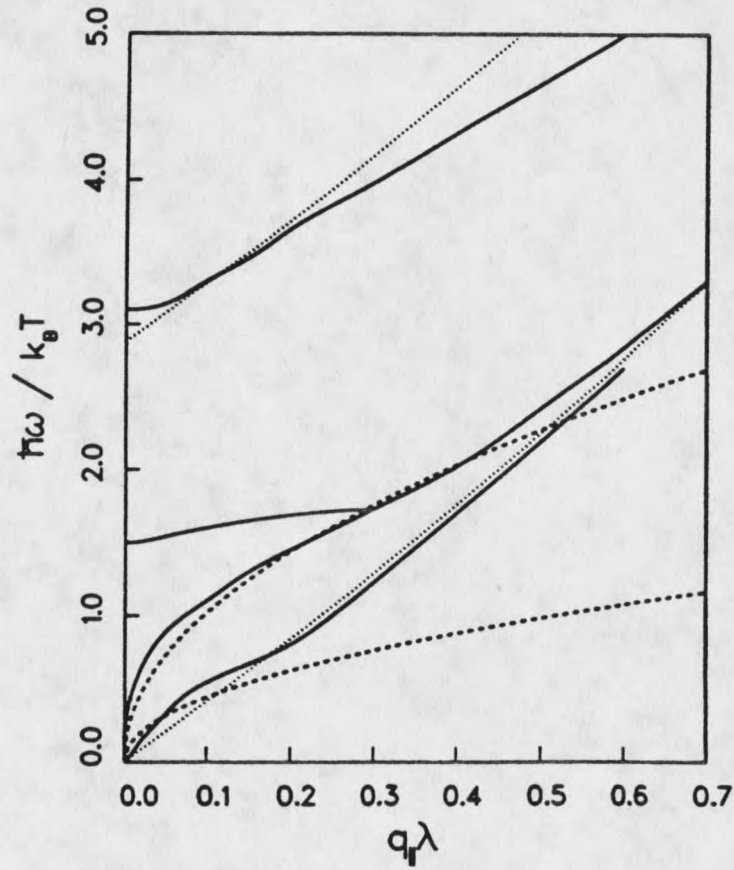


Fig. 6 Plasmon dispersion curves (solid lines) obtained from the maxima of the loss function  $P(q_{\parallel}, \omega)$ , and ideal 2-D plasmon dispersion (dashed lines). The dotted lines show the edges of the single-particle continua associated with the first and second subband. Dimensionless units are used for both axes.

acoustic plasmon<sup>54-57</sup>. At intermediate wavevectors the latter dispersion is like a 2-D plasmon; it deviates from the 2-D model when it approaches the continuum.

We now consider the intersubband plasmons in more detail at long wavelengths. The intersubband energy difference  $E_{12}=2.88$  in units of  $k_B T$  for  $n_s=1.5 \times 10^{12} \text{cm}^{-2}$ . At very small wavevector  $q_{\parallel} \lambda=0.01$  ( $q_{\parallel}=1.22 \times 10^{-4} \text{cm}^{-1}$ ), the resonance frequency obtained from the maximum of the function  $P(q_{\parallel}, \omega)$  is 3.10 in units of  $k_B T$ . This upshift of the frequency for the intersubband transition is due to the well-known depolarization effect<sup>58</sup>. We have calculated the Coulomb matrix elements  $V_{klmn}$  to better understand this frequency shift, which was first discussed by Dahl and Sham<sup>58</sup> in their mode calculations. The shifted intersubband resonance frequency is given by

$$\omega_{12}^2(q_{\parallel} \rightarrow 0) = E_{12}^2(1 + 2\Delta_{12}), \quad (54)$$

where the depolarization shift

$$\Delta_{ij} = n_s^{ij} V_{ijij} / E_{ij}, \quad (55)$$

and the Coulomb matrix elements

$$V_{klmn} = \int_0^L dz \int_0^L dz' \phi_k(z) \phi_l(z) \phi_m(z') \phi_n(z') v_e(q_{\parallel} \rightarrow 0; z, z') \quad (56)$$

In equation(55),  $n_s^{ij}$  is the charge density difference between subbands  $i$  and  $j$ . In the slab model, equation(56) can be reduced to the sum

$$V_{klmn} = \left(\frac{2}{L}\right)^2 \sum_{s_1, s_2, s_3, s_4} b_{ks_1} b_{ls_2} b_{ms_3} b_{ns_4} \int_0^L dz \int_0^L dz' \sin \frac{s_1 \pi z}{L} \sin \frac{s_2 \pi z}{L} \sin \frac{s_3 \pi z'}{L} \sin \frac{s_4 \pi z'}{L} v_e(q_{||}; z, z') \quad (57)$$

with  $v_e(q_{||}, z, z')$  given by equation(41). To derive the frequency shift we calculate the Coulomb matrix element  $V_{1212}$  for  $n_s = 1.5 \times 10^{12} \text{ cm}^{-2}$  ( $n_s \lambda^2 = 1.004$ ) and  $q_{||} \lambda = 0.01$ . The depolarization shift  $\Delta_z$  is 0.045 with  $V_{1212} = 0.31 k_B T \lambda^2$ ,  $E_{12} = 2.88 k_B T$ . Then the resonance frequency given by equation(54) is  $3.01 k_B T$ , almost identical to the result obtained from the loss function. The lower intersubband mode has energy  $\hbar \omega = 1.48 k_B T$  at long wavelength, while  $E_{23} = 1.34 k_B T$ . The depolarization shift  $\Delta_z = 0.051$ , and the corrected energy from equation(54) is 1.41 in units of  $k_B T$ , slightly smaller than the value 1.48 determined from the loss function.

By comparing the dispersion curves and pair spectra in Fig.6, we now discuss the Landau damping of the modes; see also Fig.2. The main 2-D plasmon merges into its pair-excitation continuum when  $q_{||} \lambda = 0.7$ . Beyond this intersection the mode is strongly damped. For  $q_{||} \lambda \geq 0.15$  the upper intersubband resonance enters its continuum and becomes damped as shown in Fig.3; the lower intersubband mode is strongly damped at rather small wavevector, partly due to its proximity to the dominant 2-D plasmon. Finally, notice that the acoustic plasmon mode lies outside the pair spectrum for small wavevectors, until  $q_{||} \lambda = 0.2$  when damping sets in.

We turn now to the dependence of the mode frequencies on  $n_s$ , the parameter which controls the band bending. Figure 7 shows loss functions evaluated with  $q_{||} \lambda = 0.05$ , for successively higher surface charge densities. Increasing  $n_s$  causes the potential to deepen, increasing the subband occupancies and thus the energies of the intrasubband plasmons. At the same time, the  $E_{ij}$  increase, so the intersubband plasmons also move to higher energies. The resulting loss peak positions are displayed in Fig.8. Both intrasubband plasmons

follow the 2-D behavior given in equation(53) closely at high density; note that  $n_s \approx n_1 + n_2$ . For small  $n_s$ , 3-D plasmon behavior emerges because the potential well is shallow and intersubband transitions are dominant; i.e., motion in the z-direction is important. At very low density the energy differences between subbands are so small that bulk behavior results. In Fig.8 we can see a continuous transition between 2-D and 3-D behavior. Mode coupling is not significant at high density because the frequencies are not resonant.

### Comparison with HREELS Data

We have treated the dielectric response of the slab without including the contribution of lattice vibrations; i.e., we assumed the dielectric constant of the background is given by  $\epsilon_-$ . Now InAs, a polar semi-conductor, has an infrared-active transverse optical phonon at long wavelength in bulk whose surface analog, the so-called Fuchs-Kliewer mode<sup>59</sup>, can make an important contribution to inelastic electron scattering. In order to compare our model with the HREELS observations on InAs accumulation layers<sup>15</sup> we now extend our calculations to include lattice vibrations. We describe the dynamical response of the lattice by replacing  $\epsilon_-$  in equation(41) and (50) with a frequency dependent dielectric constant

$$\epsilon(\omega) = \epsilon_{\infty} + \frac{\omega_{TO}^2(\epsilon_0 - \epsilon_{\infty})}{\omega_{TO}^2 - \omega^2} \quad (58)$$

For InAs, the long wavelength transverse optical phonon frequency  $\omega_{TO}$  is  $1.04 k_B T$  (26.9 meV). The parameters  $\epsilon_-$  and  $\epsilon_0$  were given above. In our numerical calculations a phenomenological damping term  $-i\omega\gamma$  is added in the denominator of equation(58), with  $\hbar\gamma = 0.04 k_B T$ .

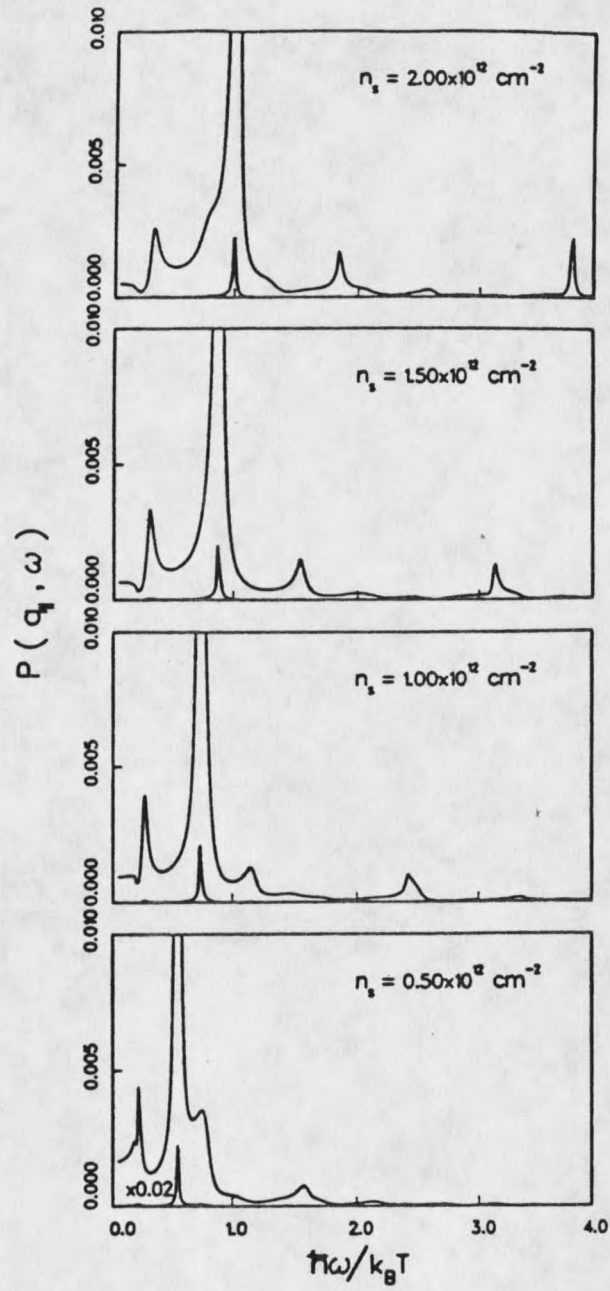


Fig. 7 Loss function evaluated at various surface charge densities  $n_s$  specified in the figure.  $q_{\parallel}\lambda=0.05$ .

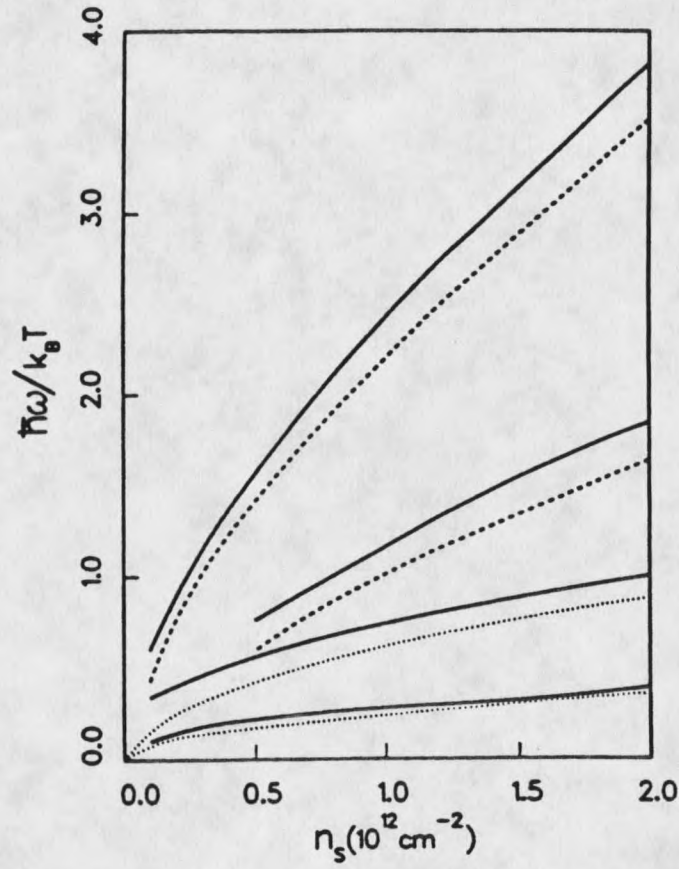


Fig. 8 Plasmon dispersion curves (solid lines) obtained from the maxima of  $P(q_{||}, \omega)$  for intersubband and intrasubband modes, as a function of surface charge density  $n_s$ . The dashed lines give the energy differences  $E_{12}$  and  $E_{23}$  and the dotted lines are the ideal 2-D dispersions.

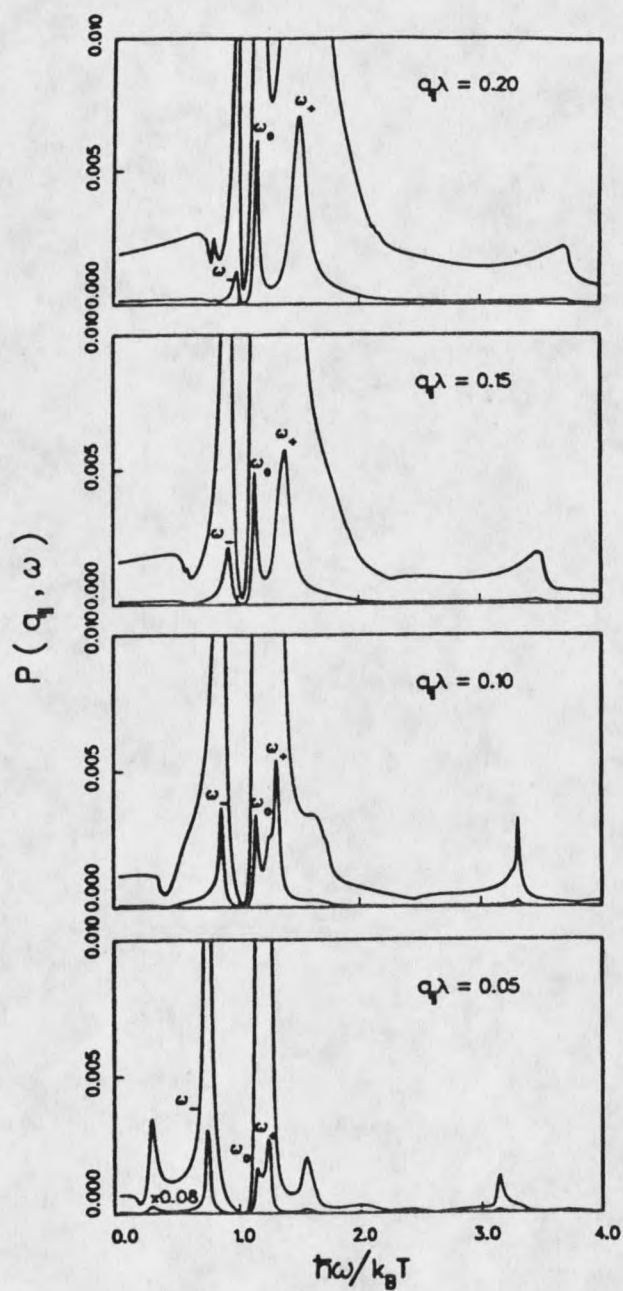


Fig. 9 The loss function including the phonon response.

When lattice vibrations are included in the dielectric response, coupled modes arise due to the interaction of the polarization fields of the phonon and plasmons; these modes have been termed "plasmareons", and may be labeled by their frequencies  $\omega_+$ ,  $\omega_0$  and  $\omega_-$ <sup>8</sup>. In general, when  $q_{||}$  increases from zero the phonon-like mode  $\omega_+$  splits off from the "bare" or unscreened Fuchs-Kliwer mode of frequency  $\omega_s = \omega_{\infty}[(1 + \epsilon_0)/(1 + \epsilon_-)]^{1/2}$ . At small  $q_{||}$ , the  $\omega_+$  and  $\omega_0$  modes are phonon-like while the  $\omega_-$  is plasmon-like. In contrast, at large  $q_{||}$ , the  $\omega_+$  mode is plasmon-like and the  $\omega_-$  mode is phonon-like. We turn now to the dispersion curves obtained in our calculation for InAs.

Figure.9 shows the loss function  $P(q_{||}, \omega)$  for the same parameters given in Fig.3 but with the inclusion of phonon response. Note that modes one, two and four in Fig.3 are almost unchanged because their energies are very different from the phonon energy in this wavevector range. Thus we focus our discussion on the coupling between the phonon and the main 2-D plasmon, which leads to three plasmareon modes labeled  $(\omega_-, \omega_0, \omega_+)$  as in Ref.8. The  $\omega_-$  mode has relatively high intensity and its frequency is well below the bare phonon frequency at small  $q_{||}$ . As  $q_{||}$  increases, the frequency of the  $\omega_-$  mode approaches  $\omega_{TO}$  and its intensity is decreased upon entering the continuum. Unlike the  $\omega_-$  mode, the intensities of the  $\omega_0$  and  $\omega_+$  modes increase with  $q_{||}$ , and the  $\omega_+$  mode is strongly Landau damped only at large  $q_{||}$  values, as indicated in Fig.6. Note that at small  $q_{||}$ , the  $\omega_0$  mode appears as a shoulder near the  $\omega_+$  mode; the separation between these modes is more apparent at large  $q_{||}$  values.

Figure.10 shows the loss function  $P(q_{||}, \omega)$  for  $n_D = 1.3 \times 10^{16} \text{ cm}^{-3}$  and  $q_{||}\lambda = 0.1$  for different surface charge density  $n_s$ . When  $n_s$  is increased, the frequencies of the  $\omega_-$  and  $\omega_+$  modes are shifted upwards and the intersubband plasmon becomes more intense. When the surface charge density has the value  $0.5 \times 10^{12} \text{ cm}^{-2}$ , the  $\omega_-$  and  $\omega_0$  modes are nearly degenerate at the surface phonon frequency. By scanning the maxima of the loss function  $P(q_{||}, \omega)$ , we obtain the dispersion curves plotted in Fig.10; we used the same

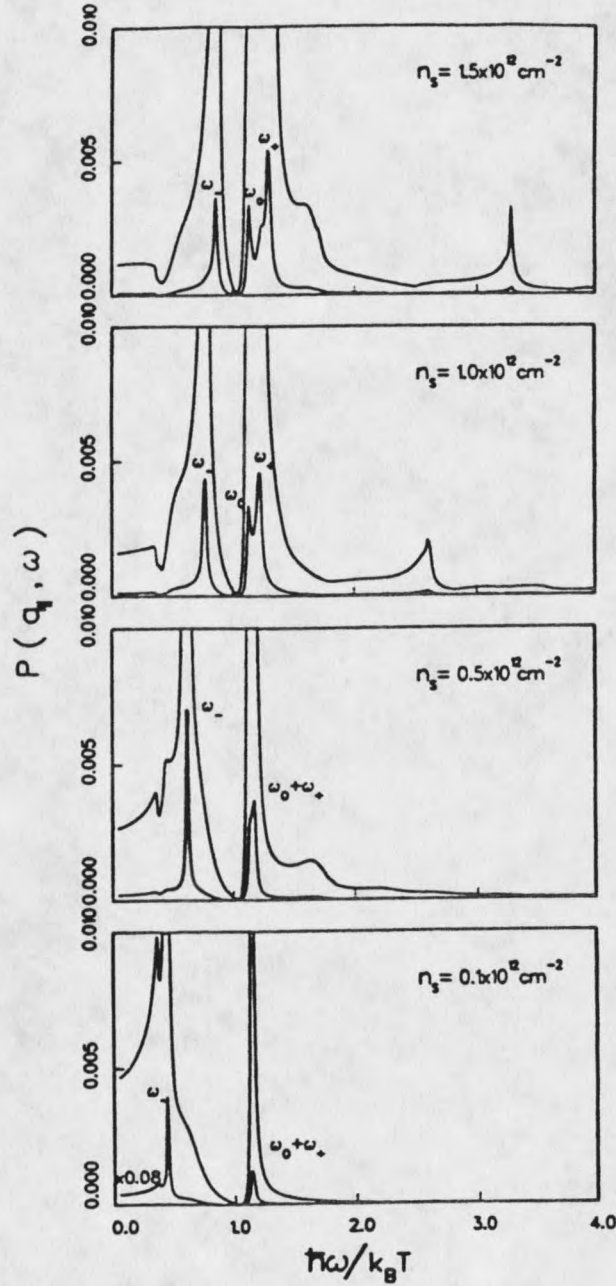


Fig. 10 Calculated loss function  $P(q_{||}, \omega)$  for different surface charge densities  $n_s$ , with  $q_{||}\lambda=0.1$ ,  $\omega_-$ ,  $\omega_0$ , and  $\omega_+$  denote the three plasmaron modes as discussed in the text. The surface charge density is given in units of  $\text{cm}^{-2}$ .

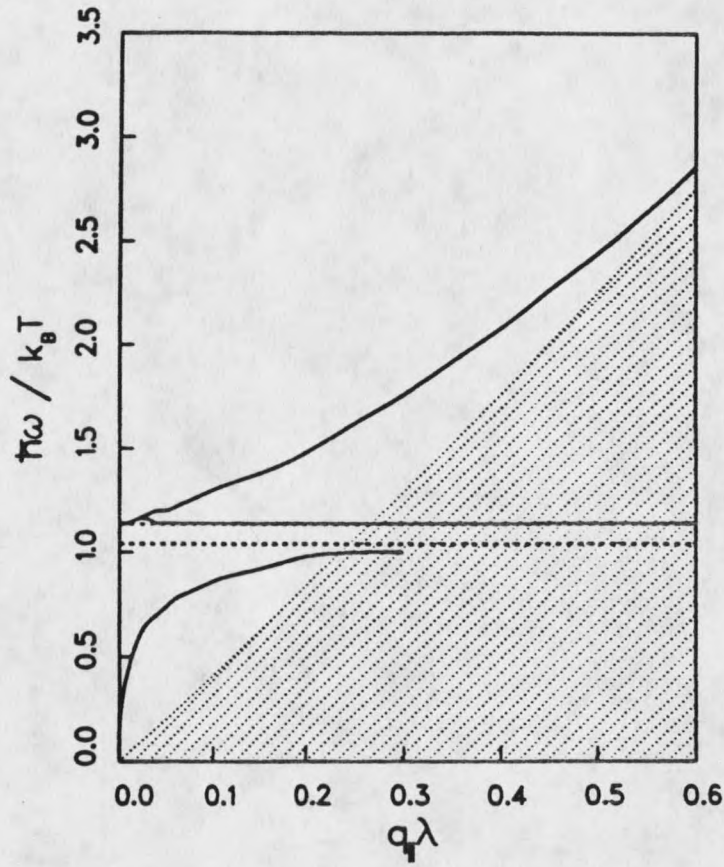


Fig. 11 Plasmaron dispersion curves for  $n_s = 1.5 \times 10^{12} \text{ cm}^{-2}$ . The solid lines give the frequencies of the maxima of the loss function  $P(q_{||}, \omega)$  as a function of the wavevector. The lowest dashed line is the frequency  $\omega_{TO}$  and the upper dashed line is the frequency  $\omega_s$ . The shaded area is the single-particle excitation region. The intersubband and acoustic plasmons are not shown.

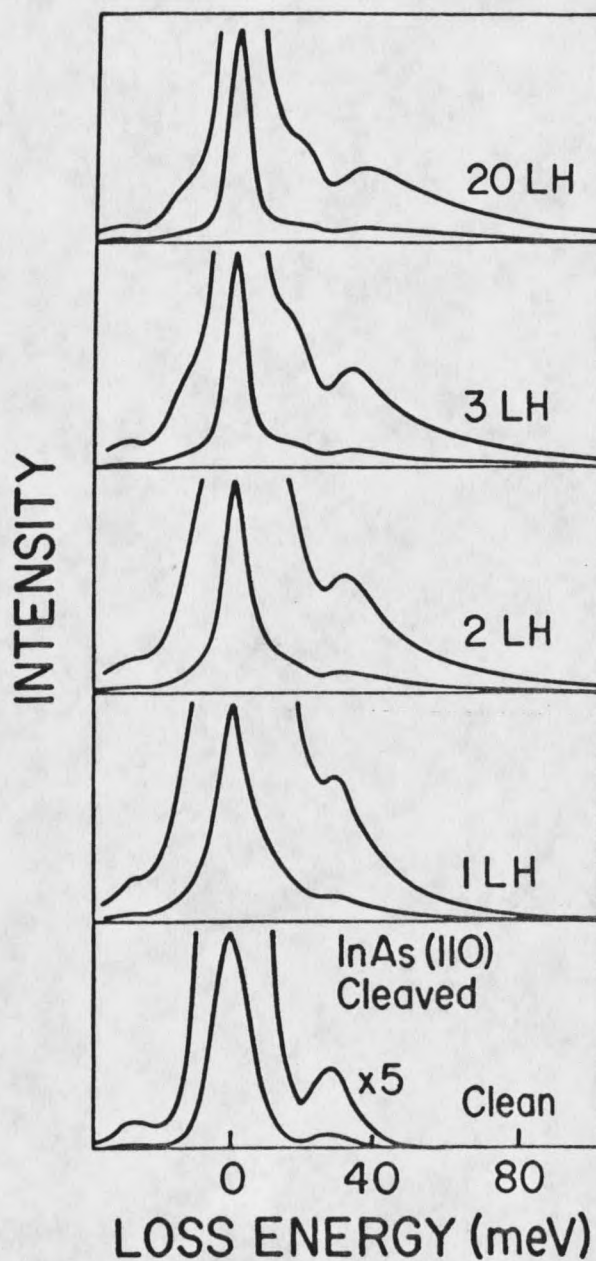


Fig. 12 Experimental HREELS spectra taken from Ref. 15 for different hydrogen exposures on InAs(110). The primary-beam energy is 21 eV and the angle of incidence is  $45^\circ$ . The intensity is given in arbitrary units.

parameters as in Fig.6. The  $\omega_-$  branch varies like  $q_{||}^{1/2}$  at small  $q_{||}$  and approaches the bulk phonon frequency at large  $q_{||}$ . When  $q_{||} \rightarrow 0$ , the  $\omega_0$  and  $\omega_+$  branches are nearly degenerate close to the surface phonon frequency, while for small wavevectors these modes are only weakly coupled. The shaded area in Fig.11 denotes the region of electron-hole pair excitations.

It is instructive to compare the present results with the collective excitations of a 3-D electron gas<sup>60,61</sup>. In the latter, the frequency of the  $\omega_-$  mode approaches a finite value as  $q_{||} \rightarrow 0$ . Also, the limiting frequency of the  $\omega_+$  mode in the 3-D case is higher than  $\omega_s$  while the 2-D  $\omega_+$  mode approaches  $\omega_s$  as  $q_{||} \rightarrow 0$ . Thus, the dispersion relations shown in Fig.11 support our interpretation that we have a 2-D rather than a 3-D electron gas in the first subband.

We now comment briefly on the experiment which was performed recently by our colleagues at Montana State University<sup>15</sup>. Figure.12 shows the HREELS spectra of the InAs(110) surface at different stages of accumulation layer formation achieved by hydrogen exposure. In these experiments, the bulk free carrier density was  $1.3 \times 10^{16} \text{ cm}^{-3}$  and the energy of the incident electron beam was  $E_0 = 21 \text{ eV}$ . The instrumental FWHM was about 9 meV, as judged from the width of the quasielastic peak. Figure.13 shows the spectra obtained after the surface was exposed to 100 L of hydrogen, producing saturation coverage. Hydrogen adsorption induces donor levels which inject charge below the surface, bending the bands downward to create the accumulation layer. The primary-beam energy  $E_0$  was varied in these experiments in order to sample the space-charge layer at different depths; recall that the effective probe depth is proportional to  $E_0^{1/2}$  in HREELS. The behavior of the peaks observed in Fig.12 and 13 may be described as an evolution from one major loss peak to two peaks and an upward shift of their frequencies with either hydrogen exposure or primary energy. In addition, a rather broad loss peak is seen near 50 meV in Fig.13 for low values of  $E_0$ .

In order to interpret the experimental loss peaks, we calculated the scattering efficiency for energy loss  $\hbar\omega$  by integrating  $P(q_{\parallel}, \omega)$  over  $q_{\parallel}$ <sup>26</sup>:

$$P(\omega) = \frac{4m^*e^2}{\omega^2 \cos \theta_I} \int_0^L dq_{\parallel} q_{\parallel}^{-1} M(\xi) P(\omega) \quad (59)$$

where  $\xi = v_0 q_{\parallel} / \omega$  and the factor  $M(\xi)$  is given by

$$\begin{aligned} M(\xi) = & \xi^{-1} [(\xi^2 - 1)^2 + 4\xi^2 \cos^2 \theta_I]^{-3/2} \\ & \times \text{Re}\{(\xi^2 - 1 + 2i\xi \cos \theta_I)^{1/2} [(1 + 2\xi^2 \cos \theta_I + i\xi \cos \theta_I)(1 + \xi^2 \cos^2 \theta_I) \\ & + \xi^2 \sin^2 \theta_I (3\xi^2 \cos^2 \theta_I - 2 - i\xi \cos \theta_I) + \xi^4 \sin^4 \theta_I]\} \end{aligned} \quad (60)$$

The limit of integration in equation(59) was set to a large value because the HREELS detector, with an aperture of a few degrees, collects essentially all inelastically-scattered electrons within the dipole approximation. In equations(59) and (60),  $\theta_I$  is the angle of incidence of the electron beam and  $v_0$  the incident electron velocity.

Typical energy loss spectra calculated by equation(59) are given in Fig.14. Instrumental broadening was not included in these calculations. The angle of incidence of the electron beam is assumed to be  $45^\circ$ . Note that, since a typical wavevector  $q_{\parallel} \approx E_0^{-1/2}$ , the loss peaks are shifted downwards and become narrower as the primary energy  $E_0$  increases. Comparison of Fig.14 with Fig.9, noting again that  $q_{\parallel} \lambda$  is in the range 0.1 to 0.3, leads to the conclusion that the low frequency peak in Fig.14 is due to the  $\omega_-$  plasmon mode while the high frequency peak is due to the  $\omega_+$  mode. The surface phonon mode  $\omega_0$  always appears in the middle and does not disperse, as expected from our discussion above. The small peak at nearly fixed energy  $\hbar\omega \approx 1.7k_B T$  is ascribed to the intersubband plasmon(2 $\rightarrow$ 3); see Fig.6. Finally, the tiny peak at very low energy is due to the acoustic mode.

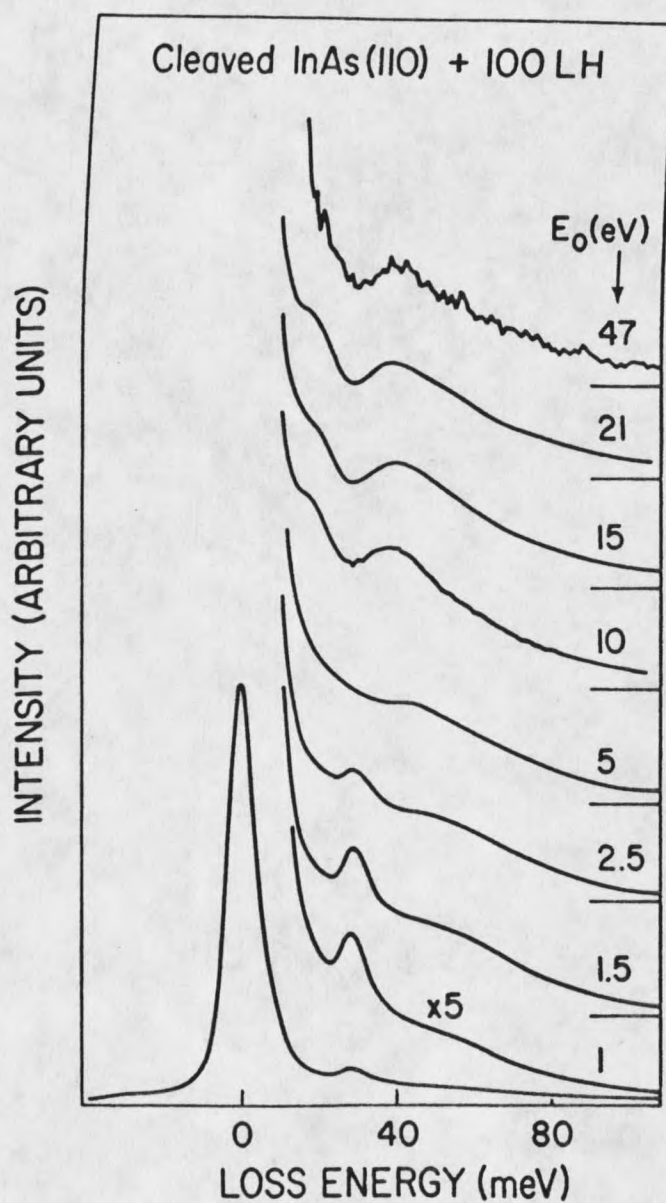


Fig. 13 Experimental HREELS spectra for InAs(110) with 100 L hydrogen exposure and various primary-beam energies as shown in the figure. The angle of incidence is  $45^\circ$ .

Several line-shape changes are evident in Fig.14 as the primary energy is varied. When the  $E_0$  is as low as about 1 eV only the bare phonon mode is observed ( $\omega_0$ ) in the calculated spectrum. As  $E_0$  increases up to 3 eV, two prominent modes ( $\omega_-$  and  $\omega_0$ ) emerge. When  $E_0$  increases further all three plasmon modes become apparent. In general, as  $E_0$  increases, the intensity of the  $\omega_-$  and  $\omega_+$  modes increases while the bare phonon peak becomes weaker. This crossover behavior is associated with the increase of the probe depth, which is essentially  $q_{||}^{-1} \propto E_0^{1/2}$ . To allow a closer comparison with the data shown in Fig.12 and 13, we simulated a 9 meV FWHM instrumental broadening through Lorentzian convolution; the results are displayed in Fig.15 for a range of primary-beam energies. The correspondence of loss features between our calculations and the experimental data of Fig.13 is quite satisfactory, with respect to both the positions and line shapes of the  $\omega_-$  and  $\omega_+$  modes. On the other hand, the "bare" phonon mode at 29 meV persists at higher primary energy in the calculations than in the data. Because this mode is strongest at low primary energy (small probe depth), we infer that it oscillates mainly in the near-surface region where the free-carrier density is low because of the boundary condition on the wavefunction. Since multiple excitations were not considered in our calculations we do not comment on the lineshape of the quasielastic peak.

Finally, some of our calculated HREELS spectra are presented in Fig.16, for different surface charge densities  $n_s$ ; these spectra also include instrumental broadening. It is instructive to compare this set of spectra with the loss functions shown in Fig.10. For weak accumulation layers (small  $n_s$ ) the frequency of the  $\omega_-$  mode is very close to zero at small values of  $q_{||}$ . The  $\omega_-$  mode is shifted upwards with increasing surface charge density  $n_s$  and the mode weakens as  $q_{||}$  increases. After integrating  $P(q_{||}, \omega)$  over  $q_{||}$  and including the instrumental broadening, we find that for small  $n_s$  the  $\omega_-$  mode is buried under the quasielastic peak. If one considers the clean surface ( $n_s=0$ ), the phonon peak is dominant as shown in Fig.16. Increasing the surface charge density deepens the potential well,

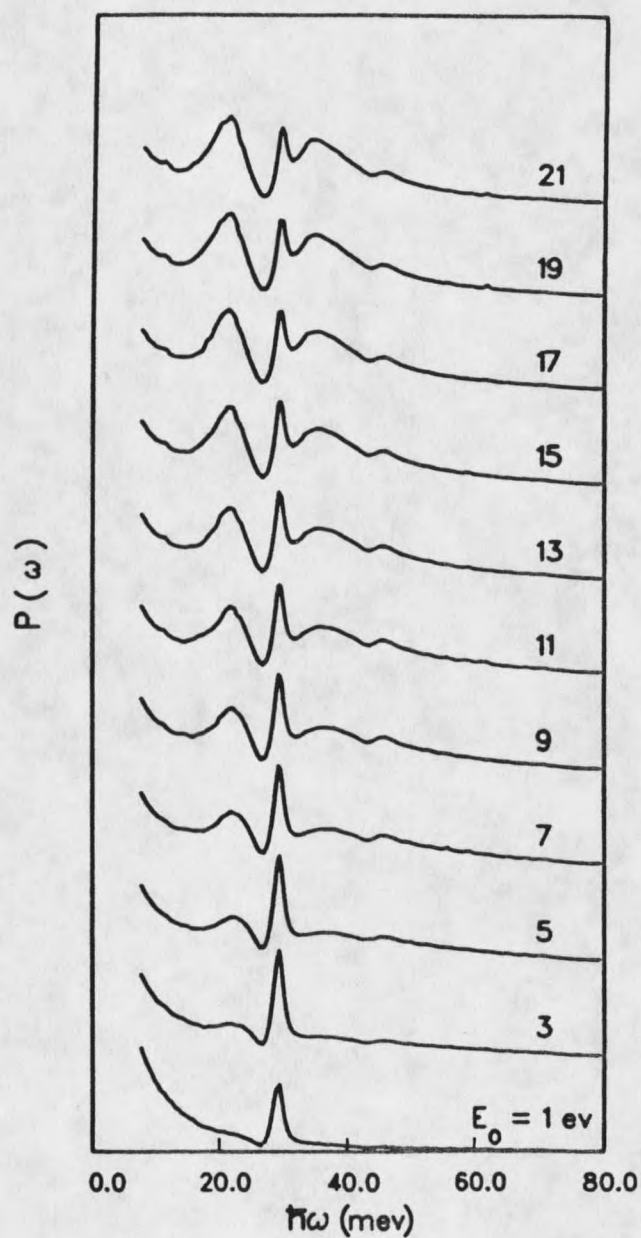


Fig. 14 Calculated HREELS spectra for surface charge density  $n_s = 1.5 \times 10^{12} \text{ cm}^{-2}$  and various primary energies  $E_0$  as shown; without instrumental broadening. The angle of incidence of the electron beam is  $45^\circ$ .  $P(\omega)$  is given in arbitrary units.

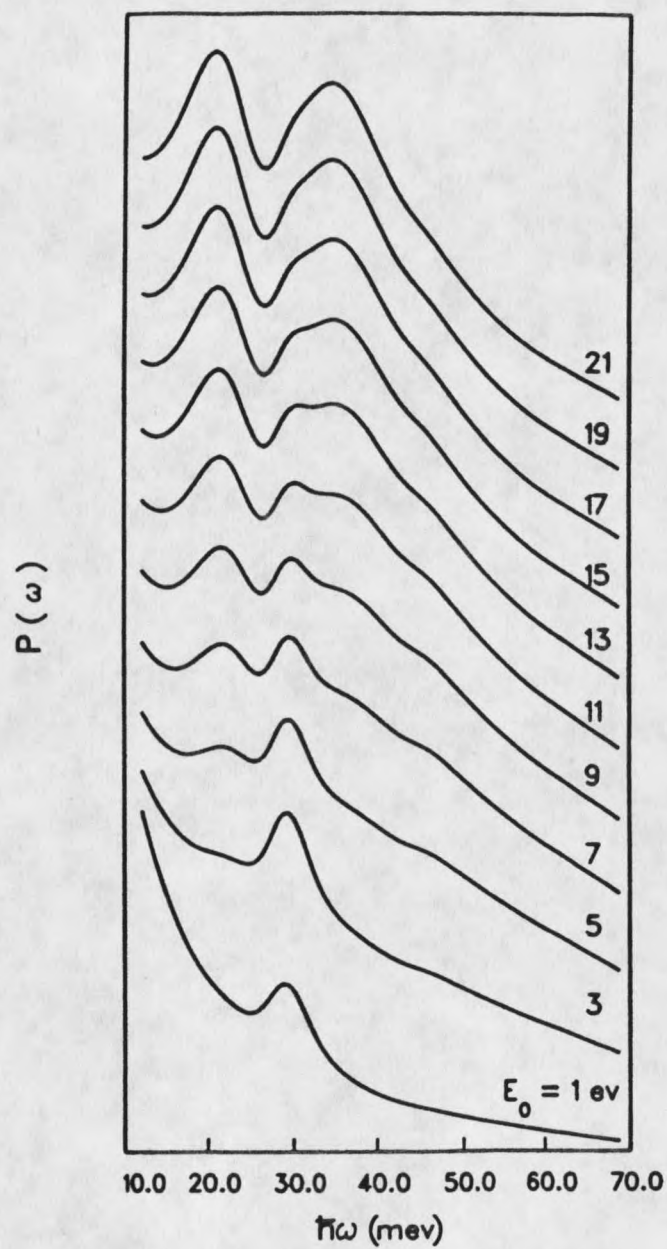


Fig. 15 Calculated HREELS spectra including a simulated 9 meV FWHM instrumental broadening. Other parameters are given in Fig. 14.

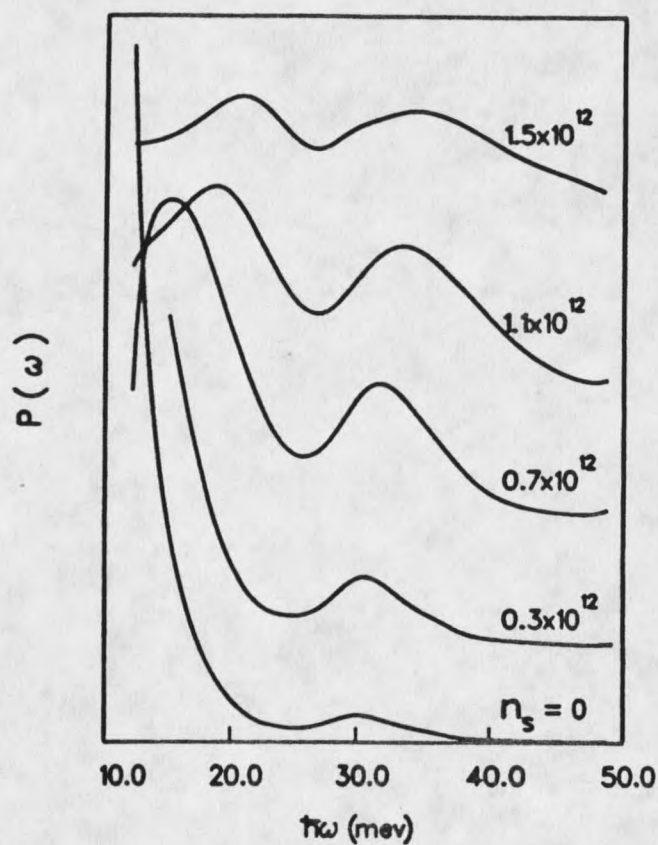


Fig. 16 HREELS spectra calculated for various surface charge densities  $n_s$ , in units of cm $^{-2}$ . Broadening is included as in Fig. 15. The primary energy is 21 eV and the angle of incidence is 45 $^\circ$ .

producing a strong accumulation layer; the intrasubband plasmon oscillating in this layer corresponds to the wing on the left side of the phonon peak. Thus, for large  $n_s$  two peaks corresponding to the  $\omega_-$  and  $\omega_+$  modes should be observed in HREELS. These trends are consistent with the experimental spectra given in Fig.12. Our results confirm the interpretations given in Ref.15, which were based on a simple picture of local dielectric response. The apparent success of the local response model is somewhat surprising given the strength of the inhomogeneity that characterizes the accumulation layer in InAs.

### Accumulation layer on ZnO surface

#### Subband Electronic Structure

The ZnO accumulation layer possesses a strongly localized electron gas with a length scale of about 10 Å. 2-D behavior of such a system would be anticipated because a large majority of the electrons occupy the lowest subband. Because of the small length scale (10 Å) of the extension of the electron gas in the z-direction, the microscopic details, such as electron tunneling effect, position dependent effective mass, and exchange and correlation effect<sup>62</sup>, may affect the electronic subband as well as the dynamics of the electron gas of the accumulation layer of ZnO.

To include the effect of electron tunneling into vacuum<sup>68</sup> we fill the two regions of width  $z_0$  between the slab and the infinite barriers with finite potential barriers of height  $E_b$  equal to the electron affinity; i.e., the energy difference between the vacuum level and the conduction band minimum. We choose  $z_0$  sufficiently large compared with the decay length of the wavefunction that the shape of the charge density and potential are independent of it.

The conduction electrons inside the dielectric slab are treated as a free electron gas characterized by an effective mass  $m^*$  just as we did for the InAs surface. Immediately

outside the slab we place infinitely thin sheets of charge of area density  $n_s$  representing the positively charged layer induced by exposing the surface to atomic hydrogen. We introduce a position-dependent effective mass  $m^*(z)$  varying continuously between  $m^*$  deep inside the slab and  $m_e$ , the free electron mass, in the vacuum. Specifically,  $m^*(z)=m_e$  inside the finite barriers and  $m^*$  in the dielectric slab, except that in a thin layer of width  $a$  immediately outside the slab an exponential interpolation is inserted to connect the inner and outer masses in a continuous manner.

The wavefunctions and energy levels of the electrons satisfy equations(30) and (31). But, given a position-dependent longitudinal mass as defined above, the wavefunction  $\phi_i(z)$  satisfy a one-dimensional Schrodinger equation of the BenDaniel-Duke form:<sup>69</sup>

$$-\frac{\hbar^2}{2} \frac{d}{dz} \left[ \frac{1}{m^*(z)} \frac{d\phi_i(z)}{dz} \right] + v_{\text{eff}}(z)\phi_i(z) = \epsilon_i \phi_i(z) \quad (61)$$

In this equation, the effective potential is given by

$$v_{\text{eff}}(z) = v_B(z) + v_H(z) + v_{xc}(z), \quad (62)$$

where  $v_B(z)$  represents the potential barrier of height <sup>70-72</sup>  $E_b=4.1$  eV,  $v_H(z)$  is the Hartree potential of the conduction electron system(see equation(33)), and  $v_{xc}(z)$  is the exchange-correlation potential. The boundary conditions

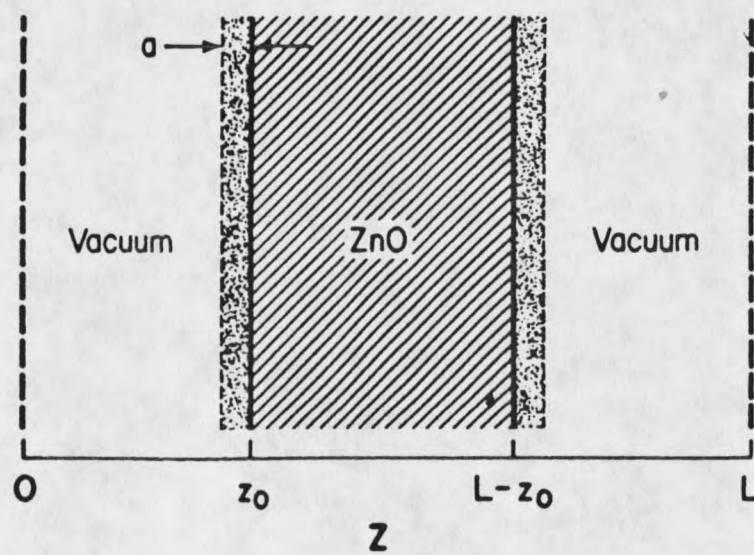


Fig. 17 Slab model of ZnO; see text for explanation.

$$v_H(L/2) = 0$$

$$\frac{dv_H(z)}{dz}\bigg|_{z=z_0} - \frac{dv_H(z)}{dz}\bigg|_{z=L-z_0} = \frac{8\pi e^2 n_s}{\epsilon_\infty} \quad (63)$$

ensure that we measure all energies from the bottom of the bulk conduction band. The free carrier density at depth  $z$  is given at zero temperature by

$$n(z) = \frac{m^*}{\pi \hbar^2} \sum_i \theta(E_F - \epsilon_i)(E_F - \epsilon_i) |\phi_i(z)|^2, \quad (64)$$

with  $\theta$  the unit step function, equal to unity (zero) for positive (negative) values of its argument, and  $E_F$  the Fermi energy, determined by the condition of charge neutrality,

$$n_{\text{tot}} = \frac{m^*}{\pi \hbar^2} \sum_i \theta(E_F - \epsilon_i)(E_F - \epsilon_i) \quad (65)$$

In equation(64),  $n_{\text{tot}}$  is the areal electron density given by

$$n_{\text{tot}} = \int_0^L n(z) dz = n_D(L - 2z_0) + 2n_s \quad (66)$$

In equations(64) and (65), the sum over subband index  $i$  includes occupied states only and a constant effective mass is assumed. The generalization of equation(64) to finite temperature is given by equation(34).

The exchange-correlation potential  $v_{xc}$  was adopted from the local density approximation;<sup>69</sup> We chose the simple analytic form of Hedin and Lundqvist<sup>73</sup>:

$$v_{xc}(z) = -[1 + 0.7734x \ln(1 + x^{-1})] \left( \frac{2}{\pi \alpha r_s} \right) R_y^* \quad (67)$$

with

$$\begin{aligned}\alpha &\equiv \left(\frac{4}{9\pi}\right)^{1/3}, x \equiv x(z) = \frac{r_s}{21} \\ r_s &\equiv r_s(z) = \left[\frac{4}{3}\pi(a^*)^3 n(z)\right]^{-1/3} \\ a^* &= \frac{\epsilon_\infty \hbar^2}{m^* e^2}, R_y^* = \frac{e^2}{2\epsilon_\infty a^*}\end{aligned}\quad (68)$$

As in our earlier work, we use a Fourier representation for the wavefunction. The Schrodinger equation is transformed to a matrix equation of the same form as equation(37) except that for the ZnO case the matrix element is rather lengthy because of the position-dependent effective mass.

In the calculations for ZnO, the parameters to be specified included  $n_s$  and  $n_D$ , the slab thickness  $L-2z_0$ , the number of sine waves  $N_{\text{sine}}$  kept in the expansion of  $\phi_l(z)$ , the width  $z_0$  of the finite potential barrier, the width  $a$  of the interpolation region for the effective mass, and the dielectric constant. The total thickness  $L$  was chosen large enough to simulate a semi-infinite geometry:  $q_{||}\lambda \gg 1$ , where  $q_{||}=0.01 \text{ \AA}^{-1}$  is a typical surface wavevector transfer in HREELS for ZnO. The optimum choice for  $N_{\text{sine}}$  depends on the slab thickness. A typical width of the accumulation layer is of the order of  $10 \text{ \AA}$ ; thus the smallest half wavelength in the sine expansion basis must be of this order, or  $L/N_{\text{sine}} \approx 10$ . After careful examination we chose the values  $L=500 \text{ \AA}$  and  $N_{\text{sine}}=60$  to provide an accurate simulation of semiinfinite geometry.

The charge density profile and self-consistent electron-electron potential ( $v_{\text{eff}}-v_B$ ) are shown in Fig.18 for ZnO with  $n_s=3 \times 10^{13} \text{ cm}^{-2}$  and  $n_D=0$ . The solid lines show our results including exchange and correlation, while the dotted lines were obtained by neglecting these effects. The calculations shown in Fig.18 included the effects of the

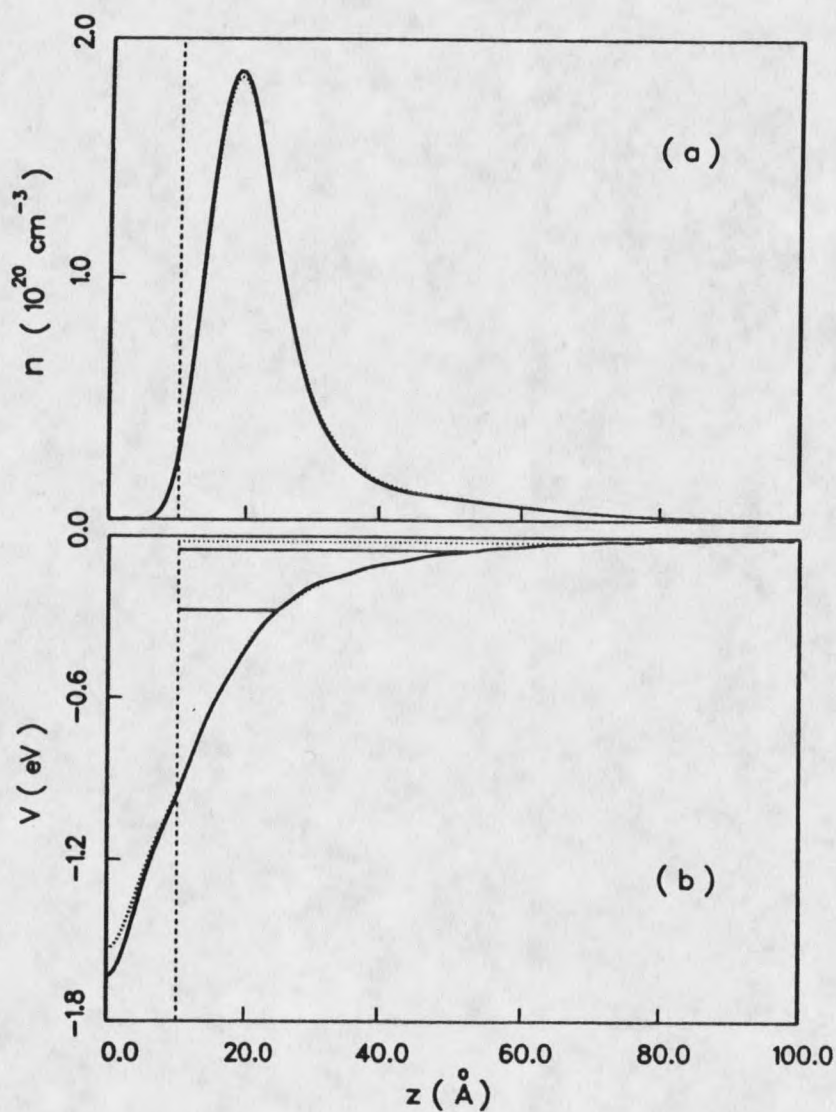


Fig. 18 (a) Self-consistent conduction-electron density and (b) electron-electron potential for ZnO accumulation layer with  $n_s = 3 \times 10^{13} \text{ cm}^{-2}$ ; other parameters are explained in the text. In both panels the results calculated with (without) exchange and correlation effects are given by solid (dotted) lines. The vertical dashed lines indicate the position of the surface plane.

surface barrier but assumed that the effective mass is independent of position:  $L=500\text{\AA}$ ,  $z_0=10\text{\AA}$  and  $a=0$ . The present results are in good agreement with previous results<sup>51,52</sup> on the subband electronic structure of ZnO accumulation layers computed with a different method. We find that more than 90% of the conduction electrons occupy the lowest subband, the remainder occupying mainly the second subband. The charge density is highly localized within  $8\text{\AA}$  of the surface plane because of the strong electric field, as seen in Fig.18. Electrons in the second subband exhibit very little tunneling into the vacuum region; the electron density found outside the surface comes primarily from the first subband. Since the density essentially vanishes within  $3\text{\AA}$  of the surface plane, our choice of surface barrier width, namely  $10\text{\AA}$ , is adequate to ensure that the results do not depend on the boundary condition. Although exchange and correlation affect the charge density only slightly on the scale seen in Fig.18, they do sharpen somewhat the electron density contributed by the first subband. The calculated charge density, including the effect of the position-dependent effective mass, is shown in Fig.17 for two choices of the thickness  $a$  of the interpolation region. For comparison, the density obtained for constant  $m^*$  is indicated by the dashed line; the dotted line gives the result when tunneling is neglected ( $z_0=0$ ). A reasonable value for  $a$  is expected to be about  $2\text{\AA}$ , or roughly the interplanar separation. Fig.19 shows that tunneling shifts the peak of the charge density 3 or  $4\text{\AA}$  closer to the surface while keeping its shape almost unchanged.

Fig.20 shows the dependence of the subband energies and occupation numbers, and of the Fermi level and band bending, on the surface charge density and temperature. To facilitate comparison with the results of Eger *et al.*<sup>51,52</sup> we have shifted the zero of energy to the surface potential, and we have included tunneling and exchange-correlation as described above. Our room temperature results, given in Fig.20(a), are in excellent agreement with the earlier work<sup>51,52</sup>. At room temperature there are generally two

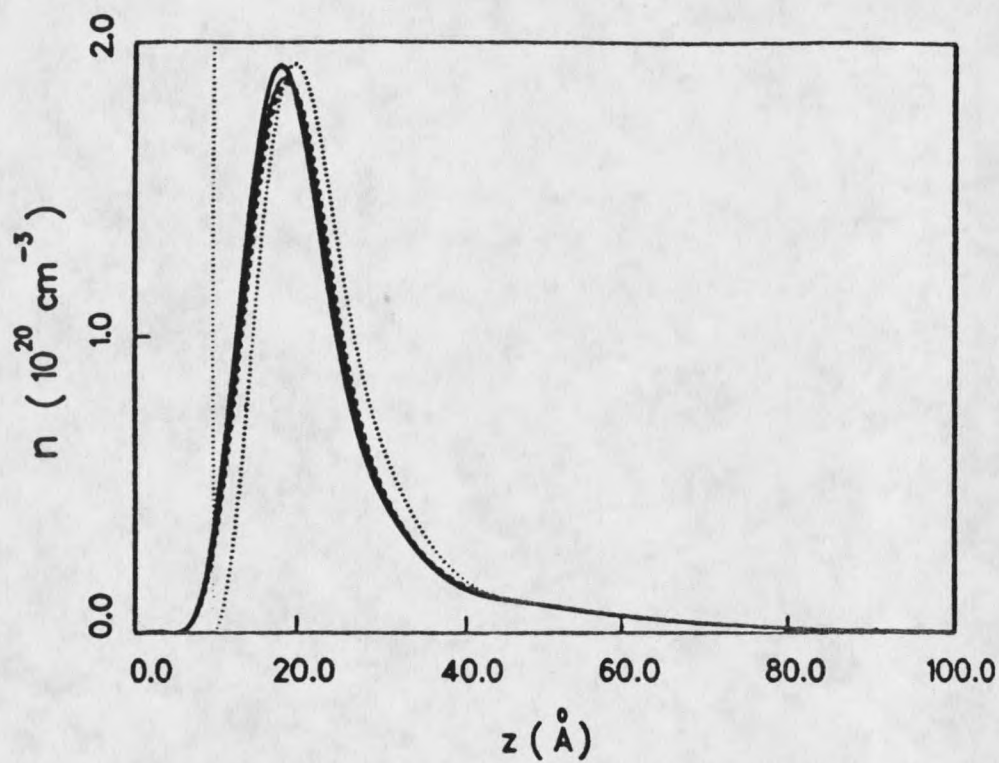


Fig. 19 Conduction-electron density for ZnO. Thick solid line: same as Fig. 18(a). Other results computed for  $a=10\text{\AA}$  (thin solid line), constant  $m^*$  (dashed line), and  $z_0=0$  (dotted line).

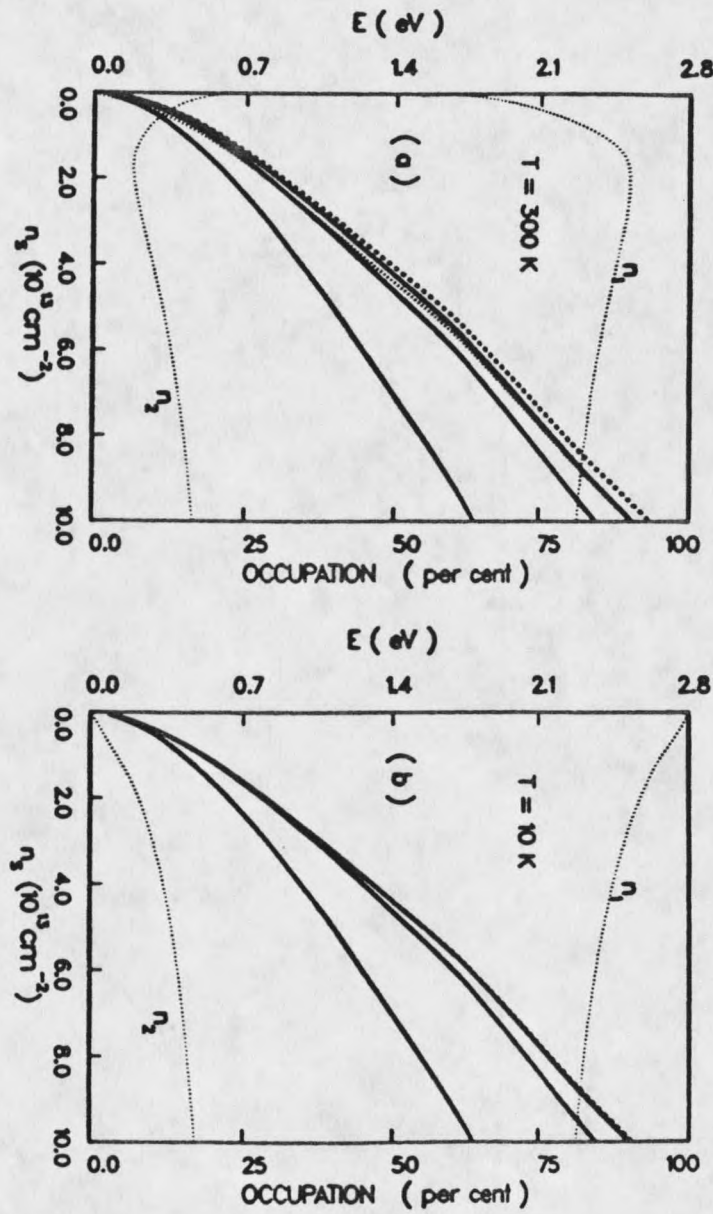


Fig. 20 Subband energy levels (solid lines) and occupation numbers (thin dotted lines) vs surface charge density for (a)  $T=300$  K and (b)  $T=10$  K. Also shown are the Fermi level (dashed lines) and band bending (thin dotted lines). The zero of energy is the surface potential.

subbands below Fermi energy  $E_F$ , and the intersubband separations and band bending increase with  $n_s$  as expected. Note the nearly linear behavior of the curves in Fig.20(a) when  $n_s > 4 \times 10^{13} \text{ cm}^{-2}$ , and the smooth behavior of the occupation numbers there. Below  $n_s = 2.5 \times 10^{13} \text{ cm}^{-2}$ , only the first subband lies below the Fermi level. The dramatic changes of the occupation numbers at small  $n_s$  are associated with the passage from the 2-D regime to the classical limit in which the thermal energy  $k_B T$ , or 26 meV, is larger than the subband separations. To explore the strict 2-D limit we show results in Fig.20(b) obtained at low temperature,  $T = 10 \text{ K}$ . For  $n_s > 3 \times 10^{13} \text{ cm}^{-2}$ , the energies of the lowest subbands agree closely with the room temperature results, although above  $6 \times 10^{13} \text{ cm}^{-2}$  three levels fall below  $E_F$ , instead of two as found at 300 K. Also for low temperature, the occupation numbers vary monotonically with  $n_s$ . Indeed, when  $n_s$  is decreased below  $10^{12} \text{ cm}^{-2}$  the strict 2-D limit is approached: essentially all of the conduction electrons are found in the lowest subband. The low temperature calculations suggest that ZnO is well suited for studies of the 2-D and quasi-2-D electron gas. Finally, we note that because the 10 K and 300 K results are so similar at and above  $3 \times 10^{13} \text{ cm}^{-2}$ , a zero-temperature theory is expected to be adequate there.

### Dynamics of the Electron Gas

Having explored the subband electronic structure of ZnO accumulation layers in the last section, we turn our attention to the dynamical response of the system. Our approach is the same as that used for the InAs accumulation layer. The energy loss function in the dipole approximation is given at zero temperature by

$$P(q_{\parallel}, \omega) = \frac{8e^2}{|1 + \epsilon|^2} \int_0^L dz \int_0^L dz' e^{-q_{\parallel}(z+z')} \text{Im}\{\chi(q_{\parallel}, \omega; z, z')\} \quad (69)$$

The response function  $\chi$  in the above expression is obtained by solving the integral equation (equation(39)). Now, however, the electron-electron interaction  $v_e$  in equation(39) contains the direct Coulomb interaction as well as exchange and correlation. It is worth mentioning that the matrix  $S_{\alpha\alpha'}(q_{||}, \omega)$  can be evaluated in closed form through contour integration<sup>74</sup>. The procedure for solving the integral equation for  $\chi$  is formally identical to that used for InAs except that the matrix elements containing the exchange and correlation potential  $v_{xc}(z)$  have to be evaluated numerically.

Fig.21 shows representative results for the energy loss function when the phonon response is omitted. These results were obtained by solving equation(69) using the parameters assumed in Fig.19, for several values of  $q_{||}$ . A rather weak intrasubband plasmon due to the first subband is seen near  $\omega=0.07$  eV for the smallest wavevector presented,  $q_{||}=0.001\text{\AA}^{-1}$ . Also, a weak peak is seen near  $\omega=0.025$  eV, associated with the interaction of surface plasmon modes on opposite faces of the slab; note that  $q_{||}L=0.5$ . The transitions from the first subband to the second subband ( $\omega\approx 0.25$  eV), and from the first to the empty subband above the Fermi level ( $\omega\approx 0.3$  eV) can be identified by comparing the energy level separation with the peak positions in Fig.21. When  $q_{||}=0.006\text{\AA}^{-1}$ , or  $q_{||}L=3$ , the interaction of plasmon modes from opposite surfaces is weak, producing only a shoulder accompanying the main intrasubband plasmon, and a well defined peak at  $\omega=0.26$  eV due to an intersubband plasmon involving the first and second subbands. Further, a rather broad background due to single-particle(sp) excitations is found near the intersubband peak, and is attributed to transitions from the lowest subband.

The finite slab simulates a semiinfinite material when the wavevector is increased to  $q_{||}=0.01\text{\AA}^{-1}$  or  $q_{||}L=5$ . Then the two intrasubband modes associated with the first band are degenerate, forming only one loss function peak as seen in Fig.21. Since less than 10% of the conduction electrons occupy the second subband, the collective response of this band is anticipated to be weak and to have low energy. This intrasubband peak, strongly screened

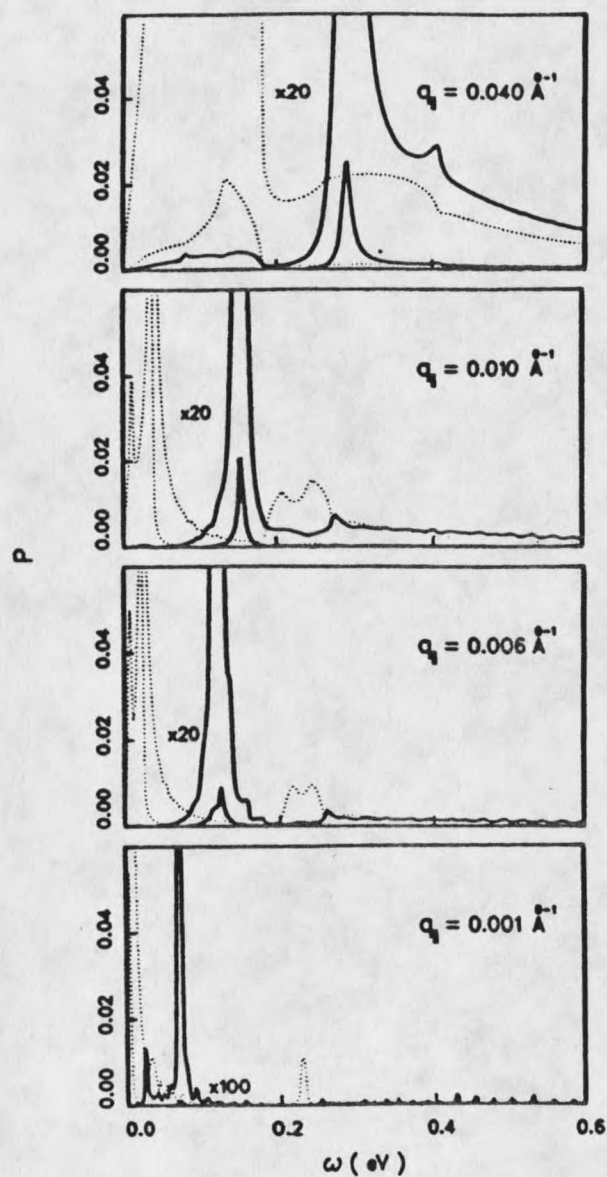


Fig. 21 Energy-loss function  $P(q_{||}, \omega)$  (solid lines) presented as a function of frequency for four surface wavevector  $q_{||}$  in units of  $2m^*e^2/\hbar^2$ . The results obtained when the electron-electron interaction is omitted are shown by the dotted lines.

by the main plasmon<sup>57</sup>, can be seen at  $\omega=0.08\text{eV}$  in Fig.21. The broad feature below  $\omega\approx 0.2\text{eV}$  is attributed to sp excitations; this conclusion is supported by comparing the position of the dip near  $\omega=0.18\text{ eV}$  with the sp edge shown in Fig.22. Note that the main plasmon gains intensity as  $q_{\parallel}$  is increased in Fig.21, and that the mode is damped at large wavevectors  $q_{\parallel}>0.1\text{\AA}^{-1}$  where it enters the sp continuum; into which it decays.

Fig.21 also presents the noninteracting (sp) spectra obtained by omitting the electron-electron interaction in equation(39) (i.e., setting  $v_e=0$ ). By energy-momentum conservation the excitation energy must fall in the ranges  $(0, q_{\parallel}v_{F1})$ ,  $(0, q_{\parallel}v_{F2})$ ,  $(E_{12}-q_{\parallel}v_{F1}, E_{12}+q_{\parallel}v_{F1})$  and peaks should be observed in the spectrum near the edges of these continua;  $v_{F1}$  and  $v_{F2}$  are the Fermi velocities of the first and second subbands. In our dispersion curves given below the two edges  $E_{12}+q_{\parallel}v_{F1}$  and  $q_{\parallel}v_{F1}$  are shown as dotted lines. The sp spectra at  $q_{\parallel}=0.001\text{\AA}^{-1}$  in Fig.21 contain only one peak near the origin arising from the first subband, because of the near degeneracy of  $q_{\parallel}v_{F1}$  and  $q_{\parallel}v_{F2}$ . The intersubband peak is seen near the intersubband energy separation  $E_{12}=0.22\text{ eV}$ . At  $q_{\parallel}=0.06\text{\AA}^{-1}$  there two sp peaks have larger energy separations. Note that, unlike the three dimensional case, 2-D intrasubband continua are peaked near their edges rather than displaying a triangle shape. Experimentally, only one intersubband transition and one intrasubband transition have been observed in Raman spectroscopy<sup>75,76</sup>.

We turn now to the dispersion relations of the plasmon modes portrayed in Fig.21. By scanning the peak positions in these and other figures we mapped out the frequency vs. wavevector plots given in Fig.22. These modes are shown: intrasubband plasmons for the first and second subbands, and the intersubband mode involving transitions between these bands. We assigned the lowest mode to an acoustic plasmon because of its nearly linear dispersion relation<sup>57</sup>; this mode may be described as an intrasubband plasmon in the second subband that is screened by the plasmon in the first subband. The middle mode in Fig.22 (intrasubband plasmon arising from the lowest band) is closely approximated by the

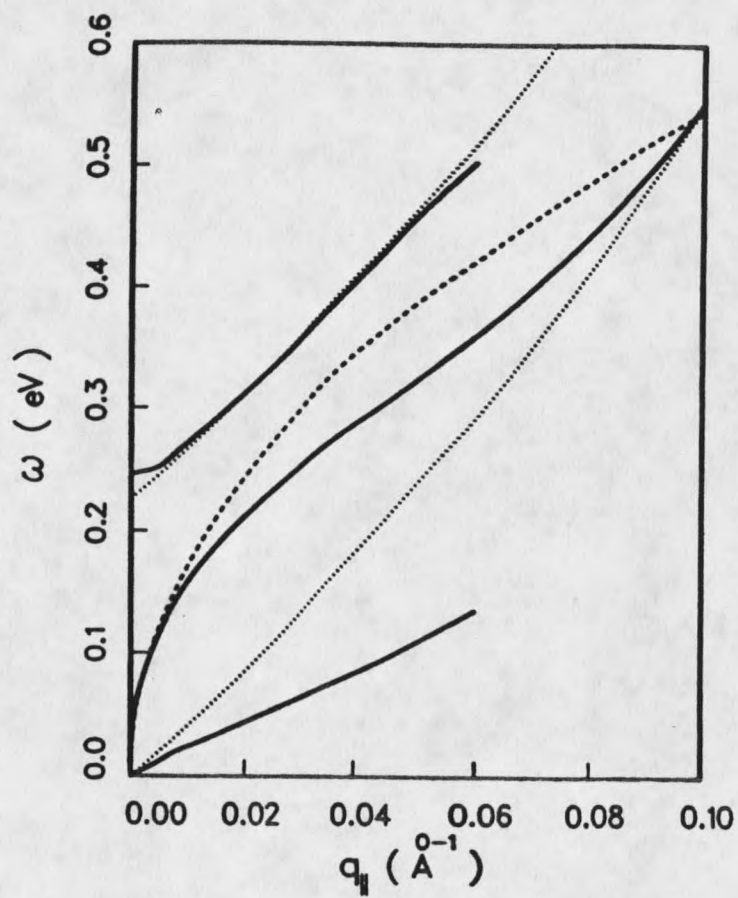


Fig. 22 Frequency-wave-vector dispersion curves (solid lines) for the plasmon peaks shown in Fig. 21. The dotted lines show the edges of the single-particle (sp) continua corresponding to intrasubband and intersubband excitations; ideal 2D dispersion is indicated by the dashed line.

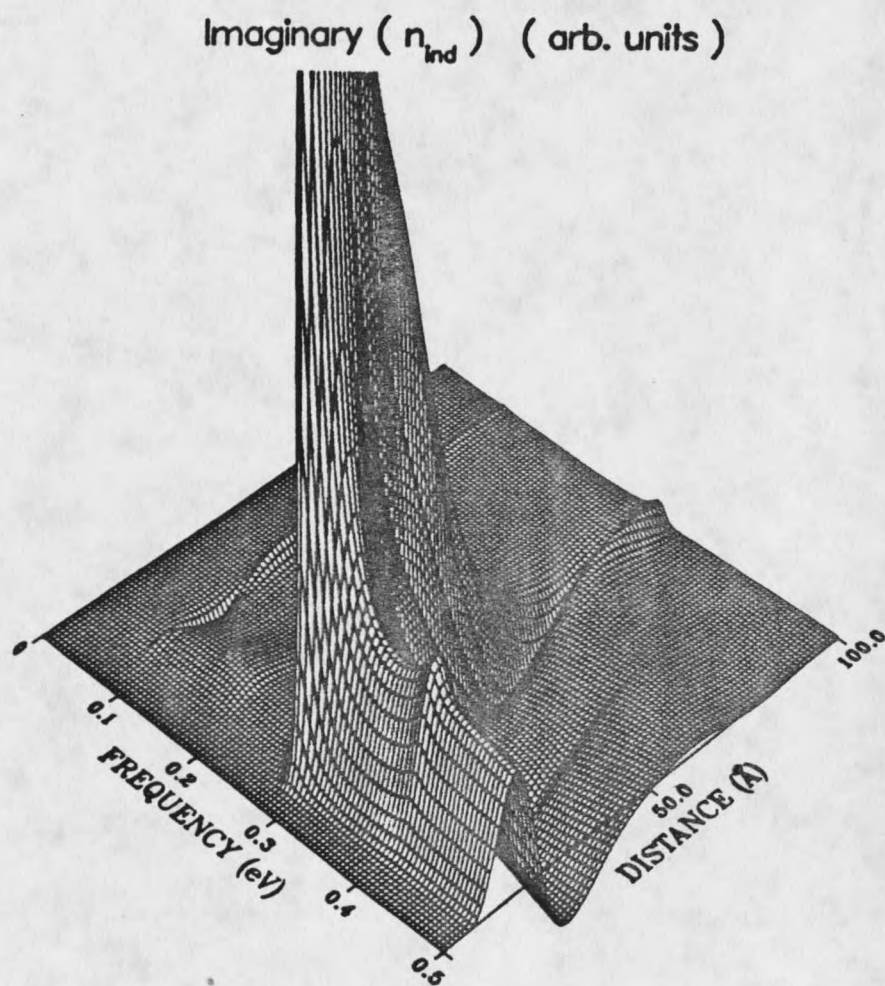


Fig. 23 Imaginary part of induced charge density shown as a function of frequency and distance below the surface for  $q_{\parallel}=0.04 \text{ \AA}^{-1}$ .

2-D frequency  $\omega_{2D}$  at small  $q_{\parallel} < 0.02 \text{ \AA}^{-1}$ , since the associated wavelength is much larger than the width of the density profile. The third mode in Fig.22, an intersubband excitation, has a noticeable coupling with the middle mode even though its amplitude in the loss function is rather small. This coupling deforms the two associated dispersion curves so that the intrasubband mode is depressed below the 2-D results; the intersubband mode follows the sp edge instead of crossing it.

The intersubband plasmon frequency  $\omega_{12}(q_{\parallel} \rightarrow 0)$ , 0.246 eV, is shifted upward from the transition energy  $E_{12}$ , 0.226 eV, because of the Coulomb interaction. This shift, which has been referred as a "depolarization effect"<sup>75,76</sup>, as we discussed above for InAs, can be estimated from the equations(54),(55),and (56). In our calculation, we obtained the values  $v_{1212}=7.01 \text{ eV-\AA}^2$  and  $\Delta_{12}=0.084$ . Thus,  $\omega_{12}(q_{\parallel} \rightarrow 0) = 0.244 \text{ eV}$ , in good agreement with the value 0.246 eV found from the peak of the loss function  $P(q_{\parallel}, \omega)$ .

The induced charge density<sup>54-56</sup> corresponding to  $q_{\parallel}=0.04 \text{ \AA}^{-1}$  is presented in Fig.23 as a function of frequency and depth below the surface. Notice the one-to-one correspondence between the peaks in this figure and the resonances shown in the top panel of Fig.21. As mentioned above, the charge density of the second subband penetrates deeply into the surface; thus the weak induced density near 0.09 eV in Fig.23 supports the conclusion that the acoustic plasmon is a collective excitation of the second subband. The main peak near 0.28 eV is associated with the first subband. Interestingly, the shape of the main peak is similar to that of the charge density shown in Fig.18, though the former lies somewhat deeper inside the surface. Near the frequency of the intersubband plasmon, the induced charge density displays a surface localized peak.

### Plasmon-Phonon Coupling

In order to compare our plasmon calculations with earlier theoretical and experimental<sup>8,13,58</sup> work the lattice vibrations as discussed in our previous calculations for InAs are included in our model. The long wavelength transverse optical phonon frequency  $\omega_{TO}$  is 0.0507 eV and the low- and high- frequency dielectric constants are  $\epsilon_0=8.5$  and  $\epsilon_\infty=4$ . A phenomenological phonon damping term  $-i\omega\gamma$  was added in the denominator of equation(58), with  $\gamma=0.003$  eV. When the phonon response is included in the dielectric function the plasmon and phonon modes may couple to form the so-called "plasmarons" denoted as  $\omega_-$ ,  $\omega_0$ , and  $\omega_+$  modes, as defined above for InAs. The calculated dispersion curves are given in Fig.24. These curves were obtained by monitoring the position of peaks in the energy loss function  $P(q_{||},\omega)$  as a function of wavevector. Though we do not present the individual loss functions we will briefly discuss some aspects of them now. At small wavevector  $q_{||}=0.005\text{\AA}^{-1}$  there are four well-defined peaks at 0.05,0.07,0.14,0.26, which are interpreted as the  $\omega_-$ ,  $\omega_0$ ,  $\omega_+$  and intersubband modes, respectively. As the wavevector is increased, the spectral weight of the  $\omega_-$  mode is decreased, as expected; its frequency approaches the bulk phonon frequency and the mode becomes weak as  $q_{||}$  is increased. An intersubband sp background and an intrasubband sp edge are seen near 0.3 and 0.13 eV, respectively. The  $\omega_0$  surface phonon mode has an almost constant frequency but increases in intensity with increasing wavevector, while both the  $\omega_+$  and the intersubband modes are shifted upwards in frequency.

Fig.24 shows the energy loss function  $P(q_{||},\omega)$  and the corresponding dispersion of the  $\omega_-$ ,  $\omega_0$  and  $\omega_+$  plasmarons but omits the acoustic and intersubband plasmons for greater clarity. Note that the lowest mode ( $\omega_-$ ) is very weak. The sp edges are seen as two dark lines in Fig.24(a). The  $\omega_+$  mode interacts only weakly with the surface phonon mode denoted  $\omega_0$ , largely because of the energy separation of these modes; the dispersion of the former mode is almost unchanged from Fig.22. The  $\omega_-$  plasmaron branch runs almost parallel to the  $\omega_0$  branch at large wavevectors. Our dispersion results are in reasonable

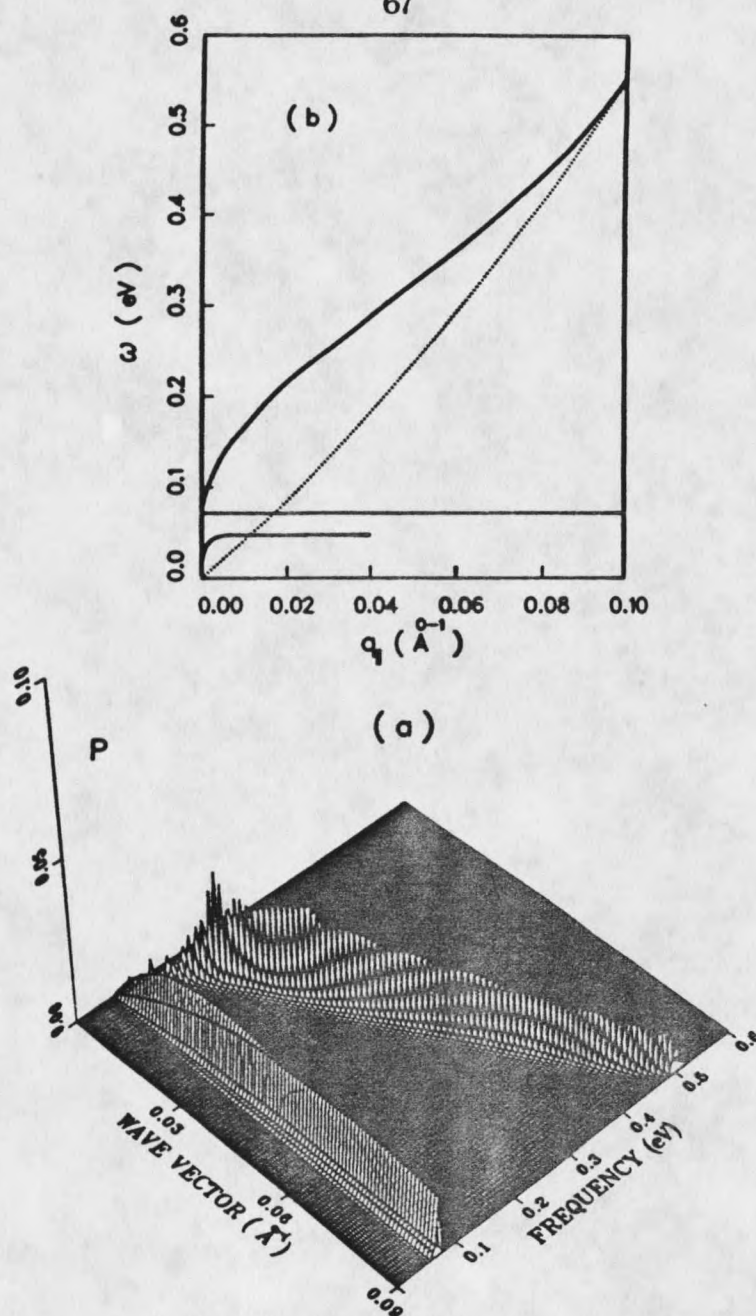


Fig. 24 (a) Energy-loss function  $P(q_{||}, \omega)$  for surface charge density  $n_s = 3 \times 10^{13} \text{ cm}^{-2}$ , including the effects of phonon response. (b) Plasmon dispersion curves obtained from (a); the intrasubband sp edge is shown by the dotted line.

agreement with earlier calculations<sup>8,13</sup> based on the strict 2-D limit. There are some discrepancies at small wavevector, where our  $\omega_+$  mode increases monotonically and the frequency of the  $\omega_0$  mode is nearly constant. Unlike InAs, for the ZnO accumulation layer there is no evidence of phonon character for the  $\omega_+$  mode, and the  $\omega_+$  plasmaron disperses upward, not downward.

The energy loss function and dispersion curves for a smaller surface charge density,  $n_s = 1 \times 10^{12} \text{ cm}^{-2}$  are presented in Fig.25. At this surface charge density only the first subband is occupied. Here the  $\omega_-$  mode is much stronger than in Fig.24, because the intrasubband plasmon has lower frequency and therefore much stronger interaction with the phonon mode. The  $\omega_+$ , on the other hand, is weaker, appearing as a shoulder on the loss peak attributed to the  $\omega_0$  mode. The dark lines represent the sp transitions from the occupied subband to higher subbands. Note that the  $\omega_-$  mode is strongly Landau damped when it enters the sp region.

So far we have discussed the spectrum of collective excitations of the electron gas, including the effects of phonon response by monitoring the peaks in the energy loss function  $P(q_{||}, \omega)$ ; the latter contains all information about the dynamical response of the medium. Now the energy loss spectrum  $P(\omega)$ , obtained from  $P(q_{||}, \omega)$  by integrating over the wavevector, is a quantity that can be directly measured in HREELS. For small wavevectors these two functions are proportional<sup>21</sup>. The surface phonon on ZnO was first observed by Ibach<sup>77</sup>. He found that the relative intensity of the corresponding energy loss peak became weaker with increasing impact energy. Noting that the probe depth is of order  $q_{||}^{-1} \sim E_I^{-1/2}$  where  $E_I$  is the impact energy, our results for the  $\omega_0$  peak intensity are consistent with the observed behavior<sup>77</sup>: the phonon peak decreases in intensity when the wavevector decreases. For large probe depths the modes that extend deeper into the material are preferentially excited (intersubband mode, for example), while for small probe depths

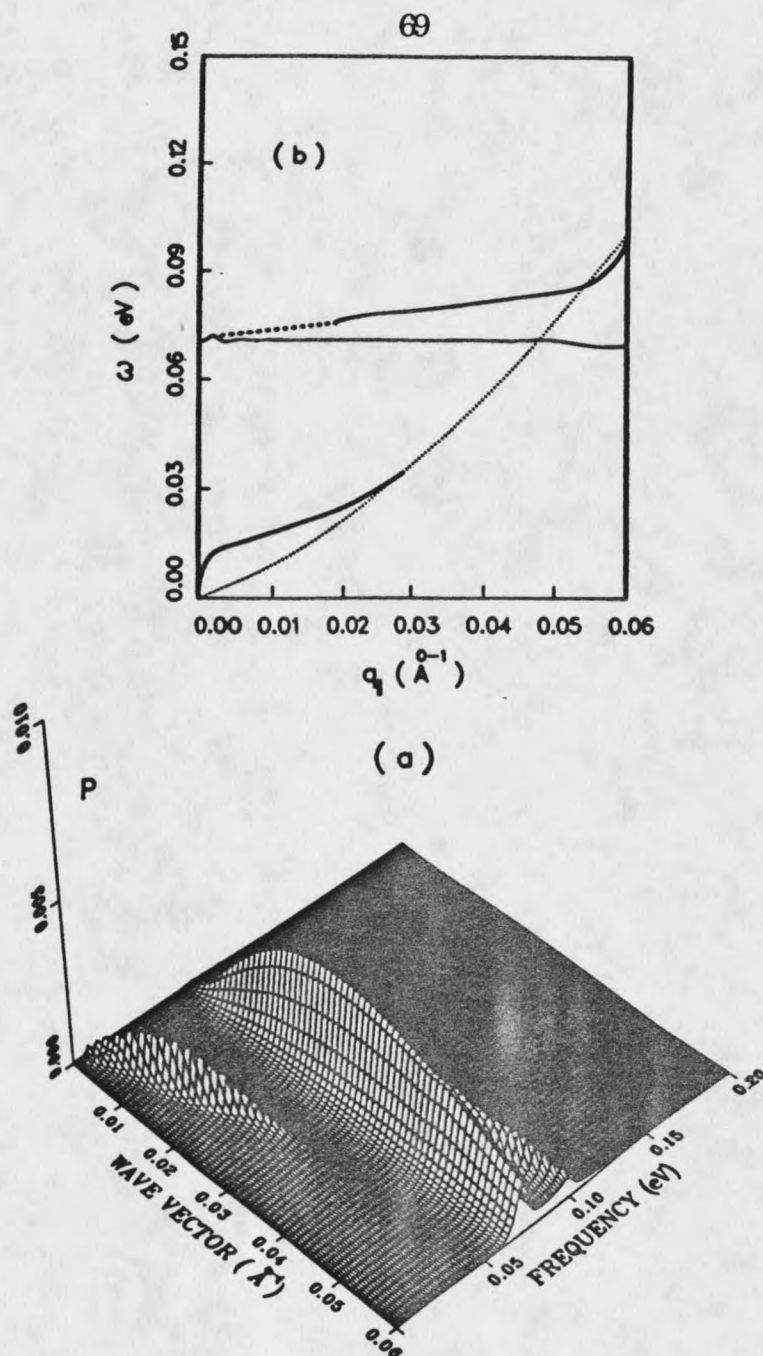


Fig. 25 The same energy-loss function and dispersion curve as shown in Fig. 24 for  $n_s=1 \times 10^{12} \text{ cm}^{-2}$ .

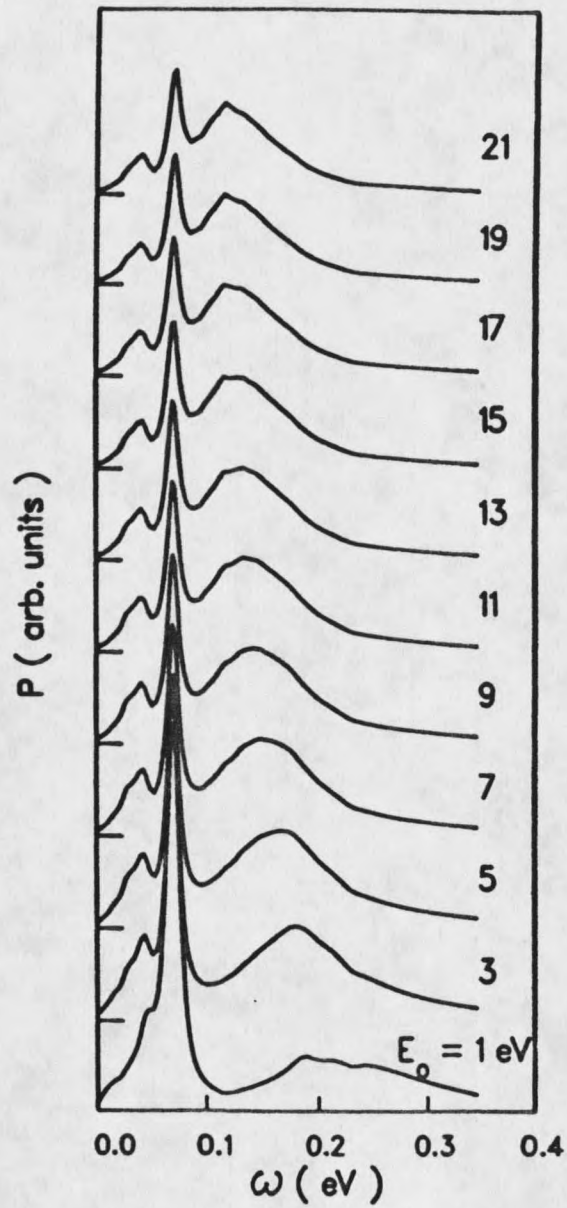


Fig. 26 Energy-loss function spectrum  $P(\omega)$  obtained by integrating  $P(q_{\parallel}, \omega)$  over  $q_{\parallel}$ , for various values of the impact energy  $E_0$ ;  $n_s = 3 \times 10^{13} \text{ cm}^{-2}$ .

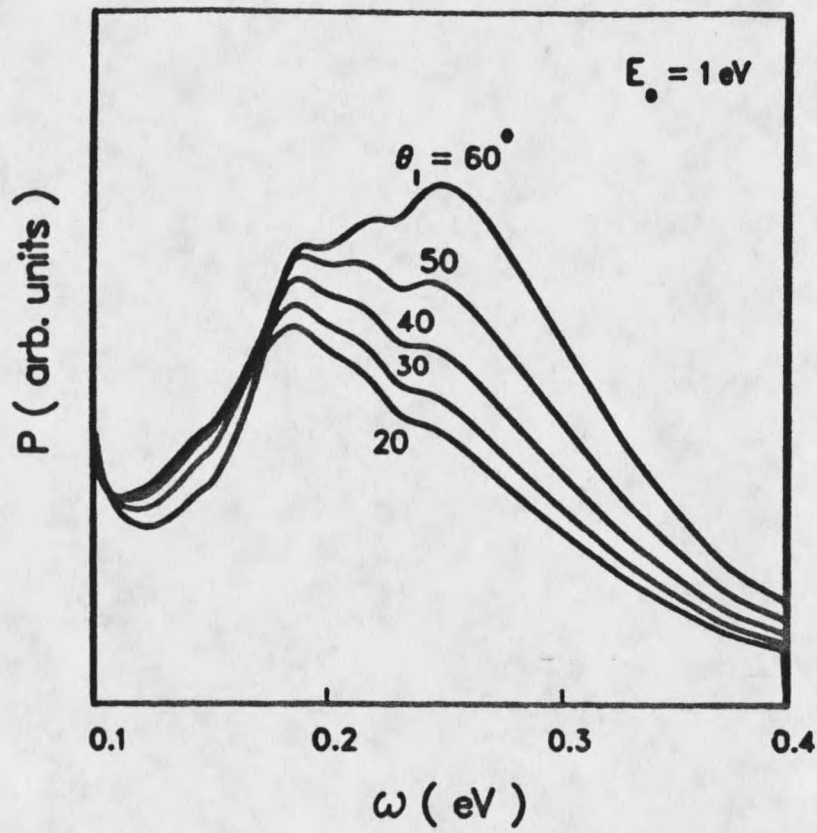


Fig. 27 Energy-loss spectrum at 1 eV impact energy for various angles of incidence  $\theta_i$  of the primary-beam;  $n_s = 3 \times 10^{13} \text{ cm}^{-2}$ .

(large  $q_{||}$ ) the surface-localized modes (surface phonon and main intrasubband plasmon, for example) are dominant.

In order to make quantitative comparison with the data<sup>8,13</sup>, we carried out the wavevector integration for  $P(\omega)$  in the same way as we did for InAs. We did not, however, convolve our spectra to simulate instrumental broadening. The resulting spectra are given in Fig.26 for various impact energies  $E_0$  and an angle of incidence of the primary electrons of  $55^\circ$ . The three plasmaron peaks are well separated as noted earlier. The lowest mode ( $\omega_-$ ) is sharp and has little dispersion; it evolves into a bulk phonon at large wavevector (small  $E_0$ ). The spectral weight of the surface phonon mode ( $\omega_0$ ) increases as the HREELS probe depth decreases. Further, the effects of the steep dispersion of the highest mode ( $\omega_+$ ) can be seen clearly in Fig.26; as the impact energy decreases, the peak maximum moves to higher energy and the peak becomes broader. The sp excitations (intrasubband as well as intersubband) provide a noticeable background in the region of the  $\omega_-$  and  $\omega_+$  peaks. Fig. 26 also contains evidence for an intersubband plasmon. Because of its surface localization this mode is only seen at low impact energies. When  $E_0$  is reduced to 3 eV a shoulder is visible on the high-frequency side of the  $\omega_+$  peak; the shoulder becomes a weak peak at a frequency of about 0.25 eV when the impact energy is 1 eV. The identification of this feature as an intersubband plasmon is suggested by noting that the intersubband transition energy is 0.24 eV. This intensity of the intersubband peak can be enhanced by increasing the angle of incidence (i.e., decreasing the probe depth), as shown by our calculations presented in Fig.27. The peak gains significant strength as the angle of incidence increases from  $20^\circ$  to  $60^\circ$  at 1 eV impact energy.

Gersten *et al.* performed HREELS experiments on the oxygen face of ZnO in the presence of adsorbed hydrogen which produces a strong accumulation layer. They observed the  $\omega_0$  and  $\omega_+$  modes only. On comparing their HREELS data, with our calculated spectra given in Fig.26, we note a similar behavior of the two peaks common to

both figures. The frequency of our  $\omega_+$  mode is slightly lower than that of the experimental peak, suggesting that the surface charge we assumed,  $n_s = 3 \times 10^{13} \text{ cm}^{-2}$ , is lower than the actual charge present in the experiment. Only the value of  $\mu n_s$ , where  $\mu$  is the surface mobility, was quoted in the experiment<sup>13</sup>.

## CHAPTER 4

## ELECTRONIC SUBBAND STRUCTURE AND DYNAMICS OF QUASI-ONE-DIMENSIONAL ELECTRON SYSTEM

Introduction

One-dimensional electronic systems have attracted considerable interest recently, following advances in high-resolution lithography which have made it possible to fabricate semiconductor structures that confine the motion of free electrons in two directions. This confinement gives rise to quasi-one-dimensional electronic properties. The electronic subbands occurring in quasi-one-dimensional structures have been studied experimentally with a variety of techniques<sup>78-83</sup>. Theoretically, few attempts have been made to study the subband electronic properties of the quasi-one-dimensional electron gas (Q1DEG) or "quantum wire". Lai and Das Sarma<sup>84</sup> proposed a variational wave function for the ground state of a Q1DEG in narrow inversion layers in metal-oxide-semiconductor field-effect-transistor (MOSFET) structures. Confinement potentials and charge densities for a Q1DEG under a narrow gate in a metal-oxide-semiconductor structure were calculated self-consistently by Laux and Stern<sup>85</sup>. Here we report, for the first time, complete self-consistent calculations of the subband electronic structure of semiconductor quantum wires, including the effects of exchange and correlation as well as quantum-mechanical tunneling into the region of the confinement potential. These results are part of an ongoing effort directed towards understanding both the subband quantum structure and the dynamical properties of Q1DEG's. The subband structure is studied by self-consistently solving the Schrodinger and Poisson equations in a rectangular wave-guide geometry. To effect the

solution the electron wavefunction is expanded in a Fourier sine series in both confinement directions. The advantages of such an expansion are that (1) the Hamiltonian matrix elements can be written analytically and (2) with the expansion coefficients in hand the study of dynamical response including plasmon excitations in the Q1DEG is greatly simplified<sup>16,17</sup>. Our sample parameters are chosen to match the heterostructure fabricated by molecular beam epitaxy and ion milling as described in Ref.81.

### Model and Self-consistent Formalism for Q1DEG

In this section we describe the physical model of a quasi-one-dimensional electronic system such as found in laterally patterned quantum wells<sup>81</sup> and derive the equations appropriate to our model. We consider an infinitely long rectangular semiconducting "wire" with a jellium-like positively charged background, and with dielectric constant  $\epsilon_{\infty}$  and electronic effective mass  $m^*$ . The coordinate system is illustrated in Fig.28, together with a cross-sectional view of the sample. The rectangular wire lying along the x-axis is enclosed by infinite potential barriers on four planes parallel to the wire's surfaces, for computational convenience. Tunneling effects are included by assuming finite potential barriers just outside the wire, as indicated by the shaded regions in Fig.28. The barrier width  $y_0$  and  $z_0$  are chosen to be large compared with the decay length of the wavefunction, so that the final results do not depend on these widths. To induce the confinement in the z-direction normal to the sample, we place positive surface charge  $n_{sy}$  on two opposite surfaces as shown in Fig.28. The latter surfaces simulate the appropriate GaAs/AlGaAs interfaces. Finally, the Q1DEG is formed by placing negative surface charge  $n_{sz}$  on two surfaces perpendicular to the latter interfaces; these charges produce depletion layers that confine the electrons in the

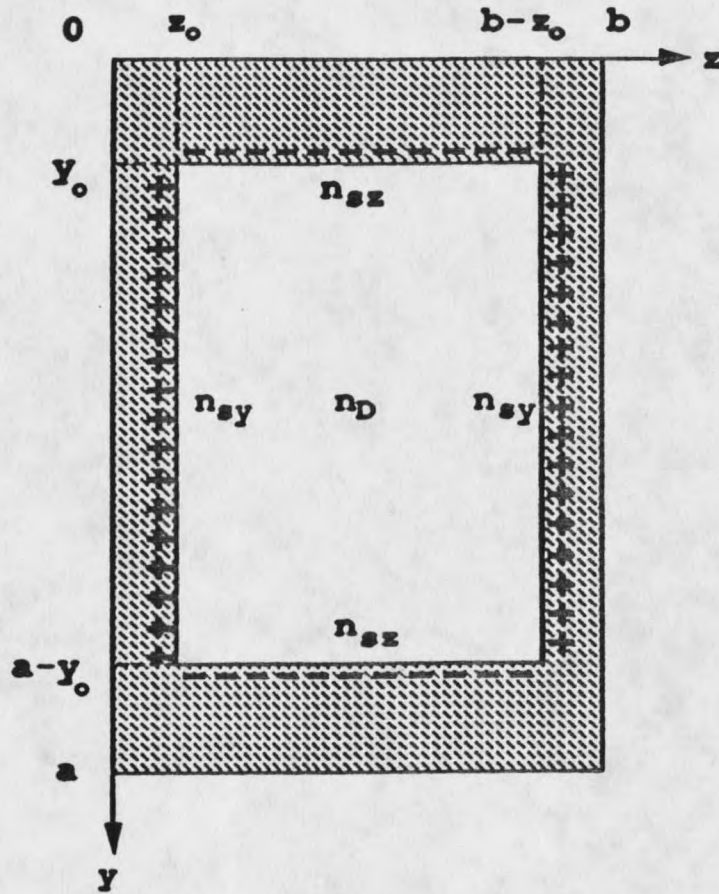


Fig. 28 Physical model considered in our calculations. A finite, constant potential barrier with height  $E_B$  is presented in the shaded region. The bulk free carrier concentration  $n_D$  is chosen to be small enough to have a negligible effect on the electronic structure.

y-direction but allow free current flow along the x-direction perpendicular to the plane of Fig.28.

Because of the translational invariance along the x-direction, the wavefunction of a conduction electron may be written as

$$\phi_{k,ij}(x,y,z) = \frac{1}{L^{1/2}} e^{ikx} \phi_{ij}(y,z), \quad (70)$$

where  $k$  and  $x$  are wavevector and position in the x-direction,  $L$  is the length of the wire and  $i$  and  $j$  are quantum numbers for motion along the confinement directions; we use the Born-von Karman periodic boundary condition for the wavefunction in the x-direction. Then the energy eigenvalue of an electron in the subband labelled by  $ij$  is given by

$$\epsilon_{ij}(k) = \frac{\hbar^2 k^2}{2m^*} + \epsilon_{ij} \quad (71)$$

Here the energy of the bottom of the subband is  $\epsilon_{ij}$ . The latter is an eigenvalue of the two-dimensional Schrodinger equation

$$-\frac{\hbar^2}{2m^*} \left[ \frac{\partial^2}{\partial y^2} + \frac{\partial^2}{\partial z^2} \right] \phi_{ij}(y,z) + v_{\text{eff}}(y,z) \phi_{ij}(y,z) = \epsilon_{ij} \phi_{ij}(y,z), \quad (72)$$

in which the effective potential is given by

$$v_{\text{eff}}(y,z) = v_B(y,z) + v_H(y,z) + v_{xc}(y,z). \quad (73)$$

The three terms on the right in equation(73) are, respectively, the finite potential barrier, the Hartree potential and the exchange-correlation potential. The potential barrier  $v_B$  is equal to the barrier height  $E_B$  in the shaded region in Fig.28, and vanishes elsewhere. The Hartree potential  $v_H$  is determined by the two-dimensional Poisson equation

$$\left[ \frac{\partial^2}{\partial y^2} + \frac{\partial^2}{\partial z^2} \right] v_H(y, z) = - \frac{4\pi e^2}{\epsilon_\infty} [n(y, z) - n_D(y, z)] \quad (74)$$

where  $n(y, z)$  and  $n_D(y, z)$  are the self-consistent charge density and the charge density of the jellium-like background of positive ions, respectively.  $n_D(y, z)$  is assumed to be the bulk carrier concentration  $n_D$  inside the rectangular "wire",  $n_{sz}$  and  $n_{sy}$  on the confinement surfaces, and zero elsewhere (see Fig.28). At finite temperature, the self-consistent charge density is obtained from the two-dimensional eigenfunctions  $\phi_{ij}(y, z)$  as

$$n(y, z) = \frac{2}{\pi} \sum_{ij} \phi_{ij}^2(y, z) \int_0^\infty dk \frac{1}{1 + \exp[\beta(\hbar^2 k^2 / 2m^* + \epsilon_{ij} - \mu)]} \quad (75)$$

where  $\beta = 1/k_B T$ ,  $\mu$  is the Fermi level, and the summation includes all subbands with significant occupation at finite temperature. The Fermi level is determined numerically from equation(75) by the condition of overall charge neutrality, taking into account the external surface charges as well as the charge of the free carriers and the positive ions. The exchange-correlation potential was approximated by an analytic form given by Hedin and Lundqvist which is the same as given in equations(67), and (68), within the local density approximation at zero temperature. Our boundary conditions for the wavefunction and Hartree potential are that both vanish on the planes  $y=0, a$  and  $z=0, b$  where the auxiliary infinite barriers begin (see Fig.28).

We solve equation(74) for the Hartree potential by first finding a Green's function  $G(y,y';z,z')$ , which satisfies the equation

$$\left[\frac{\partial^2}{\partial y^2} + \frac{\partial^2}{\partial z^2}\right]G(y,y';z,z') = -\delta(y-y')\delta(z-z') \quad (76)$$

with the boundary condition that  $G(y,y';z,z')$  vanishes at the edge of the wave guide (see Fig.28). By a standard method obtaining Green's functions, we get

$$G(y,y';z,z') = \frac{2}{\pi} \sum_{m=1}^{\infty} \frac{\sin(\frac{m\pi y}{a}) \sin(\frac{m\pi y'}{a})}{m \sinh(\frac{m\pi b}{a})} \times \begin{cases} \sinh[\frac{m\pi(b-z')}{a}] \sinh(\frac{m\pi z}{a}), & z < z' \\ \sinh(\frac{m\pi z'}{a}) \sinh[\frac{m\pi(b-z)}{a}], & z > z' \end{cases} \quad (77)$$

Then the Hartree potential is given by

$$v_H(y,z) = \frac{4\pi e^2}{\epsilon} \int_0^a dy' \int_0^b dz' G(y,y';z,z') [n(y,z) - n_D(y,z)] \quad (78)$$

On account of the boundary conditions on the wavefunctions, and the limited range of variation of the coordinates  $y$  and  $z$ , we expanded the wavefunction  $\phi_{ij}(y,z)$  in a Fourier sine series in both  $y$ - and  $z$ -directions as

$$\phi_{ij}(y,z) = \frac{2}{(ab)^{1/2}} \sum_{mn} b_{mn}^{ij} \sin(\frac{m\pi y}{a}) \sin(\frac{n\pi z}{b}) \quad (79)$$

The Schrodinger equation can then be transformed into a matrix form as

$$\sum_{m'n'=1}^{\infty} \left\{ \frac{\hbar^2}{2m^*} \left[ \left( \frac{m\pi}{a} \right)^2 + \left( \frac{n\pi}{b} \right)^2 \right] \delta_{m,m'} \delta_{n,n'} + v_{m'n',mn} \right\} b_{m'n'}^{ij} = \epsilon_{ij} b_{mn}^{ij} \quad (80)$$

The matrix elements can be written in closed form.

Next, we discuss a few details of the numerical calculations. To achieve an accuracy of 0.001 meV for the energy eigenvalues it is necessary to include at least 10 terms in the sine wave expansion of the Green's function (equation(77)). The number of sine waves kept in the expansion of the wavefunction itself is chosen large enough to satisfy two conditions: (1) the energy eigenvalues must be converged and (2) there must not be significant oscillations in the potential and charge density. These criteria can be satisfied with 10 to 20 sine waves in each direction, y and z; as a result the hamiltonian matrix has dimensionality 100 to 400. A simple convergence factor technique is used to achieve self-consistent between the potential and charge density, as in our earlier work for InAs and ZnO. The criterion for convergence is that the Fermi levels of consecutive iterations must differ by no more than 0.0001 meV.

### Results and Discussion of Electronic Subband Properties

In the physical model described above, some of the sample parameters were fixed throughout our numerical work. The length parameters a and b (see Fig.28) were chosen as 1200 Å and 700 Å, respectively, to match the experimental situation<sup>81,82</sup>; also,  $m^*=0.069 m_e$  and  $\epsilon_{\infty}=12.9$  appropriate to bulk GaAs. The quantum numbers i and j are, by definition, the number of nodes for the wavefunction in the y and z directions, respectively. In

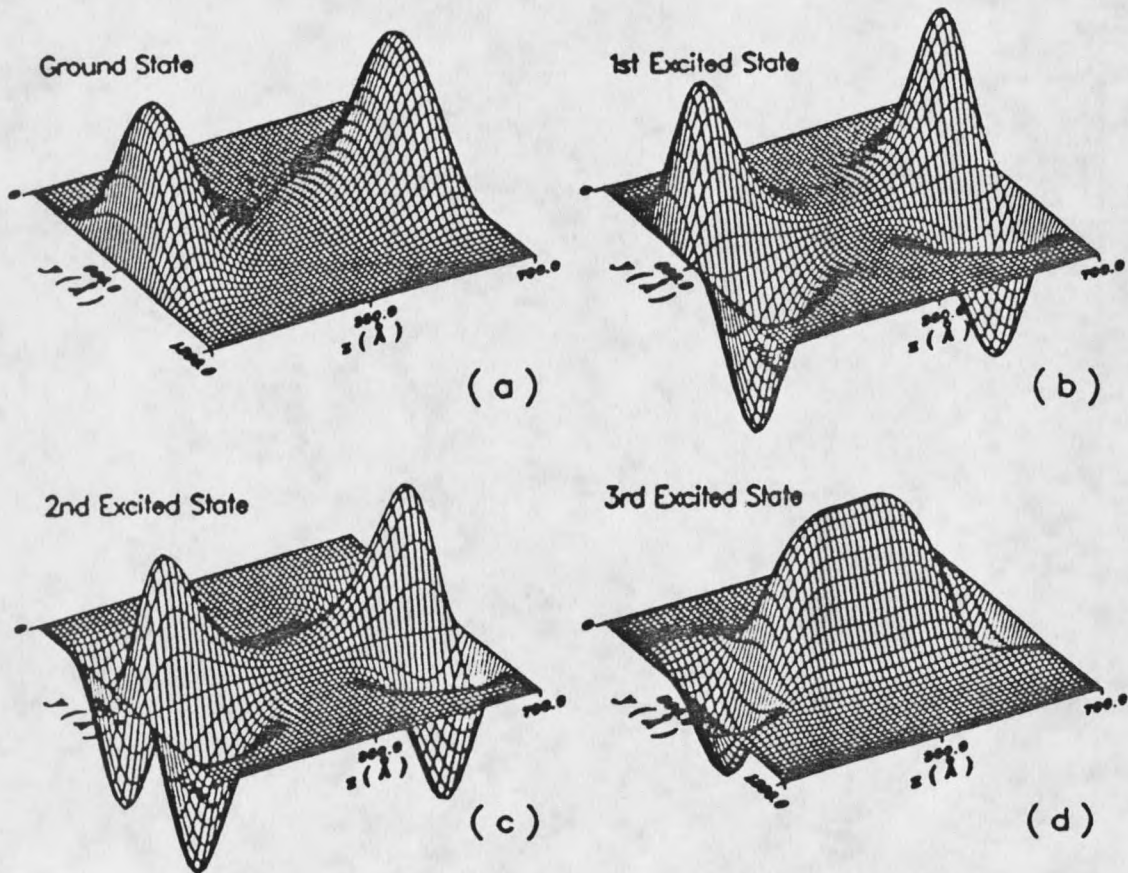


Fig. 29 Wave functions of the first four subbands calculated for a rectangular quantum wire of dimensions  $a=1200 \text{ \AA}$  and  $b=700 \text{ \AA}$ . the barrier widths  $y_0$  and  $z_0$  are  $40 \text{ \AA}$  and the temperature  $t=3 \text{ K}$ . Nearly degenerate partners are not included.

Fig.29(a)-(d) are illustrated the wavefunctions for the lowest four subbands of the quantum wire. The barrier widths  $y_0$  and  $z_0$  are both equal to 40 Å, while the finite barrier height  $E_B$  was chosen as 4 eV. The wavefunctions of the ground state and first three excited states are shown in Fig.29. There we see a significant accumulation(depletion) of electronic charge in the  $z$ -( $y$ -) direction, induced by the planar charge densities introduced as discussed above. The ground state wavefunction is very similar to the one found by Lai and Das Sarma<sup>84</sup>, who used a variational approach in which the extreme quantum limit (i.e., the neglect of all excited states) was assumed. We find that about 70% of the electrons occupy the ground state. This results indicate that excited states may play an important role in real quantum wires. Only the lowest three or four subbands have significant occupation in our model; a more detailed discussion will be given below. The accumulation effects clearly outweigh the effects of depletion in all but the highest excited state shown in Fig.29. Because of the relatively small occupation of the fourth subband (see Fig.29(d)) the charge density is expected to be low in the center of the quantum wire.

The self-consistent charge density and potential in the quantum wire are shown in Fig.30. Note that there is almost no overlap between charge densities associated with the two accumulation regions parallel to the surface. As a result, the lowest occupied states in the well are nearly two-fold degenerate. Also, the charge density is sharply reduced within about 100 Å from the edges of the rectangular box in the  $y$  direction. These results show that the dimensions we assumed for the quantum wire are large enough to simulate a typical quantum wire. The slight dip in the charge density halfway along the  $y$  direction is believed to be real, since a sufficient number of sine waves was used to avoid artificial oscillation, as discussed above. This dip should correspond to a rather flat potential or perhaps a minor hump; its existence indicates the importance of including excited states since the ground state wave rises monotonically to a peak in this region. From Fig.30 we see that only a

small fraction of the electrons tunnel out of the box in the  $z$  direction (the accumulation direction) and a negligible fraction in the  $y$  direction. Thus, the effect of the tunneling on the charge density in the box is not significant.

Fig.30(b) shows the self-consistent potential. This drawing clearly exhibits the attractive potential well at the accumulation surfaces and the repulsive barrier potential within the depletion layers. Most importantly, this result indicates the inadequacy of a square well potential for simulating a real quantum wire. The potential may be described as a saddle, with a deep, wide valley running along the  $y$  direction. The weak oscillations in Fig.30(b) are an artifact of the finite sine expansion of the wavefunctions. We have performed calculations with increasing numbers of sine waves and find that our results for the energy levels, band bending and Fermi level are well converged. Band bendings for the case presented in Fig.39(b) were determined by comparing the potential values at the appropriate interfaces with the values at the center of the wire. The interface planes are all defined as the locations where the finite barrier potential drops discontinuously to zero:  $z=z_0$ ,  $b-z_0$  and  $y_0$ ,  $a-y_0$ . The band bending is found to be -38.1 meV for the accumulation layers and 63.4 meV for the depletion layers.

We now compare our subband energies and the Fermi level with recent observations<sup>81,82</sup>. The energy level difference  $E_{12}$  between the ground state and the first excited state is 2.58 meV, while the separation between the first and the second excited state  $E_{23}=3.54$  meV. The lowest three subbands, which are doubly degenerate because of the two equivalent but non-overlapping potential wells, are all occupied, although the occupation number for the second excited state is very small. The Fermi level is at 6.00 meV, with only two subbands extending below this value. Two sets of data reported by Weiner *et al.*<sup>81</sup> and Brinkop *et al.*<sup>82</sup> correspond closely to the case we investigated. The pertinent parameters and results of theory and observations are given in Table 1. We did

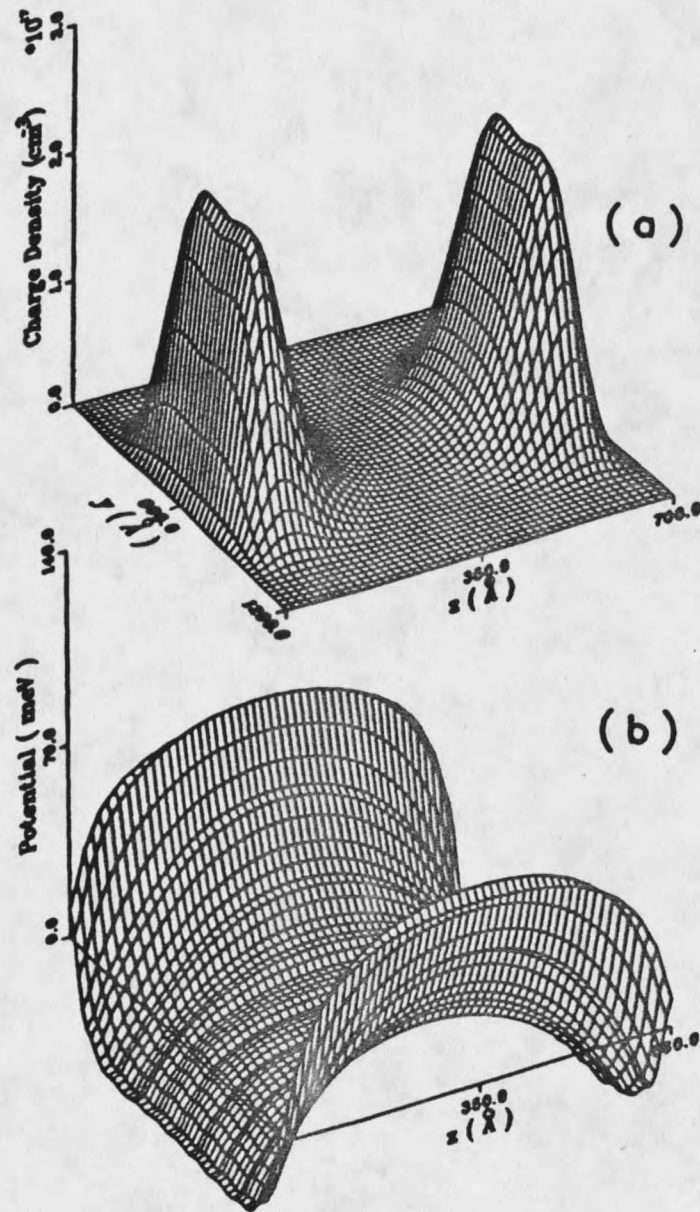


Fig. 30 Self-consistent charge density (a) and (b) potential in the quantum wire. Model parameters are the same as in Fig. 29.

not adjust our sample parameters to agree precisely with those reported earlier because of the uncertainty in the experimental values. Our calculated results agree reasonably well with the data. From the Table 1 we see that our  $E_{12}$  is slightly higher than the observed values, while the Fermi level is below the experimental value. We also calculated the resonant intersubband energies within the framework of an RPA response formalism. To estimate the shift of the intersubband resonance frequency we calculated the Coulomb matrix element  $v_{12,12}(k \rightarrow 0)$  that expresses the interaction between electron-hole pairs; the label 12 refers to an intersubband excitation from the ground state to the first excited state. The intersubband resonance frequency can be written as

$$\omega^2 = (E_{12})^2 + \omega_d^2, \quad (81)$$

where the depolarization frequency  $\omega_d$  is given by

$$\omega_d^2 = 2E_{12}n_{1D}^{12}v_{12,12}(k \rightarrow 0). \quad (82)$$

Here  $n_{1D}^{12}$  is the difference of charge densities between the first and second subbands. For the parameters used in Fig.30 and Table 1, the Coulomb matrix element  $v_{12,12}=0.23e^2/\epsilon_\infty$  and the depolarization frequency is 5.0 meV as given in Table 1.

Fig.31 shows the dependence of the energy levels, occupation numbers and the Fermi level on the total one-dimensional charge density  $n_{tot}$  in the quantum wire. The solid lines give the energy levels and the dashed line the Fermi level. Note that the number of energy levels below the Fermi level increases from one to four as  $n_{tot}$  increases over the range plotted. For  $n_{tot} > 0.025 \text{ \AA}^{-1}$ , there are four (doubly degenerate) subbands below the Fermi level; the spacing of consecutive levels increases with the energy. The level spacing

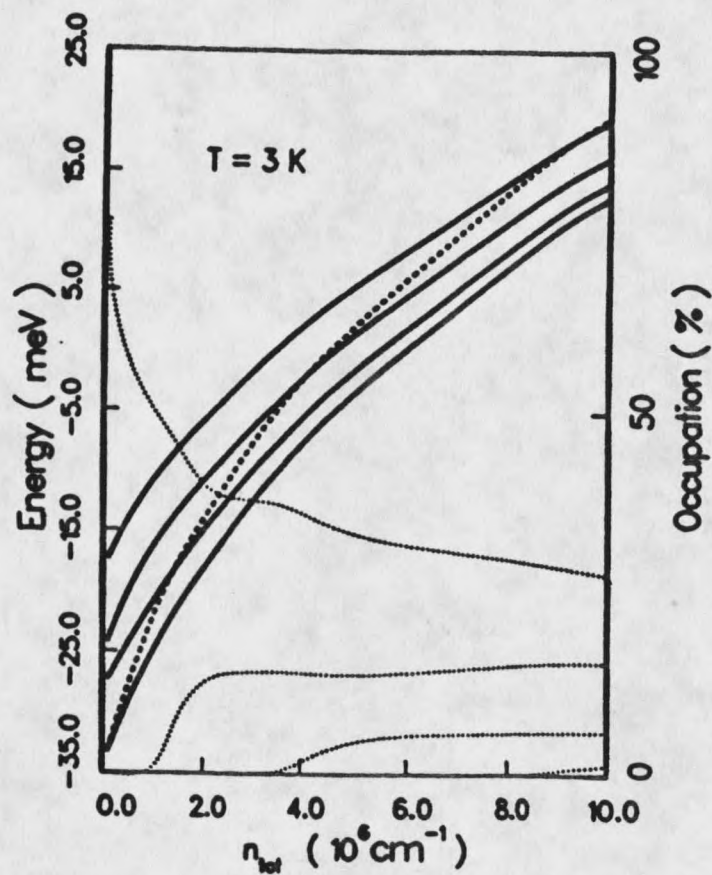


Fig. 31 Density dependence of energy levels, Fermi energy and occupation numbers in the quantum wire. Only the surface charge density  $n_{\text{sz}}$  and hence  $n_{\text{tot}}$  was varied in these calculations. The energy levels (solid lines) and Fermi energy (dashed line) are given in units of meV. The corresponding occupation numbers (dotted lines) are given in per cent.

Table 1. Comparison of representative experimental result<sup>81,82</sup> and our calculated result for a model quantum well. Most symbols are explained in the text. In the second row,  $d$  is the effective width of the conducting channel, i.e. the accumulation layer; the overall width  $b$  of the model "wave guide" was chosen to be 700 Å. Some of the experimental parameters are estimated.

|                                 | Present results      | Weiner <i>et al</i> 81 | Brinkop <i>et al</i> 82 |
|---------------------------------|----------------------|------------------------|-------------------------|
| $a(\text{Å})$                   | 1200                 | 1000                   | 1600                    |
| $d(\text{Å})$                   | 240                  | 250                    | -                       |
| $n_s(\text{cm}^{-2})$           | $7.0 \times 10^{11}$ | $2.9 \times 10^{11}$   | $6.0 \times 10^{11}$    |
| $T(\text{K})$                   | 3                    | 3                      | 3                       |
| $n_{\text{ID}}(\text{cm}^{-1})$ | $3.0 \times 10^5$    | $2.6 \times 10^5$      | $3.7 \times 10^5$       |
| $E_p(\text{meV})$               | 6.0                  | 9.0                    | 11.7                    |
| $\Delta E^{1,2}(\text{meV})$    | 2.6                  | 2.2                    | 2.0                     |
| resonance<br>energy (meV)       | 5.6                  | -                      | 5.8                     |
| depolarization<br>energy (meV)  | 5.0                  | -                      | 6.3                     |

and their degeneracies depend on the charge density because the potential does. The occupation numbers also depend on  $n_{\text{tot}}$ , of course. The saturation of the occupation numbers at high charge density is a consequence of the fact that the energy level spacings become fixed.

### Dynamical Structure Factor of a Quantum Wire in the Random-Phase-Approximation

Utilizing the electronic subband structure of a quantum wire in the GaAs/AlGaAs heterostructure studied by us<sup>86</sup>, we present, for the first time, self-consistent calculations of the single-particle and collective excitations in a quasi-one-dimensional electron gas<sup>87</sup>. The dynamical structure factor, evaluated in the random-phase-approximation (RPA), displays unique line shapes and dispersion relations which are in sharp contrast with the results expected for higher-dimensional electron gases. A well defined spectral feature is attributed to an acoustic plasmon. The calculated shift of the intersubband resonance frequency, caused by the depolarization effect, agrees well with previous experiments.

Concerning the dynamical screening properties of a Q1DEG, a few groups have measured the intersubband plasmon resonance frequency in the long wavelength limit ( $k \rightarrow 0$ ). Brinkop *et al*<sup>82</sup>, recently reported the measurement of intersubband resonance energies of a narrow inversion channel in  $\text{Al}_x\text{Ga}_{1-x}\text{As}/\text{GaAs}$  heterojunctions with static magnetotransport and infrared spectroscopy. Almost at the same time, measurements of the far-infrared(FIR) response of arrays of periodic single- and double-layer quantum wires were reported by Demel *et al*<sup>88</sup>. The most striking results of these experiments is that the intersubband resonance energies are significantly higher than the corresponding energy differences of the subbands, due to the depolarization effect. Theoretically, studies have been made of the collective motion of electrons for lateral multiple-quantum-wire

superlattices<sup>89-91</sup>. Most recently, Li and Das Sarma<sup>92</sup> presented results of the dispersion curves for both intersubband and intrasubband plasmons. To provide a more detailed interpretation of experimental results and more accurate predictions of the electronic properties of the Q1DEG, self-consistent calculations of the potential and subband structure as well as density response function are highly desirable.

If we suppose that an external probing beam of particles interacts weakly with the Q1DEG system under consideration, the differential scattering cross section is expressed by equation(15) in Chapter 2. In that equation  $S(q,\omega)$  is the dynamical structure factor, which contains all the dynamical properties of the Q1DEG. Griffin and Harris related the dynamical structure factor to the density response function  $\chi(r,r';\omega)$ , just as given in equation(16).

In the random-phase-approximation (RPA), the density response function for an inhomogeneous electron gas can be obtained by solving the integral equation

$$\chi(\vec{r},\vec{r}';\omega) = \chi^0(\vec{r},\vec{r}';\omega) + \int d\vec{r}_1 \int d\vec{r}_2 \chi^0(\vec{r},\vec{r}_1;\omega) v(\vec{r}_1,\vec{r}_2) \chi(\vec{r}_2,\vec{r}';\omega), \quad (83)$$

which is the same as equation(27), where  $\chi^0(r,r';\omega)$  is the "free-electron" response function and  $v(r_1,r_2)$  is the Coulomb interaction. In our quasi-one-dimensional case, with translational invariance in the x-direction only, we introduce the Fourier transformation

$$\chi(\vec{r},\vec{r}';\omega) = \int \frac{dk}{2\pi} e^{ik(x-x')} \chi(y,y';z,z';k,\omega), \quad (84)$$

where  $\chi(y,y';z,z';k,\omega)$  is the reduced density response function, which is the key ingredient for the calculation of the dynamical structure factor  $S(q,\omega)$ , and  $k$  is the x-

component of the wavevector. Upon Fourier transforming the RPA integral equation(equation(83)), we are led to the integral equation

$$\begin{aligned} \chi(y, y'; z, z'; k, \omega) = & \chi^0(y, y'; z, z'; k, \omega) \\ & + \int_0^a dy_1 \int_0^b dz_1 \int_0^a dy_2 \int_0^b dz_2 \chi^0(y, y_1, z, z_1; k, \omega) \\ & \cdot v_e(y_1, y_2; z_1, z_2; k, \omega) \chi(y_2, y'; z_2, z'; k, \omega), \end{aligned} \quad (85)$$

where the integration limits  $a$  and  $b$  are defined by the wave-guide geometry shown in Fig28, and  $v$  is the electron-electron interaction in the wire. As in the surface problems of InAs and ZnO (see equation(42)), the reduced noninteracting density response function  $\chi^0(y, y', z, z'; k, \omega)$  can be described by the electronic wavefunctions of the quasi-one-dimensional electronic system, by applying equation(19) to the Q1DEG, as

$$\chi^0(y, y'; z, z'; k, \omega) = \sum_{i_1, i_2, j_1, j_2} S_{i_1 i_2, j_1 j_2}(k, \omega) \phi_{i_1 i_2}(y, z) \phi_{j_1 j_2}(y, z) \phi_{i_1 i_2}(y', z') \phi_{j_1 j_2}(y', z') \quad (86)$$

where the coefficients  $S_{i_1 i_2, j_1 j_2}(k, \omega)$  are given, at finite temperature, by

$$\begin{aligned} S_{i_1, i_2, j_1, j_2}(k, \omega) = & 2 \int_{-\infty}^{\infty} \frac{dq}{2\pi} \frac{1}{1 + \exp[\beta(\epsilon_{i_1, i_2} + \hbar^2 q^2 / 2m^* - \mu)]} \\ & \left[ \frac{1}{(\epsilon_{i_1, i_2} - \epsilon_{j_1, j_2}) - \hbar^2 qk / m^* - \hbar^2 k^2 / 2m^* - \hbar(\omega + i\eta)} \right. \\ & \left. + \frac{1}{(\epsilon_{i_1, i_2} - \epsilon_{j_1, j_2}) - \hbar^2 qk / m^* - \hbar^2 k^2 / 2m^* + \hbar(\omega + i\eta)} \right] \end{aligned} \quad (87)$$

Here the coefficient 2 in front of the integral is due to the spin degeneracy. The procedure for obtaining the dynamical properties is the same as the one in the surface problem. The above integral equation (equation(85)) is discretized by defining the Fourier transformation,

$$\chi(y, y'; z, z'; k, \omega) = \sum_{m_1} \sum_{n_1} \sum_{m_2} \sum_{n_2} \chi_{m_1 n_1; m_2 n_2} \cos \frac{m_1 \pi y}{a} \cos \frac{n_1 \pi z}{b} \cos \frac{m_2 \pi y'}{a} \cos \frac{n_2 \pi z'}{b} \quad (88)$$

The same transformation was applied to  $\chi^0(y, y'; z, z'; k, \omega)$ , and we find the result

$$\chi^0(y, y'; z, z'; k, \omega) = \sum_{m_1} \sum_{n_1} \sum_{m_2} \sum_{n_2} \chi^0_{m_1 n_1; m_2 n_2} \cos \frac{m_1 \pi y}{a} \cos \frac{n_1 \pi z}{b} \cos \frac{m_2 \pi y'}{a} \cos \frac{n_2 \pi z'}{b} \quad (89)$$

where the Fourier coefficients  $\chi^0_{m_1 n_1; m_2 n_2}$  can be obtained explicitly with use of the expansion coefficients for the wavefunction (see equation(79)):

$$\begin{aligned} \chi^0_{m_1 n_1; m_2 n_2}(k, \omega) = & \frac{\mu_{m_1} \mu_{n_1} \mu_{m_2} \mu_{n_2}}{16a^2 b^2} \sum_{i_1 i_2} \sum_{j_1 j_2} \sum_{k_1 k_2} \sum_{l_1 l_2} \sum_{k'_1 k'_2} \sum_{l'_1 l'_2} S_{i_1 i_2; j_1 j_2}(k, \omega) \\ & \cdot b_{k_1 k_2}^{i_1 i_2} b_{k'_1 k'_2}^{j_1 j_2} b_{k_1 k_2}^{i_1 i_2} b_{k'_1 k'_2}^{j_1 j_2} \cdot (\delta_{m_1, k_1 - l_1} + \delta_{m_1, l_1 - k_1} - \delta_{m_1, k_1 + l_1}) \\ & \cdot (\delta_{n_1, k_2 - l_2} + \delta_{n_1, l_2 - k_2} - \delta_{n_1, k_2 + l_2}) \\ & \cdot (\delta_{m_2, k'_1 - l'_1} + \delta_{m_2, l'_1 - k'_1} - \delta_{m_2, k'_1 + l'_1}) \\ & \cdot (\delta_{n_2, k'_2 - l'_2} + \delta_{n_2, l'_2 - k'_2} - \delta_{n_2, k'_2 + l'_2}), \quad (90) \end{aligned}$$

where the coefficients  $S$  are given by equation(87) and the coefficients  $b_y^u$  are the solutions of the self-consistent Kohn-Sham problem (see equations(79) and (80)).

With the sample parameters chosen to simulate the experimental situation, representative results of the calculated RPA dynamical structure factor  $S(q, \omega)$ , using the parameters given in Fig.30, are shown in Fig.32. In the calculation we fixed the  $y$  and  $z$

components of the wavevector such that  $q_y a = q_z b = 0.1$ . The dynamical structure factor shown in Fig.32 is a function of the wavevector  $q$  along the quantum wire and of the frequency. We focus first on the single-particle excitation spectra as represented by the dotted lines, which were calculated from the non-interacting density response function  $\chi^0(y, y'; z, z'; k, \omega)$ . When  $q = 0.0001 \text{ \AA}^{-1}$  (see Fig.32(a)), the three lowest intrasubband sp excitation peaks are nearly degenerate, forming a single peak near zero frequency. The peaks at  $\omega = 2.68 \text{ meV}$ ,  $3.69 \text{ meV}$  and  $4.69 \text{ meV}$  are attributed to intersubband sp transitions corresponding to  $E_{12}$ ,  $E_{23}$  and  $E_{34}$ , respectively. The high frequency modes (above  $\omega = 7 \text{ meV}$ ) are also intersubband resonances, involving transitions between the ground state and states above the Fermi energy. When  $q = 0.001 \text{ \AA}^{-1}$  we see three intrasubband sp excitation peaks at low frequency; the intersubband sp transitions overlap to form a rather broad structure. The three intrasubband peaks and the intersubband modes become broader when the wavevector is increased further.

From Fig.32(d), we see that when  $q$  is  $0.003 \text{ \AA}^{-1}$  the three intrasubband modes produce a step-function-like structure. The peak near  $3 \text{ meV}$  is caused by the overlapping of the first and second intrasubband continua. Fig.32 shows that there is an energy gap for all of the intrasubband sp excitations at finite wavevector ( $q$  is finite but smaller than twice the Fermi wavevector  $k_F$ ). The gaps are wider and the spectral structure of  $S(q, \omega)$  are stronger and broader at larger wavevectors. The step-like line shape of the RPA  $S(q, \omega)$  and the energy gaps for sp intrasubband excitations at finite wavevector are general features of the Q1DEG and are absent in three- and two-dimensional electron gases. For the latter cases there is no gap and  $S(q, \omega)$  displays a triangle shape (3D) or a peak (2D) near the upper limit of the sp continuum at finite  $q$ . In our calculation we find that for a given subband, the dynamical structure factor of the intrasubband sp excitations is small outside region I, shown in the insert of Fig.33, defined by the inequalities

$$v_F^i q \left(1 - \frac{q}{2k_F^i}\right) < \omega < v_F^i q \left(1 + \frac{q}{2k_F^i}\right) \quad q < 2k_F^i, \quad (91)$$

where  $k_F^i$  and  $v_F^i$  are the Fermi wavevector and the Fermi velocity for the  $i$ th subband. At zero temperature the structure factor vanishes outside this region. We show the sp excitations of the lowest subband by the vertically shaded area in Fig.33, while the sp excitations of the second subband are represented by the horizontally shaded area. The cross shaded area is the region of overlap between two sp regions. The diagonally shaded area shows the sp excitations of the third subband. For finite wavevectors, the frequency of the intersubband sp excitation is limited by the relations

$$v_F^i q \left(-1 + \frac{q}{2k_F^i}\right) + E_{ij} < \omega < v_F^i q \left(1 + \frac{q}{2k_F^i}\right) + E_{ij} \quad (92)$$

In Fig.33 we show the upper limit of the intersubband sp band with the energy  $E_{12}$  by a dashed line.

We turn now to the collective intersubband and intrasubband excitations of the Q1DEG, whose spectra are shown in Fig.32 by solid lines. In the long wavelength limit,  $q \rightarrow 0$ , the energies of all three intrasubband plasmons vanish at the origin. For  $q=0.0001$

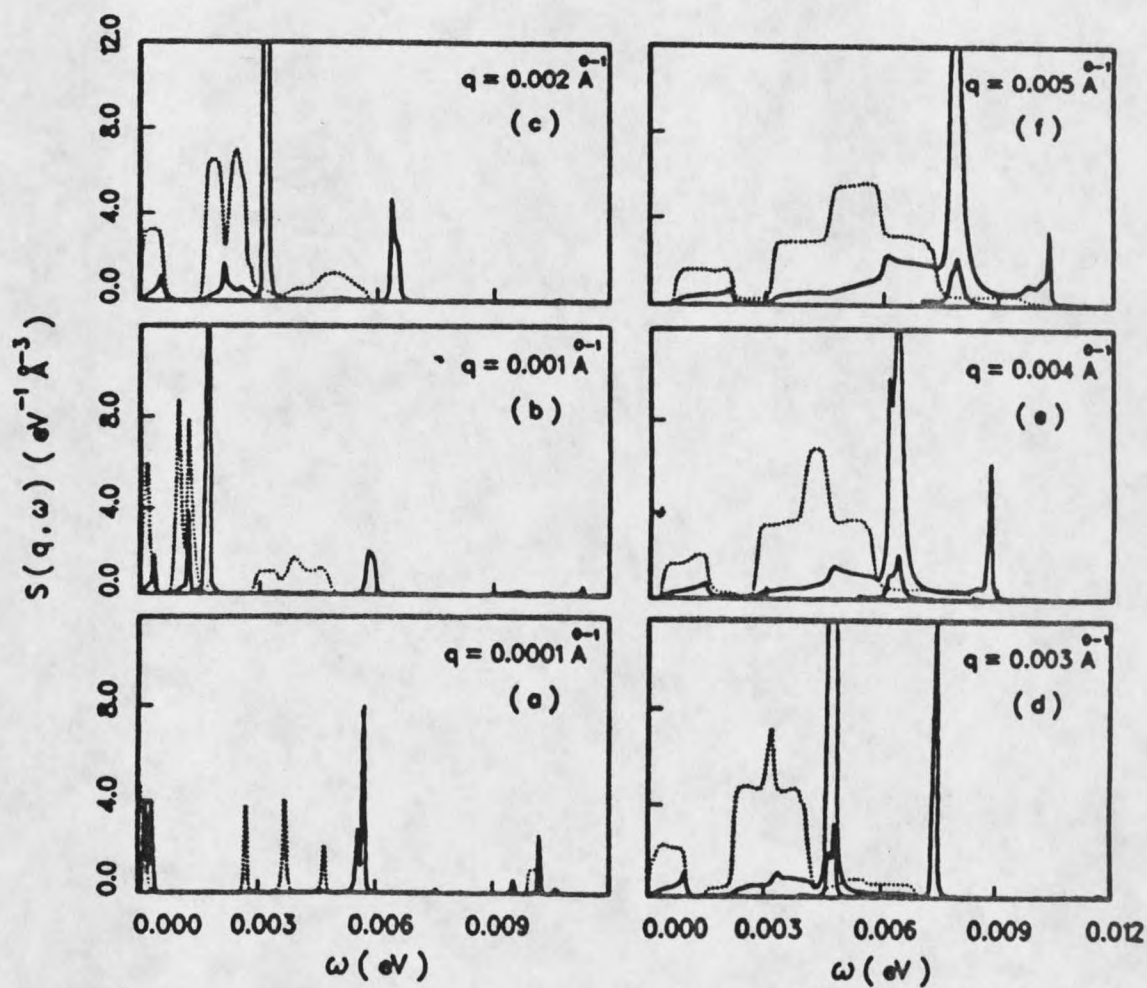


Fig. 32 Dynamical structure factor based on the results shown in Fig. 30. The charge density (spin density) excitations are represented by solid (dotted) lines.

$\text{\AA}^{-1}$ , two intrasubband plasmon peaks attributed to the lowest two subbands can be seen at low frequency. The intersubband plasmons associated with  $E_{12}$  and  $E_{23}$  are peaked at  $\omega=5.69$  meV and  $\omega=9.55$  meV, respectively. A weak peak split off the left of the intersubband plasmon is attributed to the weak interaction of modes generated by the two surfaces. This interaction is important at small  $q$  because the range of the associated potentials is inversely proportional to the wavevector. In the high frequency region (above 10 meV), there is a collective mode coinciding with the upper edge of the intersubband continuum that corresponds to excitations between the ground state and a state above the Fermi level. When  $q=0.001 \text{ \AA}^{-1}$ , three well-defined intrasubband plasmon peaks are distinguishable and their frequencies are slightly above their respective sp continua; the latter are shown with dotted lines in Fig.32.

In our calculations of the ground state properties of the Q1DEG, we found that about 70% of the electrons occupied the lowest subband while about 20% of the electrons occupied the first excited subband. The intensities of the intrasubband plasmon reflect roughly this electron occupation ratio. As the sp continua broaden, the collective intrasubband modes due to the second and third subbands are buried in them, and the peaks are Landau damped, as seen in Fig.32(c). The weak coupling between the modes on different surfaces is responsible for the small peak on the low frequency side of the main intrasubband mode (see Fig.32(d) and (e)). Note that the intersubband plasmon and the upper edge of the intersubband sp continuum move closer together as  $q$  increases. Consequently, the intensity of the collective intersubband transition is decreased when Landau damping sets in (Fig.32(d) to (f)). Throughout Fig.32 we see that only the plasmon associated with intrasubband excitations within the lowest subband is well above the sp continuum. In this case the plasmon is quite conspicuous because of its large intensity and weak Landau damping.

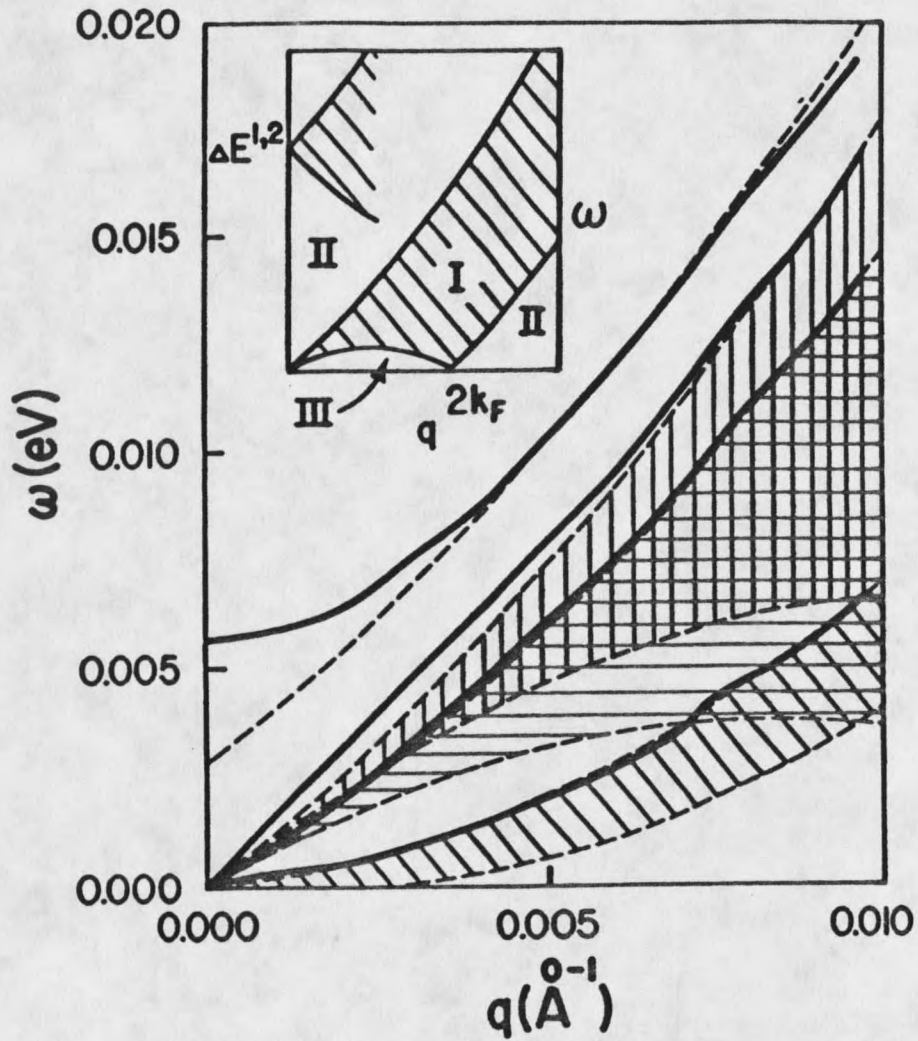


Fig. 33 Dispersion relations of collective modes in the quantum wire. The vertically (horizontally) shaded region depicts the intrasubband single-particle continuum associated with the first (second) subband at zero temperature. The upper limit of the intersubband continuum  $1 \rightarrow 2$  is given by the uppermost dashed line. The insert shows the continua belonging to the lowest subband on a more extended wavevector scale.

Note that for the Q1DEG the intersubband plasmon frequency in the long wavelength limit ( $q \rightarrow 0$ ) can be as large as three times the energy difference between subbands or perhaps even larger<sup>83</sup>, due to the effect of the depolarization field. Indeed, Brinkop *et al*<sup>82</sup> reported a case in which the energy difference of the lowest two subbands is 2.0 meV but the resonance energy is 5.8 meV. We performed calculations simulating their experimental situation and found that the collective intersubband resonance energy associated with  $E_{12}$  is upshifted from 2.58 meV to 5.69 meV while that of  $E_{23}$  is shifted from 3.54 meV to 9.55 meV as shown in Fig.32(a). Our results agree reasonably well with the observations<sup>83</sup>.

Finally, we discuss the frequency vs. wavevector dispersion relations of the collective excitations. The dispersions of the intersubband and intrasubband plasmons, shown in Fig.33 by solid lines, are defined as the locations of maxima in the RPA dynamical structure factor  $S(q, \omega)$ . The dispersion curves for intrasubband plasmons of the second and third subbands nearly coincide with the edges of their respective sp continua. The intersubband plasmon dispersion is qualitatively the same as that we found for a quasi-two-dimensional electron gas except that here the depolarization shift is much larger. The intrasubband plasmon dispersion of the lowest subband is almost linear as expected for an acoustic plasmon. For our low but finite temperature case,  $T=3K$ , the acoustic plasmon mode is only weakly Landau damped at very large wavevector. We anticipate that at higher temperature the mode will still be well-defined.

## CHAPTER 5

## CONCLUSIONS

We have applied an accurate and efficient theoretical method to study the collective excitations and energy-loss spectra of accumulation layers on InAs within the random-phase-approximation. Spectra calculated for several surface charge densities were used to interpret HREELS observations taken at various exposures to atomic hydrogen and at a range of primary-beam energies. Our interpretation focused on the behavior of three "plasmaron" modes of the coupled plasmon-phonon system, whose energies and intensities are sensitive to the band bending. We also performed loss function calculations in which the phonon response was omitted in order to gain insight into the possible 2D behavior of the accumulation layer on InAs. The computed loss function contains intersubband as well as intrasubband plasmon peaks; the dispersion relations of the intrasubband modes provide strong evidence of 2D character. A narrow peak at the edge of the pair excitation spectrum was identified as an acoustic plasmon. For a weak accumulation layer intersubband transitions lead to a breakdown of the 2D behavior.

We also determined the self-consistent electronic structure of strong accumulation layers on ZnO. The subband energy levels and wave functions were found to be weakly affected by factors such as the position dependence of the effective mass, and tunneling beneath the surface barrier. Using a nonlocal description of dielectric response within the RPA, we applied our subband calculations to predict the collective excitations of the accumulation layer on ZnO surfaces. Our results for ZnO surfaces are in qualitative agreement with the experimental data. As yet, neither the  $\omega_{\text{L}}$  plasmon mode nor the intersubband plasmon has been observed in HREELS. Higher resolution may yield better understanding of these excitations. Also, experiments done at varying impact energies and

angles of incidence would be useful in clarifying the localization properties of the modes.

For the first time, we report first-principles calculations of electronic subband structure and collective excitations of a quantum wire based on sample parameters chosen to match the GaAs/AlGaAs heterostructure recently fabricated by molecular beam epitaxy. The Poisson and Schrodinger equations in a wave-guide geometry were solved self-consistently. Significant discrepancies between our selfconsistent results and the prediction of the simple particle-in-a box model were found due to the presence of both depletion and accumulation regions. We find that the quasi-one-dimensional electron gas occupied three or four subbands whose spacings are in good agreement with experiment. The dynamical structure factor and the dynamical density response function for our inhomogeneous electron gas were evaluated in the RPA. We obtained the spectra and the dispersion relations of single particle excitations and collective excitations for both inter- and intra-subband transitions in the quantum wire. The spectra display unique line shapes and dispersion relations which are in sharp contrast with the results expected for higher-dimensional electron systems. The intra-subband single-particle excitation spectrum shows a step-function-like shape as well as overlapping sp continua of different subbands. The calculated shift of the intersubband plasmon resonance frequency, caused by the depolarization effect, agrees well with experiment. Our results indicate that the RPA can be effective in the study of elementary excitations in a low-dimensional electronic system.

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