Single-particle and collective excitations of a two-dimensional electron gas at the Cs/InAs(110) surface

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The accumulation space-charge region at a semiconductor surface has been studied by a joint investigation of the plasmon excitation and the spectral density of the quasi-two-dimensional electron gas (Q2DEG). The analysis has been performed by means of high-resolution electron-energy-loss spectroscopy and high-resolution ultraviolet photoemission, respectively. The accumulation layer was produced by depositing tiny amounts of Cs on the InAs(110) surface. By using a semiclassical dielectric model, the formation of the Q2DEG in the subsurface region was unambiguously proved by a satisfactory description of the coverage and primary energy dependence of the collective excitations. The characteristic parameters of the Q2DEG, i.e., charge density and width of the space-charge region, are determined. These results are in very good agreement with the values deduced by self-consistently solving the Poisson and Schrödinger coupled equations, which also give the eigenvalue spectrum and spectral density as measured by photoemission.

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The development of micro- and nanodevices, where confinement effects play a significant role,[1] renders the understanding of the space-charge region properties important. A space-charge layer can be formed at clean or metal-semiconductor interfaces and can present either depletion or accumulation of charge. While depletion layers have been widely studied at medium- to wide-gap III-V semiconductors, where electronic levels are induced within the gap, charge accumulation often occurs in narrow-gap systems. The accumulation layer is produced by defect- or adsorbate-induced charge at the surface, attracting free carriers of opposite sign below the surface. When the width of the potential well set up by the bound charges (screened by the free carriers) is comparable to the charge carrier wavelength, the carriers in the accumulation layer are free to move parallel to the surface but are confined along the surface normal. Therefore, their electronic levels are quantized in this direction, forming a quasi-two-dimensional electron gas (Q2DEG).[1] The interaction of alkali metals with InAs (Ref. 2) or InSb (Ref. 3) surfaces constitutes a viable example of a charge accumulation layer.

High-resolution electron-energy-loss spectroscopy (HREELS) has proved to be an efficient tool to investigate the space-charge region induced at semiconductor surfaces. In particular, the study of the accumulation layer at semiconductor surfaces has been approached by the pioneering works of Matz and Lüth[4] and Many *et al.*,[5] where the HREEL spectra taken on atomic hydrogen-exposed InAs and ZnO surfaces could be only qualitatively described. The HREELS theoretical framework was mainly developed by Ibach and Mills[6] and Lambin, Vigneron, and Lucas[7] groups in the 1980s. Ehlers and Mills first gave a description of the dielectric response in space-charge layers.[8] Within this theoretical framework, a quantitative interpretation of the HREEL measurements on InAs and ZnO has been provided by Chen, Hermanson, and Lapeyre,[9] including the electron quantization and introducing the nonlocal response.[10]

From the above studies it has emerged that the major contribution to the dielectric response could be associated to that of the ideal two-dimensional gas, which can be well reproduced by using a simple three-layer dielectric model. Recently this technique has been applied to characterize the Q2DEG generated in bulk semiconductors by selective doping (δ doping)[11] or at the Cs/InSb interface.[3] In the above-mentioned cases, however, the plasma frequency of the Q2DEG was so low that the associated loss feature could not be resolved and just contributed to the quasi-elastic-peak broadening or produced very strongly coupled plasmon-phonons (plasmarons), complicating the analysis work. In InAs, a small electron effective mass combined with a strong accumulation layer at the surface when exposed to alkali metals leads to a plasma frequency sufficiently large to give rise to a well-resolved and practically uncoupled plasma feature, enabling a detailed study of the Q2DEG by means of HREELS.

In this work, we study the collective excitations and the spectral density of a Q2DEG induced by Cs adsorption at the InAs(110) surface, by high-resolution electron-energy-loss spectroscopy and high-resolution ultraviolet photoelectron spectroscopy (HRUPS). Analysis of the HREELS data by a simple dielectric model and of the HRUPS data by solving the coupled Poisson and Schrödinger equations gives the most relevant parameters characterizing the Q2DEG, namely, the spatial distribution of the accumulated charge density (as well as its integrated value) and the thickness of the space charge region.

The experiments, carried out at the SESAMO and LOTUS surface physics laboratories of the Physics Depart-
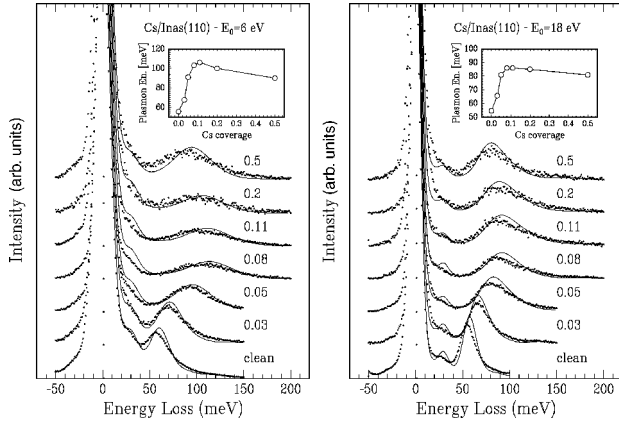
FIG. 1. High-resolution electron-energy-loss spectra of the Cs/InAs(110) interface as a function of coverage. Data taken at two different primary beam energies (6 eV and 18 eV), and in specular conditions, with incidence and scattering angles of 65° with respect to the surface normal. Results of a theoretical dielectric model fit to the experiment are superimposed as solid lines to the experimental data (dots). Insets show the plasmon energy as a function of the coverage.

ment at the Università di Modena e Reggio Emilia, were performed in an ultrahigh-vacuum (UHV) chamber containing the HREELS spectrometer and the HRUPS system, together with other ancillary facilities for sample preparation. HREELS measurements were performed in the specular geometry with primary beam energies ranging between 6 and 22 eV and with an overall resolution of about 10 meV, as derived from the full width at half maximum (FWHM) of the elastic peak. The photoelectron spectra were excited by a high-intensity He discharge lamp (He I photons, $h\nu = 21.218$ eV) and measured by a Scienta SES-200 hemispherical analyzer. The integration angle was about $\pm 6°$ with respect to the normal emission direction. The instrumental energy resolution was better than 15 meV, as determined on the Fermi level (E_F) of freshly evaporated Au in good electrical contact with the sample. The (110) clean surface was obtained by cleaving *in situ* an *n*-type InAs single crystal ($n = 4 \times 10^{17}$ cm^{-3}). Pure Cs was evaporated from well-outgassed SAES-Getters dispensers at a pressure better than 1.5×10^{-8} Pa. Cs coverage is defined as a fraction of the Cs saturation coverage ($\Theta = \Theta_{SAT}$), which corresponds to the saturation value of the Cs 5*p* core level intensity.

The evolution of the space-charge layer at the Cs/InAs(110) system was followed by studying the collective excitation of the electron gas, by means of high-resolution electron-energy-loss spectroscopy. A selected set of HREEL spectra (symbols) taken at the Cs/InAs(110) interface as a function of coverage is shown in Fig. 1. The spectra of the clean surface are characterized by two main loss features: the Fuchs-Kliewer optical phonon ($\omega_{TO} = 26.9$ meV) and the free-carrier electron-gas excitation, i.e., the plasmon loss, at higher loss energy. Due to the long-range dipole Coulomb field associated with both excitations, they are coupled, forming plasmaron modes. However, in our case, taking into account the wave vector range spanned in the experiment, together with the relatively wide energy separation between

the two unperturbed excitations, their interaction is sufficiently small to enable us to separately denote them as phonon and plasmon modes.[10] Cs deposition onto the clean surface induces an energy shift of the plasmon feature (higher-energy-loss mode) towards higher energy, indicating an increased charge density. Comparison of the plasmon energy value at the two different primary energies confirms the plasmon system to behave as a 2DEG. In fact, at higher primary beam energy the plasmon is located at lower loss energy, according to momentum and energy conservation,

$$q_{\parallel} = k_{\parallel} \frac{\hbar \omega}{2E_0},$$

and to the plasmon dispersion behavior for an ideal, single-level 2DEG sandwiched between two half-spaces characterized by the dielectric functions ε_1 and ε_2 (Ref. 1):

$$\omega_{pl} = [4\pi e^2 n_{2D}/m^*(\varepsilon_1 + \varepsilon_2)]^{1/2} \sqrt{q_{\parallel}},$$

where $k_{\parallel}$ and E_0 are the momentum and the kinetic energy of the impinging primary electrons, and $q_{\parallel}$ is the transferred momentum.

However, by analyzing the plasmon energy loss value as a function of primary beam energy (thus as a function of the transferred parallel momentum) we do not measure exactly a square-root dispersion. One reason for this is because the environment of the actual Q2DEG is not semiinfinite as that of the ideal 2DEG model.

A second striking characteristic of the plasmon feature that can be observed in the spectra shown in Fig. 1 is that it becomes broader and broader with increasing coverage, together with the increase of a characteristic tail on the higher-energy-loss side. This behavior is not expected in the collective excitation of a 3D electron gas, where the plasmon feature is symmetrical.[12]

The assumption of a 2D electron gas, therefore, can qualitatively account for the evolution of the HREEL spectra with coverage. A more quantitative characterization of the accumulation layer can be achieved by reproducing the HREEL spectra by means of a dielectric model, where the 2D carrier density is considered as an adjustable parameter. Assuming that the dielectric response can be reasonably approximated by the ideal 2DEG, a method for simulating the parabolic dispersion consists in modeling the system as a "thin" 3D electron gas, sandwiched between a depleted surface region (dead layer) and a semi-infinite substrate having the bulk doping level (unscreened substrate plasmon frequency of 160 meV).[11] The "thin" requirement for the 3DEG thickness is with respect to the characteristic spatial extension of the electric fields generated by the excitation, i.e., $\mathbf{q}_{\parallel}^{-1}$. In the momentum region typically spanned by a HREELS experiment performed in dipole geometry, a thickness of about 5 Å can satisfactorily simulate the behavior of a two-dimensional electron gas. Notice that its thickness is fictitious, while only the electronic area density $n_{2D} = n_{3D} d_{3DEG}$ has a physical meaning.[11] The presence of a depleted layer just at the surface comes from the boundary condition imposed on the electronic function to vanish at the surface.
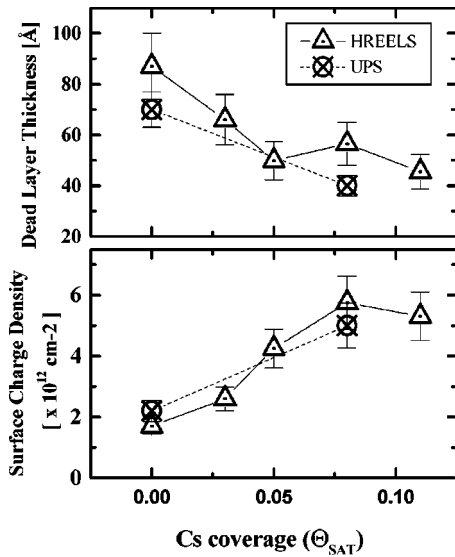
FIG. 2. Dead layer thickness (upper panel) and accumulated charge density (lower panel) as determined by HREELS and HRUPS.
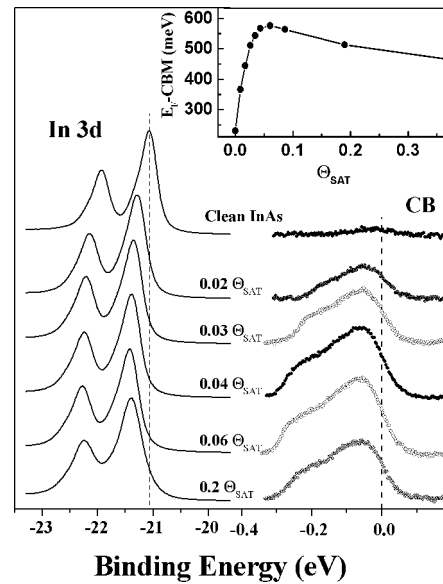

FIG. 3. HRUPS: Fermi-level region (right side) and core-level (left side) data at the Cs/InAs(110) system, as a function of Cs coverage. Photon energy of 21.218 eV and the binding energy is referred to the Fermi level. The inset shows the band bending behavior, as derived from the core-level shift.

The theoretical "fitting" of spectra to the experimental data is shown in Fig. 1 as superimposed solid lines. The calculations have been based on the model described above and performed using the Lambin code.[13] This program calculates—using a semiclassical approach—the dipolar HREEL spectrum of an arbitrary layered structure, where each layer is characterized by its dielectric properties. The only free parameters in the fitting procedure were the dead layer thickness and the electron-gas area density. The agreement is very good at all Cs coverages. The reproduction of the clean InAs spectrum required also the presence of a small accumulation layer. This can be explained assuming that a small density of cleavage defects was present on the clean surface, so that the bands are bent downwards even before Cs adsorption. The dead layer thickness and accumulated surface charge-density, extracted from the best-fit procedure for all the measured spectra, are shown in Fig. 2 as a function of the Cs coverage. In the charge density determination, we need to take into account that the strong nonparabolicity of the InAs conduction band affects the electron effective mass at the Fermi level. As the bulk density of the electron gas in an accumulation layer is in the 10^{18} cm^{-3} range, we adopt $m^* = 0.032 m_0$.[14] The charge density on the clean surface is 1.7×10^{12} cm^{-2}. It increases with Cs coverage, reaching the maximum value of 5.7×10^{12} cm^{-2}, at about $\theta = 0.08$; afterwards, it starts slowly decreasing. In the same coverage range, the dead layer thickness decreases from 87 to 57 Å. The uncertainty in these values is estimated to be about 15% for the charge density and 20% for the dead layer thickness, respectively.

The effects of the Cs deposition on the InAs(110) surface were also investigated by means of high-resolution ultraviolet photoelectron spectroscopy. The HRUPS data taken in the valence-band region close to the Fermi level are shown in Fig. 3 (right side) as a function of Cs coverage. The conduction-band minimum (CBM) is pushed down in the surface region, so that clear population of the conduction band is observable upon increasing coverage, and the spectral band shape presents evidence for a steplike signal. This behavior can be interpreted as due to the formation of a Q2DEG confined in the surface region.[3,15,16] Quantum confinement accounts for the resolved steplike Q2DEG occupied eigenstates, with progressive filling of the subbands upon increasing Cs coverage. The corresponding In $4d_{3/2}$ core-level evolution as a function of coverage is also shown in Fig. 3 (left side). The clean InAs surface spectrum shows the Fermi level to lie about 200 meV above the CBM, indicating a small downwards band bending and accumulation layer, in agreement with the previously discussed HREELS results and suggesting the presence of a small density of cleavage defects on the clean surface. Dosing very low amounts of Cs induces a much larger band bending, bringing the Fermi level deep into the conduction band and producing a deeper potential well, with a maximum depth of 600 meV at Cs coverages between 0.05 and 0.1. The inset of Fig. 3 shows the Fermi-level position with respect to the CBM as derived from the In $4d$ bulk component energy shift. We notice that the steplike spectral intensity follows the band-bending behavior; in fact, the Q2DEG is confined within the potential well between the strongly bent bands and the vacuum.

From a theoretical point of view, once the electron effective mass and bulk doping are known, the space-charge layer is completely defined by the well depth. In fact, the potential well profile, the subband levels and the wave functions can be obtained by self-consistently solving the Schrödinger and Poisson equations, so to have an alternative way to determine the characteristic parameters of the 2DEG. Thus, from the measured well depth we can obtain the 2DEG density of states as a function of the binding energy to be compared with the experiments. The details of the spectra reproduction
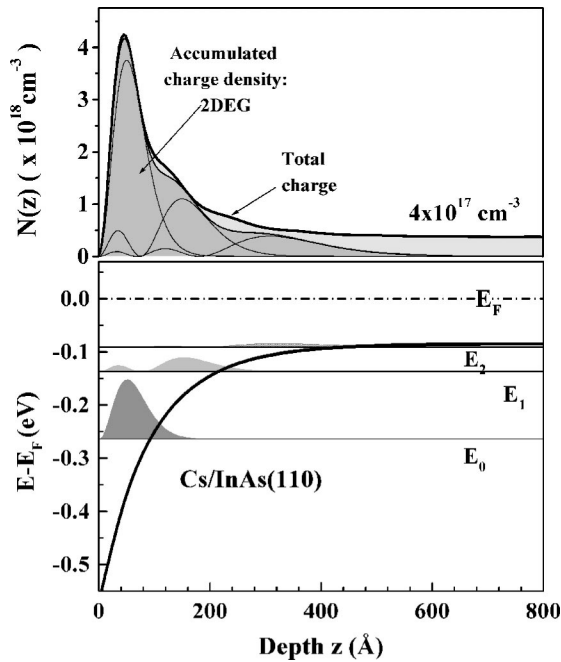
FIG. 4. Potential well shape, subband energy eigenstates, accumulated (total and partial for each eigenlevel), and total charge density, as determined by solving the Schrödinger and Poisson equations at the maximum accumulation.

have been reported elsewhere.[15,16] Here we only consider the 2DEG area electron densities obtained from the calculations and compare them with the results obtained by HREELS.

The 2DEG charge-density values obtained from photoemission are 2.2×10^{12} cm^{-2} and 5.0×10^{12} cm^{-2}, for the clean and maximum-accumulation surface, respectively. These values compare very well to the corresponding values obtained by HREELS analysis, namely, 1.7×10^{12} cm^{-2} and 5.7×10^{12} cm^{-2}. Also the "dead" layer thickness provided by HREEL agrees with the width of the depleted region at the surface, determined as the distance from the surface where the calculated 2D charge density reaches half of its maximum value. The comparison must also take into account the inevitable differences among the different surfaces used in the various experimental runs.

In Fig. 4 we plot the charge density calculated through the square modulus of the eigenfunctions obtained from the solution of the Schrödinger equation for each eigenstate and their sum, i.e., the charge density accumulated in the space-charge region. This constitutes the 2D part of the total free-carrier density. In particular, the given numerical results correspond to the filling of the quantized states up to the Fermi level. Another contribution has 3D character, and its value decreases as the well deepens, whereas the accumulated 2D charge density increases. As clearly depicted in Fig. 4, when the accumulation reaches its maximum, the bulk 3D charge density contribution in the space-charge region almost vanishes. The bulk contribution to the space-charge density is more important at lower well depth, as for the clean cleaved surface, yet its contribution is much lower than the two-dimensional part.

Notice that both UPS and HREELS are able to discriminate between the two contribution of the free-carrier charge density, leading to its characterization. This is an important point, because it contains the peculiarity of the techniques used, making them useful to study an accumulation layer. Photoemission measures directly the density of filled states of the accumulated electron gas and allows one to extract the the charge density related to the confined two-dimensional states and the nonconfined three-dimensional (bulk) ones extended to the accumulation region. On the other hand, the HREELS technique is sensitive to the collective excitations of the electronic gas, in particular to that of the two-dimensional confined part, which is the feature which dominates the spectra. Photoemission is sensitive to the single-particle character of the electron gas, whereas HREELS is sensitive to its collective character. The UPS and HREELS analysis of the space-charge region are thus completely independent and alternative. In fact, each analysis does not use any results of the other. Moreover, they are based on two different aspects of the same property of the electron gas: its density.

In conclusion, we have studied the effects of exposing the InAs(110) surface to submonolayer amounts of Cs by using HREELS and HRUPS. We have shown that a charge accumulation layer is produced by the deposition of a few hundredths of monolayers of Cs on the InAs surface, proving the two-dimensional character of the electron gas. We have determined the total and subband resolved accumulated charge density, its spectral density, and the dead layer thickness. The 2DEG characterization has been performed independently by means of the two techniques, finding full agreement between their results. We also proved that the plasmon excitation of a quasi-two-dimensional electron gas is very well approximated by the collective excitation of the ideal one. This result indicates the possibility of analyzing the HREELS data by using a very simple model, confirming this technique as a precious tool for investigating the space-charge region properties.

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