Fermi-level pinning on ideally terminated InP(110) surfaces

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The deposition of one monolayer of Sb followed by annealing of about 300 °C gives nearly the flat-band condition for both *n*- and *p*-type InP(110). Schottky barriers were formed on these surfaces by using nonreactive metals, Au and Ag, and the surface-Fermi-level positions measured by photoemission spectroscopy converge at about 0.52 eV above the valence-band maximum (VBM) for both *n*- and *p*-type InP. A defect model using a donor level due to phosphorus deficiency at 0.55 eV (±0.05 eV) above the VBM is proposed as a most probable candidate responsible for this Schottky-barrier formation. However, it is also noted that the Sb interlayer could shift the metal-induced gap state's pinning position.

I. INTRODUCTION

One of the challenging problems in surface physics is determining a way to ideally terminate the surfaces of compound semiconductors. For Si this is done by SiO_2, but to date no similar termination has been found for the compound semiconductors despite enormous amounts of research. Perhaps the closest we have come is with the Sb termination of III-V compounds such as GaAs and InP. Many researchers have shown that on the (110) faces of these materials, a monolayer (ML) of Sb is epitaxial and satisfies all III-V surface and Sb bonds. This is very different from the termination of Si by a thick layer of SiO_2; however, it is an intriguing possibility.

As for structures of the Sb terminated III-V (110) surfaces, Skeath *et al.*[1] reported possible structures on GaAs(110): the so-called epitaxial continued-layer structure (ECLS) and p^3 structure with zigzag chains. As for InP(110), Duke *et al.*[2] studied the atomic geometry of the Sb(1 ML)/InP(110) interfaces using elastic low-energy electron diffraction (LEED). The LEED pattern revealed that the surface keeps the same $p(1\times1)$ pattern as the clean cleaved InP(110) surface. They suggested that the two Sb species in the saturated monolayer are located at sites analogous to those which would be occupied by In and P at an unreconstracted InP(110) surface such as the ECLS. We observed the same $p(1\times1)$ pattern as Duke *et al.* in this experiment. The ECLS was confirmed by theoretical total-energy model predictions by Mailhiot, Duke, and Chadi[3] and by comparison of the predictions of this structure with angle-resolved photoemission by Maani, McKinley, and Williams[4] and inverse photoemission by Drube and Himpsel.[5] Recently, LaFemina, Duke, and Mailhiot[6] proposed another possible structure called the epitaxial on-top structure (EOTS). However, a recent development in x-ray standing-wave experiments by Kendelewicz *et al.*[7] suggests that the ECLS is much more probable than the EOTS. Figure 1 shows InP(110) so terminated with a monolayer of Sb.

The thrust of this paper is the use of such Sb termination to attack a very old problem, the production of higher barrier heights on *n*-type InP and Ohmic contacts on *p*-type InP. Typically, metal contacts on InP produce a Fermi level pinned at the metal-InP interface inconveniently close to the conduction-band minimum (CBM). We will report experiments designed to use Sb termination to move this pinning position toward the valence-band maximum (VBM). We will also examine possible physical models in attempting to explain the results reported.

In addition to defects, Fermi-level pinning can be effected by so-called metal-induced gap states (MIGS's).[8] Such states are induced in Schottky barriers by the tunneling of electrons from the metal into the semiconductor.[9]

By imposing a thin nonmetallic layer between the metal and semiconductor, "imaginary" states through which the tunneling takes place can be changed and thus the pinning position due to MIGS's changed.

There is a growing consensus that both defects and MIGS's can be important in determining the Fermi-level pinning position and thus the barrier height at the Schottky barrier.[10]

In this paper we report the termination of an InP(110) surface with Sb, subsequent thermal annealing to acquire almost flat-band conditions on both *n*- and *p*-type InP, and deposition of certain metals to give a new surface Fermi-level position almost 0.8 eV below the CBM. If these results can be translated into practical device tech-

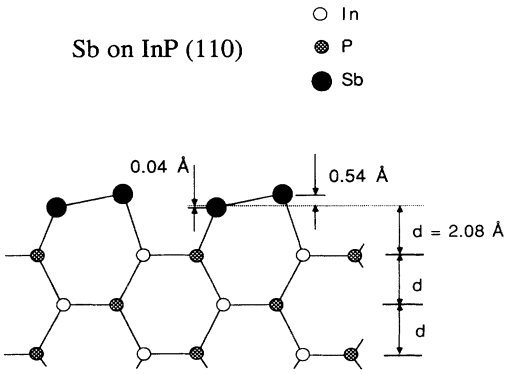

FIG. 1. Suggested structure of Sb-passivated InP(110) interfaces using x-ray standing-wave experiments by Kendelewicz *et al.* (Ref. 7).

nology, they can have a major effect on InP and related technologies by providing sufficiently larger barrier heights on n-type InP to allow for field-effect transistor (FET) gate technologies to be used. Conversely, moving the Fermi-level pinning position closer to the VBM should make it easier to form low-resistance electric contacts on p-type InP and thus contribute to the solution of a major problem.

A critical way of judging the ideality of an interface is Fermi-level pinning. If sufficient interface states are present, the Fermi level at the interface is "pinned" at a position different from that away from the surface. When Sb is placed on (110) surfaces, the Fermi level becomes pinned away from the bulk position, indicating that interface states have been created. However, annealing to around 300 °C can reduce the number of interface states created so that the Fermi level on both n- and p-type samples moves to almost the bulk position.[11]

To our knowledge this is as close as anyone has obtained with the termination of a III-V compound to the ideal case of SiO_2-terminated Si.

Furthermore, we have found that when nonreactive metals (e.g., Au or Ag) are then deposited on Sb-terminated InP, Schottky-barrier heights, which are quite different from those on unpassivated surfaces, can be obtained. For example, Au/n-type InP interfaces which have a Schottky-barrier height of about 0.5 eV when Sb interlayers are used yield barriers as high as 0.82 eV.[12] Therefore, this approach using Sb interlayers opens avenues for engineering Schottky barriers and fundamentally understanding them.

As for a method reducing surface-state defect densities other than this, we would like to pay attention to recent progress in the passivation of III-V semiconductors by group-VI atoms. Sandroff *et al.*[13] indicated that a simple chemical treatment using aqueous solutions of sulfur compounds can reduce surface-state densities considerably. However, Spindt *et al.*[14] and Hasegawa *et al.*[15] found that the surface Fermi level of the treated sample is still pinned at the near midgap, contrary to everyone's expectations. Recently, Chambers and Sundaram[16] reported that Se-passivated GaAs(001) surfaces yield impressive low band bending with approximately 0.2 eV. They explained this low surface-state density assuming that Se undergoes diffusion-limited anion exchange down to a depth of five to seven anion layers below the surface, thereby creating a $Se_xAs_{1-x}Ga$/GaAs heterojunction region that possesses a very low interface-state density. As compared with these passivated surfaces with VI atoms, Sb-passivated surfaces still seem to be much closer to the ideal termination of III-V semiconductors.

II. EXPERIMENT

Photoemission experiments were performed with a monochromated He discharge lamp (He II, $h_\nu = 40.8$ eV). A standard ultrahigh-vacuum chamber (base pressure of 1×10^{-10} Torr) equipped with a double-pass cylindrical mirror analyzer was used. Atomically clean (110) surfaces were prepared by cleaving InP crystals in high vacuum. Crystals used were n-type (Sn doped at $9.3\times10^{17}/cm^3$) and p-type (Zn doped at $2\times10^{18}/cm^3$). Metals were evaporated from tungsten coils. The coverage was measured with a quartz-crystal monitor, and one monolayer was defined as the surface density of atoms on the (110) face of InP (8.2×10^{14} atoms/cm^2). In order to form a one-monolayer passivation layer of Sb, we first deposited 2.0 ML Sb (the same as for a quartz oscillator monolayer) and then annealed at 300 °C for 10 min to remove the excess Sb coverage and anneal the interface. To make Schottky barriers, metals (Au and Ag) were deposited at room temperature. Other metals (Pd, Cu, and Al) were also deposited which reacted strongly with InP through the Sb differing with Au and Ag. These results will be discussed briefly. In order to measure surface-Fermi-level positions, all photoemission spectra were analyzed using a computer curve-fitting routine in order to deconvolve surface, bulk, and reacted components. The surface-Fermi-level shifts were obtained from shifts in the bulk component. The accuracy of this method in determining the surface-Fermi-level position is ±0.05 eV.

III. RESULTS

Figure 2 shows the Fermi-level shifts at Sb/InP(110) interfaces as a function of Sb and subsequent thermal annealing temperature. Although the deposition of two monolayers of Sb results in a Fermi-level position about 0.35 eV below the CBM for n-type InP and 0.85 eV above the VBM for p-type InP, annealing moves the Fermi level to nearly the flat-band condition for both n- and p-type InP. The residual band bending is less than 0.2 eV for both n- and p-type InP. As shown in Fig. 2, at 200 °C, Fermi-level shifts begin to saturate and remain constant from 250 °C up to 350 °C.

The deposition of Au or Ag on these InP(110) surfaces has no effect on the In 4d line shapes, except for the reduction of the In 4d emission intensity. Actually, all spectra are well fitted with the same Gaussian width of

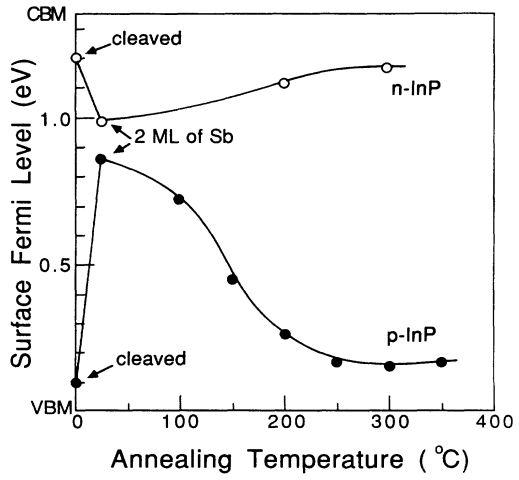

FIG. 2. Surface-Fermi-level positions as a function of annealing temperature at Sb/InP interfaces, showing that the interface gives nearly the flat-band condition after annealing above 250 °C.

0.43 eV ($<\pm 0.02$ eV) as the clean cleaved surfaces.[11,12] For convenience, a series of photoemission spectra of the In 4*d* core level at Au/Sb (1 ML)/InP(110) and Ag/Sb/InP(110) interfaces is shown in Figs. 3 and 4, respectively. Dumas *et al.*[17] reported similar results, suggesting increased stability of Ag/Sb/InP interfaces from Auger studies. Figure 5 shows that the In 4*d* core-level emission intensity decreases very slowly, especially with Ag deposition. Since the photoelectron escape depth is so short (5–6 Å),[18] this indicates that clusters are formed on these surfaces.

The Fermi-level shifts plotted as a function of metal coverage for both *n*- and *p*-type InP(110) are shown in Fig. 6. We find that (1) the Fermi level for *p*-type InP strongly converges at about 0.52 eV above the VBM, starting from the early stage of the Schottky-barrier formation, irrespective of the metal work function [Ag, 4.6 eV; Au, 5.1 eV (Ref. 19)]; (2) the Fermi level at Au/Sb/*n*-type InP interfaces slowly moves toward the same energy level as *p*-type InP; and (3) the Fermi level at Ag/Sb/*n*-type InP interfaces does not saturate even at 10 ML, but shifts very gradually and still seems to be moving at 10 ML, the highest coverage used.

We would like to recall that the slow movement of the Fermi level often suggests that there might be some clustering-induced effects.[20] Since the surface is not still completely covered by metals, one monitors by photoelectron spectroscopy both covered and uncovered areas simultaneously. For this reason the Fermi level at the Ag/Sb/*n*-type InP interface does not saturate even at 10 ML. Fundamentally, the sum of the barrier heights on any *n*- and *p*-type semiconductors must equal to the band gap. Therefore, if the Fermi-level positions on *n*- and *p*-type semiconductors do not converge at the same level, this also means usually that there is a clustering-induced effect. Miyano *et al.*[21] suggested that the band bending measured on clustered systems is interpreted in terms of intercluster surface states and that, when the overall surface potential approaches the value beneath the clusters, one can obtain the exact surface Fermi level. It is also true that separations between *n*- and *p*-type Fermi-level positions cannot be maintained at the metallized surface as suggested by Zur, McGill, and Smith.[22]

From these photoemission studies, we conclude that the Fermi-level pinning position can be moved to about 0.55 eV ($\pm$ 0.05 eV) above the VBM for Au on both *n*- and *p*-type InP. We also expect that Ag will bring the surface Fermi level on *n*-type InP to the same position on *p*-type InP. For *n*-type InP, however, further experiments are necessary to see if it certainly comes to the same position. From the present data, Ag is found to be approaching 0.55 eV ($\pm$ 0.05 eV), but is 0.1–0.2 eV away at the highest coverage (10 ML) used.

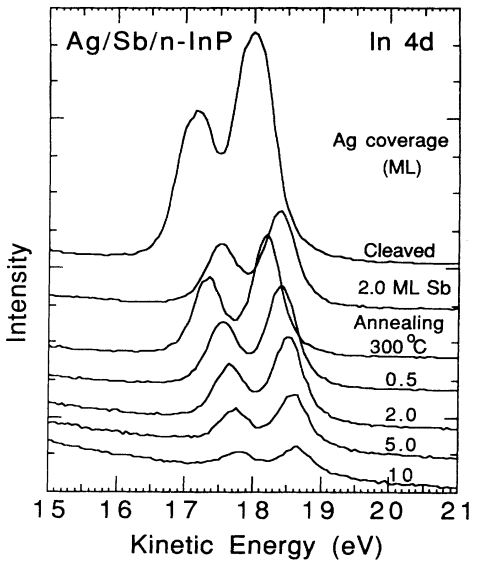

FIG. 4. Series of photoemission spectra of the In 4*d* core level at Ag/*n*-type InP(110) interfaces with Sb interlayers.

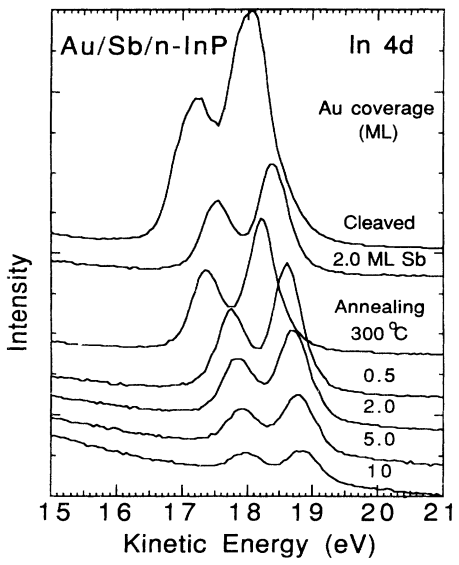

FIG. 3. Series of photoemission spectra of the In 4*d* core level at Au/*n*-type InP(110) interfaces with Sb interlayers.

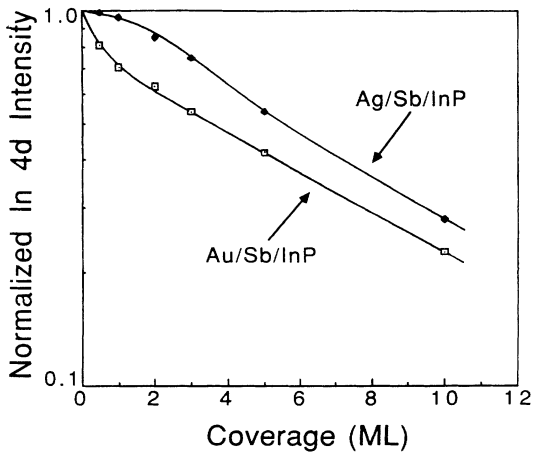

FIG. 5. Attenuation of the In 4*d* core-level emission intensities as a function of metal coverage for Au and Ag.

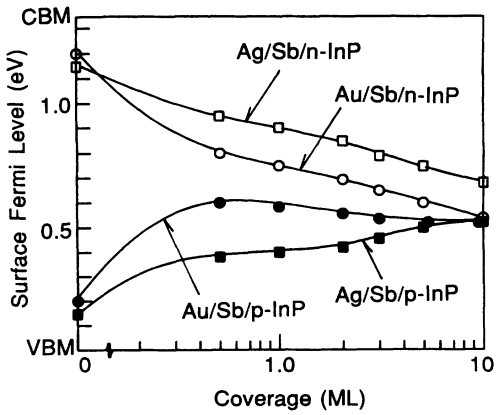

FIG. 6. Surface-Fermi-level positions as a function of metal coverage for both n- and p-type InP passivated with one monolayer of Sb at room temperature, showing that the surface Fermi levels for p-type InP strongly converge at 0.52 eV above the VBM and those for n-type InP slowly converge toward the same level as p-type InP.

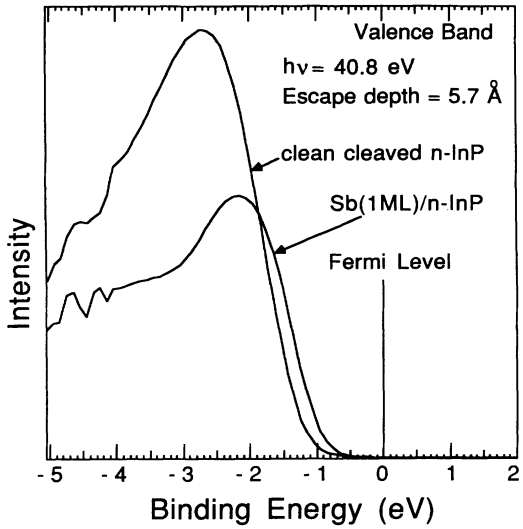

FIG. 7. Photoemission spectra of around the valence-band maximum for a clean cleaved n-type InP(110) surface and that passivated with one monolayer of Sb, showing that the Sb interlayer is not metallic.

IV. MECHANISM FOR SCHOTTKY-BARRIER CHANGE

We will consider three mechanisms for Schottky-barrier pinning: defect, MIGS's, and a version of the Schottky model based on Bardeen's concepts. Increasing attention is being given to the possibility that defects and MIGS's may jointly be responsible for Fermi-level pinning. Mönch[23] suggested this, and Cao *et al.*[24] have argued that a wide range of data can be best understood in this way. von Schilfgaarde and Newman[25] have recently presented theoretical results which reinforce these arguments.

Let us first examine the Sb-terminated surface. In principle, the Sb monolayer produces a surface in which all chemical bonds are satisfied. The fact that after annealing almost flat-band conditions can be achieved on both n- and p-type Sb-terminated material is consistent with few dangling-bond states in the gap. The flat-band condition also indicates that the density of defect levels in the gap is insufficient to cause pinning well away from the band edges.

The deposition of a metallic overlayer could produce defects[26] and MIGS's. In order to explain the unpinned nature observed on GaAs(100), Freeouf *et al.*[27] introduced the concept of an insulating layer like surface reconstruction which reduces pinning due to the MIGS's. However, in Sb-terminated cases, instead of saying so simply, we would like to say that the MIGS's would be different from those without the Sb present, because electrons have to tunnel through the forbidden gap of the Sb monolayer. The tunneling through the Sb layer and thus the MIGS will be determined by the electronic structure of that monolayer as well as of the InP. Figure 7 gives photoemission spectroscopic results of the valence-band electronic structure of the system before and after Sb was added. It appears that the valence-band maximum with Sb present has moved to higher kinetic energy by about 0.2 eV. Thus the Sb VBM may be located 0.2 eV higher than the VBM for InP. This experiment can only put a lower limit on the CBM with Sb present. It cannot lie below the Fermi level shown in the diagram. Since the band structure of thick Sb is semimetallic, one would not expect a large band gap for the monolayer of Sb on InP. The conduction-band structure of InP pulls the MIGS position to 0.76 eV above the VBM, a position unusually high in the band gap. If the Sb electronic structure has more strength near the VBM, this would move the MIGS pinning position toward the VBM, as is found in the experiment.

If defects are important, the change in pinning position indicates that they are dominated by different defect energy levels than were present in the absence of the Sb overlayer. For example, without the presence of Sb, the Au pinning position is at 0.95 eV above the VBM, whereas, with Sb, it is pinned near 0.53 eV above the VBM. However, it may be that different defects are present in the two cases. Interestingly, a donor defect near 0.55 eV (±0.05 eV) has been induced by annealing a clean cleaved (110) surface of InP.[11] This defect has been associated with stoichiometric loss of P. Figure 8 shows the spectrum of the In 4d core level at Sb/p-type InP(110) interfaces annealed at 300 °C. In order to get a good curve fitting, a small component is always required on the low-kinetic-energy side of the main bulk component. The main bulk component shows that almost all parts of the interface return to approximately the flat-band condition with a residual band bending of about 0.16 eV by annealing at 300 °C. We have to note that (1) this small component does not appear on the In 4d core-level spectra on n-type InP, and (2) the energy level of this component is 0.55 eV (±0.05 eV) above the VBM. These findings suggest that this small component is a pinned component due to some donor defects which are created locally during annealing Sb/InP(110) interfaces. One would explain the p-type pinning in terms of such a defect. For n-type

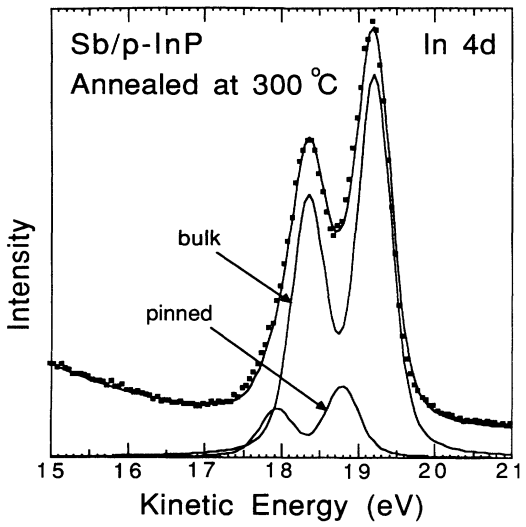

FIG. 8. Photoemission spectrum of the In 4*d* core level at Sb/*p*-type InP(110) interfaces annealed at 300 °C, showing that a small pinned component appears on the low-kinetic-energy side of the main bulk component no matter what the initial cleave quality is.

material one would have to empty the donors to pin near the 0.55-eV level. This would be done by introducing acceptor-type defects below 0.55 eV. Another mechanism would be the transfer of electrons from the semiconductor to the Au as a result of the high electronegativity of the Au.

This latter mechanism can be explained in terms of the Schottky mechanism of charge transfer between the metal and semiconductor. This was first developed by Schottky in terms of differences in work function between metal and semiconductor.[28] However, as Bardeen[29] pointed out, it is better to think of this in terms of a difference in absolute potential energies in the metal and semiconductor. In this way confusion due to surface dipoles, which are inherent to vacuum work functions but not metal semiconductor contacts, do not confuse the problem.[30] The absolute potentials are related to the electronegativity. Electrons will flow from a material of low to one of high electronegativity. Newman *et al.*[31] suggested that metals with electronegativity higher than 1.9 (Au, 2.4; Ag, 1.9) have a tendency to pin *n*-type semiconductors at donor levels for both GaAs and InP. Thus one would expect Au (with higher electronegativity than Ag) to pin *n*-type semiconductors more effectively than Ag, as is observed. However, one must be cautious in taking this agreement seriously until corrections are made for the fact that Ag clumps more strongly than Au does; that is, Ag clusters more strongly than Au does.

To summarize, an Sb interlayer may modify the charge neutrality point and may also modify the nature of dominant defects. Both mechanisms as well as a combination of them should be examined in order to explain the shift in pinning position due to the Sb. We identified the dominant defects as donors with phosphorus vacancies as the simplest form of phosphorus loss. Note that this energy level does not coincide with the pinning level of so-called P-deficient metal interfaces (e.g., noble metals) suggested by Brillson *et al.*[32] However, we have recently shown that the surface Fermi level at the Pd/InP barrier which is formed on InP(110) with intentionally modified surface stoichiometry by vacuum annealing moves toward 0.6 eV above the VBM from the common level of about 0.9 eV above the VBM with increasing phosphorus loss.[33] Stair and Chung[34] also reported that the surface Fermi level has a tendency to move toward the CBM with increasing phosphorus. These findings suggest that defects responsible for the Fermi-level pinning of so-called P-deficient metal interfaces are structurally different with P-deficient defects which are thermally introduced at Sb-passivated surfaces.

Let us briefly introduce Fermi-level pinning for reactive metals on Sb-passivated surfaces (e.g., Pd, Cu, and In). In this case the surface Fermi level ranges from 0.6 to 0.9 eV above the VBM, depending on metal electronegativity, and the separations between *n*- and *p*-type surface-Fermi-level positions are smaller than 0.05 eV. These findings suggest that (1) in reactive cases the clustering-induced effect is smaller than nonreactive cases (especially smaller than Ag) and (2) other defects at around 0.9 eV above the VBM may be created by chemical reactions which destroy interfacial perfection. Accordingly, this Fermi-level pinning would be best explained by the defect model using the simplest defect model with only two defect levels dominant (i.e., a donor level at about 0.55 eV and an acceptor level at about 0.9 eV above the VBM).

V. CONCLUSIONS

On the basis of these experimental studies, we summarize as follows: In order to control the Fermi level of InP(110), we have proposed a method using one monolayer of Sb as an interlayer of metal-semiconductor interfaces. The deposition of one monolayer of Sb followed by annealing about 300 °C gives a nearly flat-band condition for both *n*- and *p*-type InP. Au and Ag are nonreactive on these surfaces and show strong clustering. The Fermi-level positions for *p*-type InP measured by photoemission spectroscopy strongly converge at about 0.52 eV above the VBM, and those for *n*-type InP slowly converge toward the same level as *p*-type InP. The MIGS's modified by Sb is proposed as a possible candidate responsible for Schottky-barrier formation on ideally terminated surfaces. However, a defect model involving a donor level at about 0.55 eV ($\pm$ eV) above the VBM is proposed. This is attractive because such a level in this energy range has been independently produced by annealing InP.

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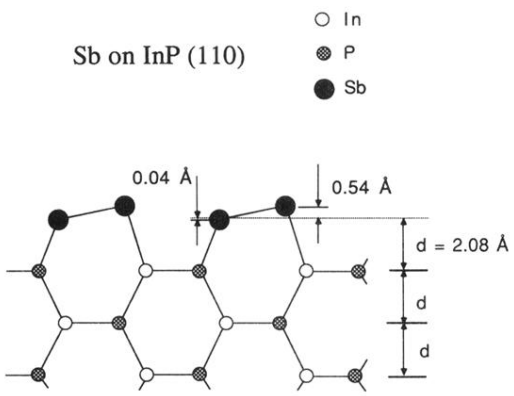

FIG. 1. Suggested structure of Sb-passivated InP(110) interfaces using x-ray standing-wave experiments by Kendelewicz *et al.* (Ref. 7).