

## Resonant inverse-photoemission study of layer-dependent surface states at the epitaxial GaAs(110)-Bi interface

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The unoccupied electronic structure of the ordered GaAs(110) $1\times 1$ -Bi system has been studied with use of inverse photoemission. In the Bi-coverage range from 1 to 2 monolayers, two unoccupied electronic surface states are observed, 0.9 and 1.9 eV above the valence-band maximum of GaAs at  $\Gamma$ . From their coverage-dependent intensities, they are assigned to the outer (Bi-Bi) and inner [Bi-GaAs(110)] interfacial layers, respectively. The states are characteristic of the bilayer and vanish for thicker, bulklike Bi films. A resonant enhancement of these two states, and an additional image state, is observed when the energy of the emitted photon coincides with the plasmon energy.

### I. INTRODUCTION

Few of the GaAs(110)-metal systems that have been studied so far are well-behaved epitaxial monolayer systems. The notable exception is Sb. It does not react disruptively with the GaAs(110) surface<sup>1</sup> and it is also fairly well lattice matched.<sup>2-10</sup> Although the atomic radius of Bi is significantly larger than that of Sb ( $\approx 9\%$ ), it has been shown recently<sup>11</sup> that Bi also forms an ordered ( $1\times 1$ ) overlayer on GaAs(110). However, images that were collected using the scanning tunneling microscope (STM) (Ref. 11) clearly show that near one monolayer (ML) there are ordered arrays of vacancies in the Bi overlayer. It is likely that these Bi vacancies relieve the strain in the ordered Bi terraces, thereby compensating for the disparity in the lattice constants. The same study identified four Bi-induced states near the Fermi level, two of which were occupied and two of which were unoccupied. In keeping with previous theoretical treatments of the GaAs(110)-Sb monolayer system<sup>9,10</sup> and a previous experimental study of the GaAs(110)-Bi monolayer system,<sup>11</sup> these states have been labeled for convenience  $S_5$ ,  $S_6$  (occupied),  $S_7$ , and  $S_8$  (unoccupied), respectively (see Fig. 2 of Ref. 11).

In this paper we present the results of an inverse-photoemission study of the unoccupied electronic states of the GaAs(110)-Bi system (see also the recent inverse-photoemission study of this system performed by Hu *et al.*<sup>12</sup>). In agreement with the STM spectroscopy of Feenstra,<sup>11,13</sup> we have found that there are two unoccupied, Bi-induced states above the Fermi level at adlayer coverages near 1 ML. In addition, we demonstrate that the lower of the two states ( $S_7$ ) is not detectable at coverages of 0.5 ML. Moreover, the intensity of this state continues to grow above coverages of 1 ML and its intensity saturates between 1.5 and 2 ML. This behavior allows us to make some predictions about the interfacial position of the two states.

A large enhancement of the photon intensity has recently been observed from thin Sb films ( $\approx 20$  Å), on a variety of substrates,<sup>14</sup> when the emitted photon energy

equals the plasmon energy. This was identified<sup>14</sup> as a resonance between a discrete plasmon decay channel and the inverse-photoemission continuum. In subsequent studies, other systems were found (e.g., Refs. 15 and 16) which exhibited similar behavior. In this paper we demonstrate that the unoccupied electronic states at the GaAs(110)-Bi interface also display resonant behavior. Moreover, this study of a structurally characterized, epitaxial system sheds some light on the mechanism underlying the plasmon resonance, which has remained a puzzle primarily because it does not occur on bulk crystals of the classical free-electron metals such as Al.<sup>17</sup> For Bi overlayers on GaAs(110), it appears that the plasmon resonance is strictly a thin-film effect, as it weakens when the film thickness is increased beyond two layers. Photoemission studies have demonstrated (see later) that this is the thickness at which the growth of bulklike Bi islands begins. Therefore it is possible that the thin film contains new plasmon modes, not present in bulk Bi, which couple to light.

### II. EXPERIMENT

The GaAs(110) substrates were prepared by cleaving notched and aligned GaAs bars, using the wedge-and-anvil technique, in an ultrahigh vacuum preparation chamber adjoining the analysis chamber. The Bi overlayers were evaporated from a water-cooled boron nitride effusion cell. The quality of the surface was determined from the inverse-photoemission spectra of the clean surface and from visual inspection. Both *n*- and *p*-type GaAs bars were used; however, the results presented here were obtained from *p*-type bars. Unpinned *p*-type surfaces, where the initial band-bending was less than 0.1 eV, were fairly easy to produce. In contrast, inverse photoemission from *n*-type surfaces indicated the presence of a substantial voltage drop between the surface and the back contacts during measurement. Therefore, the quality of the Ohmic contacts to the GaAs bar was checked both before and after the experiment. In each case the resistance between the two alloyed In-Ga contacts was

close to the expected value, based on estimates of the GaAs bulk resistivity, suggesting that the back contacts were not the source of the voltage drop. This indicated that the process of collecting the inverse-photoemission spectrum produces an additional dipole at the *n*-type GaAs(110) surface. For example, a buildup of electrons in surface defect states could produce an additional band-bending shift in the direction that we observed. Although we did not study this phenomenon in detail, we note that band-bending shifts have recently been observed on cleaved GaAs(110) surfaces with low-energy electron beams, He II (40.8 eV) and Ne II (27 eV) (Ref. 18) (see also Refs. 19–21).

### III. COVERAGE DEPENDENCE OF ELECTRONIC STRUCTURE

Inverse-photoemission spectra were collected from the clean GaAs(110) surface and from the same surface covered with various amounts of Bi: 0.1, 0.5, 1.0, 2.0, 3.0, and 10 ML. One monolayer is defined to be the coverage at which the atomic surface density of the Bi adlayer equals the atomic surface density of the GaAs(110) surface ( $8.85 \times 10^{14} \text{ cm}^{-2} \approx 3 \text{ \AA Bi}$ ), assuming that the sticking coefficient of the sample is the same as that of the film thickness monitor and that layer-by-layer adlayer growth occurs.

The coverage dependence of the unoccupied electronic structure is illustrated in both Figs. 1 and 2 and quantified in Fig. 3. In Fig. 1 the incident electron energy is 14.7 eV and in Fig. 2 the same coverage range has been studied using an incident electron energy of 11.7 eV relative to the valence-band maximum. For reasons that will be discussed more fully in Sec. IV, the intensity of  $S_7$  is resonantly enhanced by lowering the energy of the incoming electrons from 14.7 to 11.7 eV. At Bi coverages of 1 ML,  $S_7$  now appears as a well-resolved state rather than as an unresolved shoulder. Figure 1 also demonstrates quite clearly that for Bi coverages as low as 0.5 ML there is a substantial amount of extra emission in the GaAs band gap (the band bending has been subtracted from the spectra). This extra emission ( $S_8$ ) is situated approximately 1.9 eV above the valence-band maximum. The intensity of this feature saturates at 1 ML, and decreases above 1 ML, suggesting that the state is produced by Bi-substrate bonding. Above 0.5 ML another state appears lower in the GaAs gap, approximately 0.9 eV above the valence-band maximum. The coverage dependence of this state is quite different. For example, the state appears above 0.5 ML and its intensity continues to grow beyond 1 ML, saturating between 1.5 and 2 ML. Above 2 ML (not shown) the overlayer becomes semimetallic, displaying relatively weak emission near the Fermi level. This suggests that the Bi—Bi bonds that are formed above 2 ML are different in nature from those formed below 2 ML. This change is probably due to a structural transformation from the ordered (1×1) chainlike structure to the hexagonal structure of bulk Bi. This conclusion is supported by core-level photoemission studies, which will be discussed later, that suggest that the Bi overlayer grows in a layer-by-layer fashion only up to 2 ML.

The coverage dependence of these states suggests that although the higher of the two states ( $S_8$ ) is produced by Bi—GaAs(110) bonding, the lower state ( $S_7$ ) is related to Bi—Bi bond formation during the second-layer Bi growth. The upper of the two states is therefore analogous to the state produced by Sb bonding to the (110) surface of GaP, GaAs, and InP.<sup>9,10,22,23</sup> However these suggestions have to be verified by comparing our results with the results of other independent studies.

STM studies of Bi growth on GaAs(110) have demonstrated<sup>11,13,24</sup> that Bi forms ordered epitaxial terraces on GaAs(110). The Bi terraces nucleate in a random manner and grow laterally in size as the coverage increased. However there is a lattice mismatch induced limit for growth along [110]. The Bi terraces grow, along this direction, until they are approximately 25 Å long. Subsequent growth is along [001]. At coverages near 1 ML there are lines of Bi vacancies intersecting the ordered

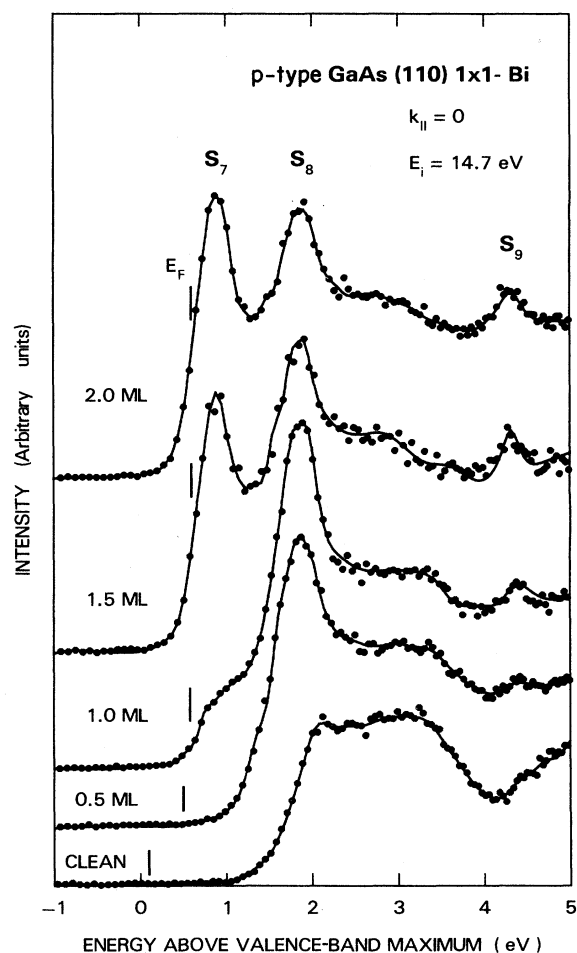


FIG. 1. Inverse-photoemission spectra from clean GaAs(110) and from the same surface covered with various Bi coverages. Three Bi-induced surface states are seen, which are assigned to the first Bi layer ( $S_8$ ), to the second Bi layer ( $S_7$ ), and to an image state at the Bi surface ( $S_9$ ). The energy of the incident electrons is 14.7 eV above the valence-band maximum.

terraces along [001].

Scanning tunneling spectroscopy has also been performed on this system at adlayer coverages of both 0.5 and 0.8 ML.<sup>11,13,24</sup> One powerful feature of the STM is its ability to spatially resolve details of the electronic structure. For example, it was shown that the electronic structure of the ordered Bi terrace is different from that of the terrace edge (e.g., Fig. 7 of Ref. 24). Above the ordered terrace two unoccupied states are seen, a peak  $S_8$  and a shoulder  $S_7$ . The energetic position of  $S_8$  is in excellent agreement with the results of this study, since both studies position  $S_8$  at 1.9 eV above the valence-band maximum. The shoulder  $S_7$  is found at 1.3 eV above the valence-band maximum in STM, whereas in our study it is positioned at 0.9 eV. This difference could be due to the low coverage in the STM data, which leaves only a weak trace of the second-layer state  $S_7$ , or due to averaging over different momenta in STM.

In order to compare our results with the inverse-photoemission study of Ref. 12 it is necessary to convert the energy scale from the valence-band maximum to the Fermi level. This is nontrivial since the Fermi-level pinning position in the gap of GaAs does vary with doping,

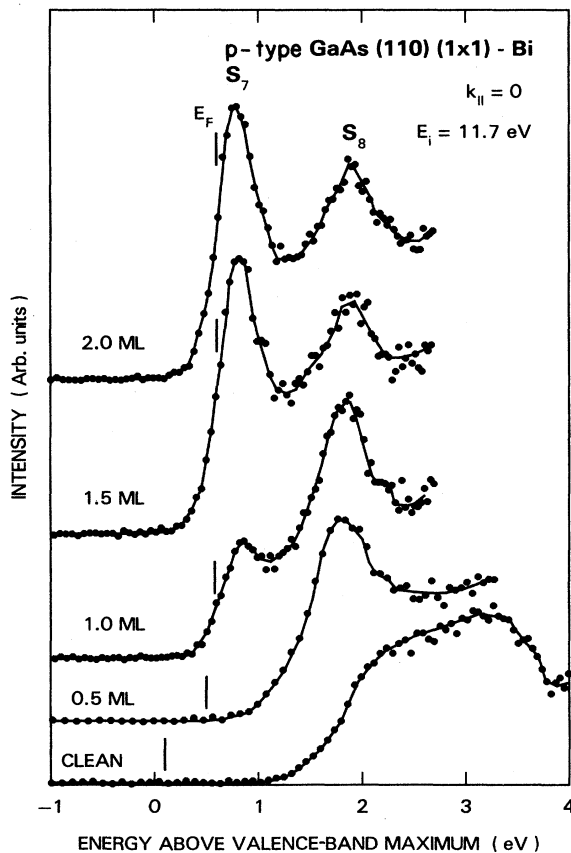


FIG. 2. As for Fig. 1, but with an incident energy of 11.7 eV above the valence-band maximum. Lowering the energy of the incident electrons resonantly enhances the intensity of state  $S_7$  relative to that of state  $S_8$  (see Fig. 4).

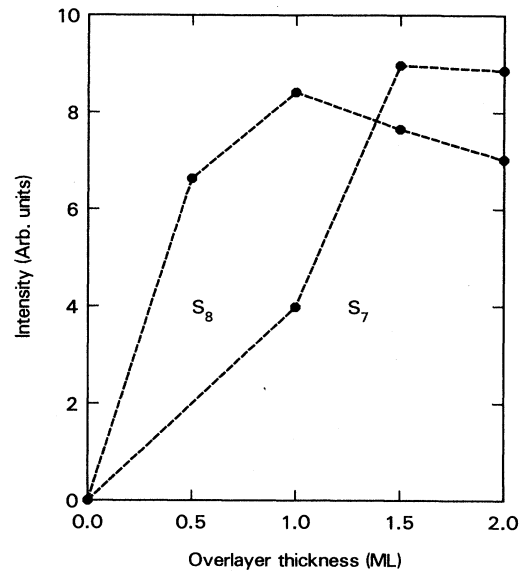


FIG. 3. Intensity of states  $S_7$  and  $S_8$  plotted against Bi coverage. The intensity of state  $S_7$  saturates near 2 ML whereas the intensity of state  $S_8$  attains a maximum at approximately 1 ML, suggesting that  $S_7$  is due to the second Bi layer and  $S_8$  to the first Bi layer. The incident electron energy is 11.7 eV above the valence-band maximum (compare with Fig. 2).

even at 2 ML coverage. We find the Fermi level 0.2 eV higher in the gap for  $n$ -type GaAs than for  $p$ -type at 2 ML. This is consistent with a previous study,<sup>12</sup> which found the Fermi level 0.4 and 0.6 eV above the valence-band maximum for 1 ML of Bi on  $n$ - and  $p$ -type materials, respectively. Using the pinning position for  $p$ -type samples we obtain good agreement in the energy position of the two surface states  $S_7$  and  $S_8$  for the two inverse-photoemission studies.

We now turn to the additional unoccupied state that is detected at the edge of the Bi terraces and above the Bi vacancies, at overlayer coverages near 1 ML, with the STM.<sup>11,13,24</sup> A similar state was observed in the GaAs band gap at the GaAs(110)-Sb and GaAs(110)-Au systems.<sup>24</sup> It has been suggested that the origins of these states may be the same.<sup>24</sup> However, the additional "terrace-edge" state is not obvious in the inverse-photoemission spectra. This is probably due to the low concentration of such defects (less than 10% of a monolayer).

Two independent core-level photoemission studies of the GaAs(110)-Bi interface have been performed.<sup>24,25</sup> Both performed a least-squares analysis of the Ga  $3d$ , As  $3d$ , and Bi  $5d$  core levels at various Bi coverages. Joyce *et al.*<sup>25</sup> performed a least-squares analysis of the Bi  $5d$  core level, using two spin-orbit split components of equal intensity at Bi coverages of 0.3 ML and three spin-orbit split components for 1 ML and above. The two spin-orbit split components of equal intensity that were observed at low coverage were identified with Bi—As and Bi—Ga bonds, whereas the third component was attri-

buted to Bi—Bi bond formation at higher coverages (1 ML). The coverage dependence of these features is similar to the coverage dependence of  $S_7$  and  $S_8$ . For example, although  $S_8$  and the Bi—GaAs(110) features are present below 0.5 ML,  $S_7$  and the Bi—Bi feature in the Bi  $5d$  core level only appear above 0.5 ML.

The other photoemission study of the GaAs(110)-Bi interface<sup>24</sup> fitted the Bi  $5d$  core level with two spin-orbit split components of equal intensity at Bi coverages of 1 and 3 Å. These were attributed to Bi—As and Bi—Ga bonds. There was no component corresponding to the Bi—Bi bond at coverages of 1 or 3 Å, and at coverages of 9 Å the total line shape was modeled again, ignoring the weak high binding-energy component, using two spin-orbit split components. However this time they were of different magnitude. The latter choice of trial functions initially appears to be the less physical of the two options as it does not provide a unified description of the entire coverage range. For example, at coverages of 9 Å and above there is a distinct component corresponding to the Bi—As bond but the Bi—Ga component is “lumped in” with the Bi—Bi feature. However, the two-component fit is clearly another viable candidate for the higher coverages, and it may be argued that it represents the simplest description of the total core-level line shape.

At this stage, it is difficult to make a detailed comparison between the results of our inverse-photoemission study and the two available core-level photoemission studies. This is partly due to the fact that the uniqueness of the trial functions which were used for the core-level analysis has not been unambiguously determined. This can be done by careful residual analysis,<sup>26</sup> and it would be helpful if this was performed. Nevertheless, we can say that there is a close correlation between the growth of  $S_7$  and the Bi—Bi feature in the Bi  $5d$  core level,<sup>24</sup> suggesting that they may be related.

#### IV. RESONANT INVERSE PHOTOEMISSION

This study of the GaAs(110)-Bi system is a natural extension of the earlier resonant inverse-photoemission studies which were undertaken using Sb overlayers on InP(110) and W(100) substrates.<sup>14</sup> For the GaAs(110)-Bi system we have the added advantage that the morphology of the films is fairly well characterized.<sup>11,13,24,25</sup> Bi is also a natural material to use for resonant inverse-photoemission studies as it has a well-defined bulk plasmon.<sup>27</sup> Due to the fact that Bi has a larger lattice constant than Sb, and consequently a lower electron density, the plasmon energy in Bi (14.7 eV) is lower than that of Sb (15.9 eV). The surface-plasmon energy of Bi is 9.9 eV,<sup>27</sup> significantly lower than the photon energies considered here.

We find that all three Bi-induced states are resonantly enhanced at an energy of 12–13 eV for the emitted photons (Fig. 4). This energy corresponds to the plasmon energy in the thin Bi film, as can be seen from the non-resonant plasmon decay that takes place when the energy of the incident electrons is raised well above the resonance energy (Fig. 4, bottom curve). All these results are completely analogous to the Sb data.<sup>14</sup>

The fact that the resonance in Bi is much less dramatic than the factor of 30 found<sup>14</sup> for Sb appears to be puzzling at first glance. However, this difference is a consequence of the different film thicknesses (6 Å for Bi, 20 Å for Sb). For Sb films, the resonance increased with film thickness until a spontaneous phase transition occurred (see below). A similar increase is seen for Bi, going from one to two layers, but the phase transition occurs near two layers, thereby limiting the magnitude of the resonance. Sb and Bi films of the same thickness exhibit comparable resonance strength.

These findings provide a clue for possible mechanisms of the plasmon resonance. Apparently, the structure of the film plays an important role. For the Sb films, the resonance effect was correlated with a metastable phase,

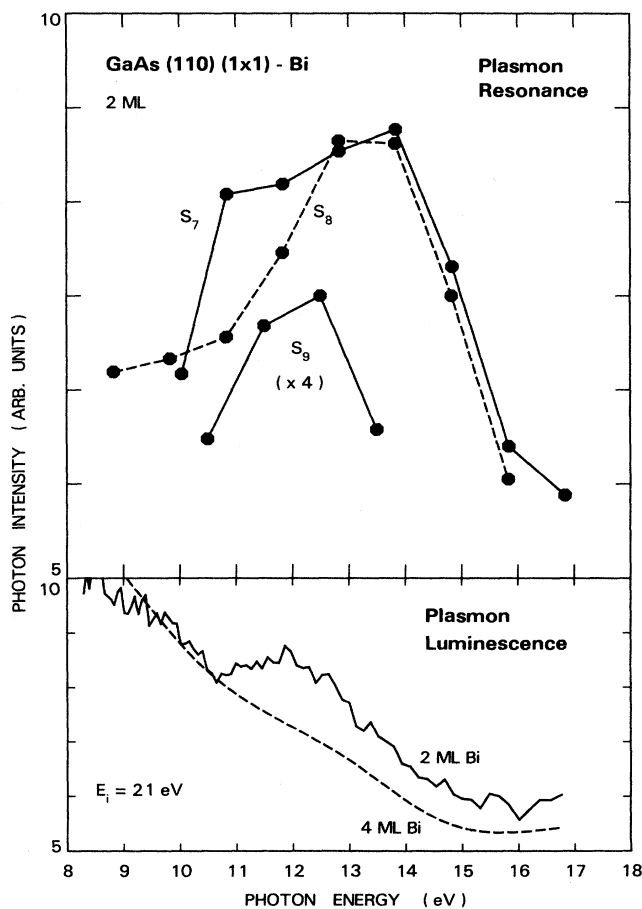


FIG. 4. Intensity of the Bi-induced states  $S_7$ – $S_9$  for 2-ML Bi on GaAs(110), plotted against the outgoing photon energy, obtained from various incident electron kinetic energies. All three states undergo a resonance when the photon energy matches the Bi plasmon energy at 12–13 eV. The latter can be inferred from the nonresonant plasmon decay shown in the bottom curve (taken with an high initial energy of 21 eV). The dashed background curve is for a thicker Bi film, where the resonance is absent (see text).

which exhibited a squared low-energy electron diffraction (LEED) pattern, both on InP(110) and W(100) substrates.<sup>14</sup> The onset of bulklike, hexagonal LEED patterns at larger film thickness caused the resonance to disappear, and it was barely visible for a Sb single-crystal surface. The conversion to the stable bulklike phase could be delayed for Sb up to a thickness of 20 Å by careful annealing. The temperature had to be high enough to go from the amorphous room-temperature deposit to the metastable structure with the square LEED pattern. On the other hand, when the sample was annealed too high, it converted to the stable bulk phase with a hexagonal LEED pattern. The bulk melting temperature of Bi is lower than that of Sb and consequently Bi may not stay in such a metastable structure at room temperature. In fact, layer-by-layer growth has been found to cease in Bi after two layers,<sup>25</sup> and this is the point where the resonance starts weakening.

Apparently, it is necessary to have a continuous thin film in order to produce the plasmon resonance. Extra plasmon modes in the thin film may couple to light. This is not possible for the bulk-plasmon mode in a half-infinite crystal. At the bulk-plasmon energy, the dielectric constant vanishes, thereby allowing no electric field in vacuum. Only above the plasmon energy does the radiative decay of bulk plasmons become possible.<sup>17</sup> The existence of special thin-film plasmon modes in the Sb and Bi films is also suggested by the deviations in the observed plasmon energies from the bulk values. For example, in Sb films the plasmon energy varied by almost 1 eV from one substrate to the other, and in the Bi layer we noticed that the state on the inner layer ( $S_8$ ) resonates about 1 eV higher than the states on the outer layer ( $S_7, S_9$ ). This observation solves the seeming contradiction with data from bulk crystals of free-electron materials like Al, where a minimum of the inverse photoemission intensity is seen at the bulk-plasmon energy.<sup>17</sup> It would be interesting to check if thin films of Al exhibited resonance behavior similar to that of Sb and Bi. We have tried to deposit such Al films on Si, but only found a strong enhancement of the inverse-photoemission intensity at the surface-plasmon energy, and not at the bulk-plasmon energy.<sup>28</sup> This is due to a completely different effect. The surface roughness couples the wave vector of surface plasmons to that of light.<sup>17</sup> The roughness implied from the surface-plasmon resonance indicates three-dimensional growth, which precludes a thin-film effect like that of Sb and Bi. The three-dimensional growth in Al is due to its high surface energy. The group-V elements exhibit saturated lone pair orbitals at the surface, making them good candidates for layer-by-layer film growth.

## V. ANNEALING STUDIES

Annealing studies of the GaAs(110)-Bi system were also performed. In contrast to GaAs(110)-Sb, the structural order of the GaAs(110)-Bi system was not significantly improved by heating. Although we did observe that some weak spots did emerge when the overlayer was gently annealed [Fig. 5(a)], indicating that an-

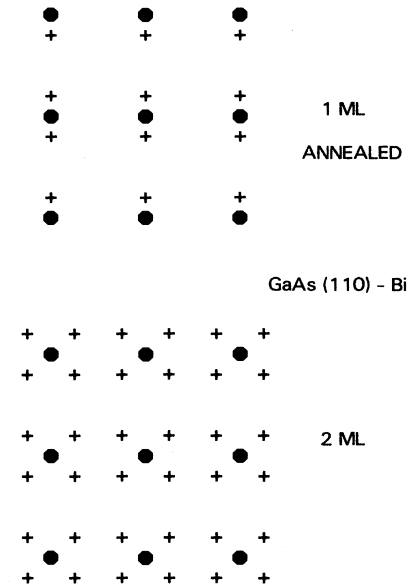


FIG. 5. Schematic LEED patterns for an annealed mono-layer coverage of Bi on GaAs(110) (A, top figure) and for an annealed (or unannealed) 2-ML coverage (B, bottom). The dots represent the  $(1 \times 1)$  pattern of the clean surface which is still visible in both cases. The crosses represent weaker spots. The diagram is not drawn to scale and only the most intense spots are shown.

nealing does have some, if not a large, affect on the ad-layer order. The spots indicated that the overlayer has a superlatticelike structure along the  $[1\bar{1}0]$  direction. This is consistent with the observation that the overlayer contains an ordered array of missing Bi atoms perpendicular to the underlying As-Ga chains. Above approximately 2 ML the LEED pattern changes [Fig. 5(b)] and a diamondlike pattern appears superimposed on the  $(1 \times 1)$  pattern. Between each  $(1 \times 1)$  spot there are now two additional visible spots of much lower intensity.

Moreover, we found that it was possible to anneal both 2 and 3 ML overlayers back to 1 ML and retrieve the 1 ML  $(1 \times 1)$  LEED pattern. However, neither the inverse-photoemission spectra nor our LEED studies indicated that the overlayer order was substantially better than that produced by depositing Bi onto a room-temperature GaAs(110) substrate. Therefore, our LEED studies suggest that, in contrast to Sb, the Bi overlayer attains its minimum-energy configuration at room temperature. This result, in itself, is not surprising since bulk Bi has a lower melting point (271 °C) than Sb (631 °C).

## VI. SUMMARY

We have studied the unoccupied electronic structure of the GaAs(110)-Bi system using inverse photoemission. Two unoccupied electronic states have been identified. The higher of the two states, which we have denoted  $S_8$ , is already present within the GaAs band gap at coverages of 0.5 ML, and it attains its intensity maximum close to the 1-ML coverage. We have suggested that this state is most likely to be produced by Bi—As and the Bi—Ga

substrate bonds of the first (inner) layer. The second Bi-induced state, which we have denoted  $S_7$ , appears at Bi coverages of approximately 0.5 ML and its intensity saturates by coverages of 2 ML. We have argued that this suggests that  $S_7$  may be produced by Bi—Bi bonding. This second-layer state is still different from bulk Bi, which exhibits a semimetallic spectrum with low emission near the Fermi level, where the second layer state resides. We have also found that it was possible to resonantly enhance the intensity of the two Bi-induced states by matching the outgoing-photon energy with the Bi-plasmon energy, thereby shedding new light on the mechanism of these plasmon-induced resonance phenomena.

*Note added in proof.* Recently, Pan *et al.* (Ref. 29) found a plasmon-related resonance in inverse photoemis-

sion from bulk Nb. If this resonance has the same origin as the resonance seen for thin Sb and Bi films, it indicates that the band structure in these metastable thin-film phases is different from that of the bulk phase and crucial to the coupling with plasmons.

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