

## Intrinsic Electron Accumulation Layers on Reconstructed Clean InAs(100) Surfaces

M. Noguchi, K. Hirakawa, and T. Ikoma

*Institute of Industrial Science, University of Tokyo, 7-22-1 Roppongi, Minato-ku, Tokyo 106, Japan*

(Received 6 December 1990)

The electronic structures of clean InAs(100) surfaces have been investigated by *in situ* high-resolution electron-energy-loss spectroscopy. Intrinsic electron accumulation layers with carrier densities strongly depending on the surface reconstruction are formed on both As-stabilized and In-stabilized surfaces. The correlation between the surface electron densities and the surface reconstructions suggests that electrons in the accumulation layers are induced by the donorlike intrinsic surface states of InAs whose energy spectrum is determined by the surface reconstructions.

PACS numbers: 73.20.At, 73.20.Mf, 79.20.Kz

It is well known that an electron accumulation layer is easily formed on InAs surfaces. Recently, this surface accumulation layer has attracted much attention because the high density of electrons on the surface has great technological importance, such as the formation of non-alloyed Ohmic contacts<sup>1</sup> and the realization of the three-terminal Josephson devices.<sup>2</sup> It is, however, not clear whether an intrinsic electron accumulation layer is present even on clean InAs(100) surfaces and how it is related with surface atomic configurations.

In this work, we studied the electronic structure of both As-stabilized and In-stabilized clean InAs(100) surfaces with *in situ* high-resolution electron-energy-loss spectroscopy (HREELS). By analyzing the HREELS spectra, it is found for the first time that electron accumulation layers are formed on both As-stabilized and In-stabilized surfaces and, furthermore, that the electron density in the accumulation layer changes reversibly with surface reconstructions. The origin of such electron accumulation layers is discussed.

HREELS is a very powerful tool to investigate semiconductor surfaces because it gives us rich information on the surface vibrational excitations which extend into semiconductors by several tens of nanometers. To pursue HREELS measurements, however, it is essential to prepare clean and undamaged semiconductor surfaces. Such techniques as cleaving,<sup>3,4</sup> ion bombardment and subsequent annealing,<sup>5,6</sup> and arsenic deposition<sup>7,8</sup> were used in the previous works. With these methods, however, it is difficult to obtain clean and undamaged InAs(100) surfaces with high reproducibility. To overcome this difficulty, the HREELS system is connected with a molecular-beam-epitaxy (MBE) chamber under an ultrahigh-vacuum condition ( $< 3 \times 10^{-8}$  Pa). This configuration keeps the surface contamination negligibly small during measurements [ $\leq 0.4$  L (1 L =  $10^{-6}$  Torr) for 2 h] and allows us to investigate clean surfaces.

The As-stabilized undoped InAs(100) surfaces were prepared as follows: 0.3–0.5- $\mu\text{m}$ -thick undoped *n*-type InAs layers were grown on undoped InAs(100) sub-

strates by MBE. The bulk electron density in the MBE-grown InAs layer is less than  $2 \times 10^{16} \text{ cm}^{-3}$ . The substrate temperature during the growth was set at 450–490 °C. The As-stabilized surfaces were obtained by cooling the arsenic-stabilized (2×4) reconstructed surfaces down to room temperature in an As<sub>4</sub> flux of  $\sim 10^{15} \text{ cm}^{-2} \text{ s}^{-1}$ . During the cooling process, the reflection high-energy electron-diffraction (RHEED) pattern changed from the (2×4) pattern to the diffusive (1×1) at 350–400 °C (see the inset of Fig. 1). Although the c(4×4) reconstruction has been reported for GaAs surfaces under high As pressure,<sup>9</sup> we could not confirm such a structure. After cooling, HREELS spectra were measured in an elastic-scattering geometry under the condition where the incident angle of the electron beam was 55°.

Figure 1 shows HREELS spectra measured on the

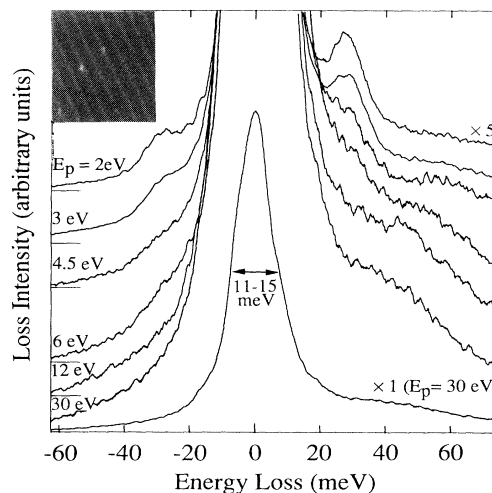


FIG. 1. HREELS spectra measured on an As-stabilized InAs(100) surface. The parameter is the incident electron energy,  $E_p$ . Inset: RHEED pattern of the As-stabilized InAs(100) surface at 300 °C. The RHEED beam direction is nearly along the [110] azimuth.

As-stabilized InAs(100) surface for various incident electron energies,  $E_p$ . By varying  $E_p$ , we can change the probing depth, which is approximately given by the inverse of the wave-vector transfer parallel to the surface.<sup>10</sup> When  $E_p$  is 2–3 eV (the probing depth is typically 20 nm), the peaks due to the Fuchs-Kliwer surface phonons are observed at  $\pm 28$  meV, as observed on the cleaved or ion-sputtered InAs surfaces.<sup>3,5</sup> However, an interesting feature appears with increasing  $E_p$  (greater probing depth). When  $E_p$  is increased up to 12 eV, a shoulder appears at  $\sim 45$  meV. This shoulder is identified as the plasma excitation of conduction electrons. This plasmon energy is much higher than that estimated from the bulk electron density ( $\leq 9$  meV), clearly indicating the presence of a natural surface accumulation layer with high electron density. Furthermore, the existence of the accumulation layer on the As-stabilized surface can be also confirmed by a drastic change in the plasmon energy (45 meV for the As-stabilized surface and less than 5 meV for In-stabilized surface as will be mentioned later); this change is much larger than that expected only by surface depletion of the  $n$ -type sample, because the plasmon energy changes only in the range from the surface plasmon energy  $\omega_{sp}$  to the interface energy  $[(\epsilon_\infty + 1)/2\epsilon_\infty]^{1/2}\omega_{sp}$  when the depletion layer width is varied.<sup>7</sup> Here,  $\epsilon_\infty$  is the high-frequency dielectric constant of InAs  $\sim 12.3$  (Ref. 11). Moreover, no time dependence was found in the shape of the shoulder during the measurement, indicating that a change in the electron densities caused by residual gas adsorption was negligible. This is in sharp contrast with the results on the cleaved InAs(110) surface, where a surface accumulation layer is absent initially and is formed only after a hydrogen exposure.<sup>3</sup> This is also different from the result on the ion-sputtered InAs(100) surface, where the electron density seems to be much lower than that on our MBE-grown As-stabilized surfaces.<sup>5</sup>

From the behavior of the peaks at  $\pm 28$  meV with various  $E_p$ , we can obtain information on the interaction between the surface phonons and plasmons. For large  $E_p$  (greater probing depth), the peaks at  $\pm 28$  meV due to the surface phonons are overridden by the large plasmon shoulder, indicating that the surface phonons couple with the plasmons and that the phonon polarization field is screened by conduction electrons.<sup>3,12</sup> With decreasing  $E_p$ , the peak becomes clearer rapidly. This fact indicates that the effective electron density decreases toward the surface and is vanishingly small at the very top of the surface, leading to the reduction of the screening effect. This is a direct evidence of quantized shape of the electron wave function. The quantization of the electron accumulation layer is also confirmed by the fact that the intensity of the plasmon peak increases with increasing  $E_p$  (increasing the probing depth). This fact cannot be explained if we assume the classical density profile of the accumulated surface electrons, because the

classical density profile has its maximum at the top of the surface.

The In-stabilized surfaces were prepared by heating the As-stabilized samples at 350–400°C under arsenic pressure lower than  $1 \times 10^{-7}$  Pa. The RHEED pattern changed from the  $(4 \times 2)$  structure to the  $(4 \times 1)$  when the samples were cooled down to room temperature (see the inset of Fig. 2). Figure 2 shows the HREELS spectra taken on the In-stabilized InAs(100) surfaces for various  $E_p$ . In contrast with the results on the As-stabilized surfaces, the peaks due to the Fuchs-Kliwer surface phonons are observed at  $\sim 28$  meV in the whole range of  $E_p$ . Furthermore, the full width at half maximum (FWHM) of the elastic peak is significantly larger than that for As-stabilized surfaces. This broadening of the elastic peak is considered to be due to the overlap of the elastic peak and the plasmon peak.<sup>12</sup> These experimental facts indicate that the electron density is low on the In-stabilized surfaces and, therefore, the surface phonons are not efficiently screened by conduction electrons.

In order to understand quantitatively the measured HREELS spectra, we simulated the HREELS spectra within the framework of the dielectric local response model developed by Lambin, Vigneron, and Lucas,<sup>13</sup> which takes account of the depth profile of conduction electrons. We used the Fang-Howard variational quantized wave function<sup>14</sup> for the accumulation layer and, furthermore, extended the model to include the Landau damping effect.

First, we calculated the  $E_p$  dependence of the HREELS spectra for the As-stabilized surfaces. In the calculation, we assume the surface electron density  $N_{ss}$

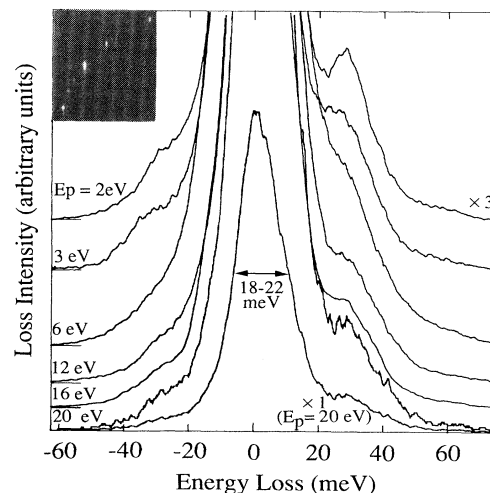


FIG. 2. HREELS spectra measured on an In-stabilized InAs(100) surface.  $E_p$  was varied from 2 to 20 eV. Inset: RHEED pattern of the In-stabilized InAs(100) surface at 300°C. The RHEED beam direction is nearly along the  $[110]$  azimuth.

to be  $1 \times 10^{12} \text{ cm}^{-2}$ , as will be shown in the next paragraph. The results are displayed in Fig. 3. When  $E_p$  is less than 6 eV, the calculated spectra show the characteristic peak due to the Fuchs-Kliwer surface phonon at  $\sim 30 \text{ meV}$  and are in good agreement with the experimental result shown in Fig. 1. This distinct surface phonon peak is the direct consequence of the low surface electron density due to the surface quantization. When  $E_p$  exceeds 6 eV, two split peaks due to the coupling between the surface phonons and plasmons are seen in the calculated spectra. With increasing  $E_p$ , both peaks shift toward the lower-energy side. The  $E_p$  dependence of the loss feature above 30 meV is in good agreement with the experimental spectra. However, the lower-energy peak was not clearly observed experimentally. This discrepancy between the experiment and the theory is reproducible and specific to the As-stabilized  $(1 \times 1)$  InAs(100) surfaces and has neither been observed on the hydrogen-exposed InAs(110) surfaces<sup>3</sup> nor on our In-stabilized InAs(100) surfaces. Although the origin of this discrepancy is not clear at present, we believe that the lower-energy peak is hidden in the elastic peak robe and the high background caused by adsorbed arsenic clusters, as suggested by Dubois and Schwartz.<sup>15</sup>

Figure 4 displays the calculated energy-loss spectra for various  $N_{ss}$ , assuming  $E_p = 12 \text{ eV}$ . By comparing the measured spectra shown in Figs. 1 and 2 with the calculated loss feature around 30–45 meV, the electron density in the accumulation layer is determined to be approximately  $1 \times 10^{12} \text{ cm}^{-2}$  for the As-stabilized surfaces,

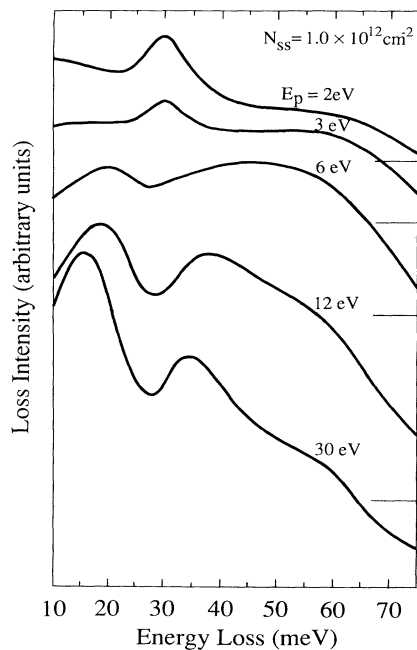


FIG. 3. Calculated HREELS spectra with different incident electron energies,  $E_p$ .  $N_{ss}$  is assumed to be  $1.0 \times 10^{12} \text{ cm}^{-2}$ .

while it is lower than  $5 \times 10^{11} \text{ cm}^{-2}$  for the In-stabilized surfaces. The former electron density corresponds to 0.27 eV downward bending of the conduction band and the latter to less than 0.17 eV. To confirm this, we performed ultraviolet photoelectron spectroscopy (UPS) measurement on the same samples and found that the valence-band maximum was located 0.58–0.65 eV below the Fermi energy ( $E_F$ ) for the As-stabilized surfaces, while it was 0.36–0.42 eV below  $E_F$  for the In-stabilized surfaces. Considering the band gap of InAs (0.36 eV) and the fact that the conduction-band minimum in the bulk is located 0.04 eV above  $E_F$ , our UPS data indicate 0.2–0.3-eV downward band bending for the As-stabilized surfaces and 0.0–0.1-eV downward band bending for the In-stabilized surfaces, well supporting the band bendings obtained by the HREELS. These results indicate that the surface electron density is strongly correlated with the surface atomic configurations.

Furthermore, it is very important to note that when the In-stabilized  $(4 \times 2)$  surfaces were converted to the As-stabilized  $(2 \times 4)$  surfaces by exposing them to an  $\text{As}_4$  flux with a pressure of  $\sim 1 \times 10^{-5} \text{ Pa}$  at the substrate temperature of 350–400 °C, the HREELS spectra also changed to those for the original As-stabilized surfaces, indicating the recovery of the high surface electron density. This conversion is reversible. The correlation between the surface electron densities and the surface reconstructions rules out a possibility that the defects in

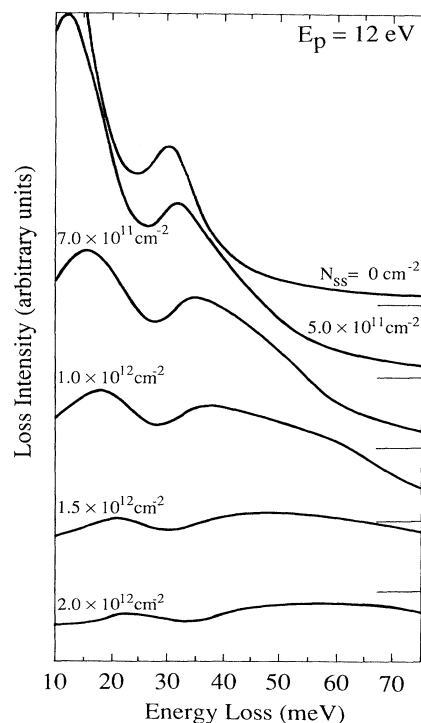


FIG. 4. Calculated HREELS spectra for different surface electron densities  $N_{ss}$ .  $E_p$  is set to be 12.0 eV.

the bulk InAs generate the surface accumulation, because the self-diffusion of In and As is so small during the low-temperature treatment in our experiment<sup>16</sup> that the densities of In- or As-related point defects are highly unlikely to depend on the surface reconstructions. Moreover, the adsorbed extrinsic atoms other than In and As cannot be the origin of the electron accumulation on the As-stabilized surface, since the adsorbed atoms are very unlikely to generate the reproducible and reversible electron accumulation which strongly correlates with surface reconstructions. Hence, it is strongly suggested that electrons in the surface accumulation layers are induced by the donorlike intrinsic surface states which are in resonance with the conduction band and whose density of states is determined by the surface reconstructions.

The above mechanism can be explained as follows: Electrons usually occupy the surface states below the charge neutrality level (CNL) where the states derive their weight equally from valence and conduction bands. However, the conduction-band minimum (CBM) at the  $\Gamma$  point in InAs is much lower than the  $X$  minima so that CNL is located slightly above CBM.<sup>17,18</sup> Therefore, the surface states between CNL and CBM can emit electrons into the conduction band, thus forming the surface accumulation layers. Our experimental finding of the correlation between the surface electron density and the surface reconstruction is the first direct evidence of the fact that the energy spectrum of the intrinsic InAs surface states are drastically modulated by the surface reconstructions.

In summary, it is found for the first time that quantized electron accumulation layers are formed on clean undoped InAs(100) surfaces grown by MBE. By analyzing the HREELS spectra taken with different probing depths, it is clarified that the coupled behavior of surface phonons and plasmons can be understood by taking into account the shape of the quantized electron wave function for the accumulation layers. By comparing the experimental results with the theoretical calculations, the electron density in the accumulation layers is determined to be  $1 \times 10^{12} \text{ cm}^{-2}$  on the As-stabilized InAs(100) surfaces and less than  $5 \times 10^{11} \text{ cm}^{-2}$  on the In-stabilized surfaces. Furthermore, the surface electron density is found to change reproducibly by controlling the surface reconstructions. This correlation between the

surface electron densities and the surface reconstructions strongly suggests that electrons in the accumulation layers are induced by the donorlike intrinsic surface states of InAs whose density of states is determined by the surface reconstructions.

This work is partly supported by the Grant-in-Aid from the Ministry of Education, Science, and Culture, Japan, by the Foundation for Promotion of Material Science and Technology of Japan (MST), and also by the Industry-University Joint Research Program "Mesoscopic Electronics."

<sup>1</sup>J. M. Woodall, J. L. Freeouf, G. D. Pettit, T. Jackson, and P. Kircher, *J. Vac. Sci. Technol.* **19**, 626 (1981).

<sup>2</sup>H. Takayanagi and T. Kawakami, *Phys. Rev. Lett.* **54**, 2449 (1985).

<sup>3</sup>Y. Chen, J. C. Hermanson, and G. J. Lapeyre, *Phys. Rev. B* **39**, 12682 (1989).

<sup>4</sup>R. Matz and H. Lüth, *Phys. Rev. Lett.* **46**, 500 (1981).

<sup>5</sup>P. A. Thiry, J. L. Longueville, J. J. Pireaux, R. Caudano, H. Muneke, and M. Liehr, *J. Vac. Sci. Technol. A* **5**, 603 (1987).

<sup>6</sup>Ph. Lambin, J. P. Vigneron, A. A. Lucas, P. A. Thiry, M. Liehr, J. J. Pireaux, R. Caudano, and T. J. Kuech, *Phys. Rev. Lett.* **56**, 1842 (1986).

<sup>7</sup>Z. J. Gray-Grychowski, R. G. Egdell, B. A. Joyce, R. A. Stradling, and K. Woodbridge, *Surf. Sci.* **186**, 482 (1987).

<sup>8</sup>R. G. Egdell, W. R. Flavell, Z. J. T. Gray-Grychowski, R. A. Stradling, B. A. Joyce, and J. H. Neave, *J. Electron Spectrosc. Relat. Phenom.* **45**, 177 (1987).

<sup>9</sup>P. K. Larsen, J. H. Neave, J. F. van der Veen, P. J. Dobson, and B. A. Joyce, *Phys. Rev. B* **27**, 4966 (1983).

<sup>10</sup>H. Ibach and D. L. Mills, *Electron Energy Loss Spectroscopy and Surface Vibrations* (Academic, New York, 1982).

<sup>11</sup>W. A. Harrison, *Electronic Structure and the Properties of Solids* (Freeman, San Francisco, 1980).

<sup>12</sup>A. Ritz and H. Lüth, *Phys. Rev. Lett.* **52**, 1242 (1984).

<sup>13</sup>Ph. Lambin, J. P. Vigneron, and A. A. Lucas, *Phys. Rev. B* **32**, 8203 (1985).

<sup>14</sup>F. Stern, *Phys. Rev. B* **5**, 4891 (1972).

<sup>15</sup>L. H. Dubois and G. P. Schwartz, *Phys. Rev. B* **40**, 8336 (1989).

<sup>16</sup>H. Kato, M. Yokozawa, R. Kohara, Y. Okabayashi, and S. Takayanagi, *Solid State Electron.* **12**, 137 (1969).

<sup>17</sup>C. Tejedor and F. Flores, *J. Phys. C* **11**, L19 (1978).

<sup>18</sup>J. Tersoff, *Phys. Rev. B* **32**, 6968 (1985).