PHYSICAL REVIEW B VOLUME 40, NUMBER 17 15 DECEMBER 1989-I

Core-level photoemission investigation of atomic-fluorine adsorption on GaAs(110)

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(Received 29 August 1989)

The adsorption of atomic F on the cleaved GaAs(110) surface has been studied with use of high-resolution core-level photoelectron spectroscopy by exposing the GaAs(110) surfaces to XeF_2, which adsorbs dissociatively, leaving atomic F behind. This surface reaction produces two chemically shifted components in the Ga 3*d* core-level emission which are attributed to an interfacial monofluoride and a stable trifluoride reaction product, respectively. The As 3*d* core level develops only one chemically shifted component and from its exposure-dependent behavior it is attributed to an interfacial monofluoride. Least-squares analysis of the core-level line shapes revealed that (i) the F bonds to both the anion and the cation , (ii) the GaF_3 component (characteristic of strong interfacial reaction) and the surface core-level shifted component (characteristic of a well ordered, atomically clean surface) are present together over a relatively large range of XeF_2 exposures, and (iii) it is the initial disruption of the GaAs(110) surface that is the rate-limiting step in this surface reaction. These results are compared with similar studies of Cl and O adsorption on GaAs(110).

I. INTRODUCTION

Although the adsorption of molecular O and molecular Cl on GaAs(110) has been the subject of several experimental studies,[1-7] the adsorption of F on GaAs(110) has not been examined so closely. The first studies of Cl adsorption on GaAs(110) (Ref. 1) indicated that the Cl atom adsorbs preferentially to the surface anions. The evidence for this bonding configuration came mainly from a comparison of valence-band photoemission spectra with calculated tight-binding valence-band density of states. This adsorption geometry subsequently found support from core-level photoemission studies,[3] where it was demonstrated that the line shape of the As 3*d* core level was dramatically modified by Cl adsorption whereas the Ga 3*d* line shape was not. This picture also provides a natural explanation for the (1×1) low-energy electron-diffraction (LEED) patterns that are observed on GaAs(110) surfaces that have been exposed to Cl (e.g., Ref. 3).

The oxidation of GaAs(110) has also been the subject of many experimental studies and the source of considerable debate. In recent years this has centered around whether the oxidation proceeds in a layer-by-layer fashion or whether it involves localized subsurface oxidation (see Refs. 4–7). Core-level photoemission studies alone (i.e., compare Refs. 3 and 4) have demonstrated that there are some striking differences between molecular Cl and molecular O adsorption on cleaved GaAs(110) surfaces.

In this study we have investigated the adsorption of atomic F onto the cleaved (110) surface of GaAs(110) using high-resolution core-level photoelectron spectroscopy with synchrotron radiation. It is demonstrated that the adsorption of atomic F onto the cleaved GaAs(110) surface is distinctly different from either the adsorption of molecular O or molecule Cl on GaAs(110). Furthermore, the study raises some interesting questions about the mechanisms of O and Cl adsorption on GaAs(110).

II. EXPERIMENTAL DETAILS

The GaAs(110) surfaces were produced by cleaving aligned and notched *n*-type and *p*-type GaAs bars in ultrahigh vacuum (2×10^{-10} Torr). Only surfaces that were visually flat and free of cleavage-induced steps were used. No initial band bending was observed on any of the *p*-type (110) surfaces. However, several of the *n*-type (110) cleaves exhibited initial band-bending shifts of the order of 50–100 meV. Since we were primarily interested in the surface chemistry, these surfaces were retained for photoemission studies. For both *n*- and *p*-type samples, the Fermi-level position was referenced to the Fermi edge of a clean metallic standard which was transferred into the analysis chamber and measured under identical conditions.

The GaAs(110) surfaces were dosed with atomic F using XeF_2. Our photoemission studies indicated that, at room temperature, the XeF_2 dissociates completely upon chemisorption leaving only F on the surface. We could not detect any residual Xe features in the photoemission spectra, even after our largest XeF_2 exposure. All of the XeF_2 exposures were performed in a custom-built stainless-steel dosing chamber which has been described in detail elsewhere.[8] The exposures were measured with a nude ion gauge which was out of line of sight of the sample. All of the XeF_2 exposures, with the exception of the highest exposure (1260 L), were performed at a pressure of 1×10^{-6} Torr [1 langmuir (L) $\equiv 10^{-6}$ Torr sec.] The last exposure (1260 L) was carried out at a pressure of 1×10^{-5} Torr. After dosing, the chamber took some time (2–10 min) to return to ultrahigh vacuum and, consequently, the exposures are almost certainly underestimated.

The photoemission experiments were performed using an ellipsoidal mirror spectrometer[9] in the angle-integrated mode. The spectrometer was coupled to a 6 m/10 m monochromator[10] and both the 6 and 10 m configurations were used during the course of the experiment. The 6 m configuration was used to study the shallow Ga and As 3*d* core levels, whereas the deeper-lying F 1*s* level was studied with the monochromator in the 10 m configuration. In this paper we shall present only 6 m data. Most of the photoemission spectra presented in this paper were collected with a total energy resolution of $\simeq 140-160$ meV.

III. Ga AND As 3*d* CORE LEVELS

The changes that are induced in the Ga 3*d* core level during the initial stages of fluorination are illustrated in Fig. 1. The dots represent the raw data after the secondary-electron background has been subtracted. This was done by fitting a smooth cubic spline to the wings of the photoemission spectrum. The other components are the results of conventional least-squares analysis (e.g., see Ref. 11) of the core-level line shape, and the solid line through the points is the sum of the individual components.

Since the analysis of the Ga and As core-level line shapes involves fitting trial functions[11] to the raw data, which are incompletely resolved and often appear only as shoulders, the assumptions used in the least-squares analysis will be discussed in detail.

For the clean surface, both the As and the Ga 3*d* core-level emission were fitted using two spin-orbit-split, Gaussian-broadened, Lorentzian doublets. The starting parameters for the clean surface were obtained from an independent, constant-final-state photoemission study of the Ga and As core-level line shapes (see Table 1 of Ref. 12). These parameters, constrained to within ±5%, provided a good description of all of the clean-surface core levels that we studied.

Upon exposure to 10 L of XeF_2, the Ga 3*d* core level developed a noticeable tail to higher binding energy and the shallow dip at the top of the core level filled in (Fig. 1). The modification of the clean Ga 3*d* core level can be simulated by introducing an additional component to the Ga 3*d* core level shifted by $\simeq 0.8$ eV from the Ga 3*d* bulk component. In this, and subsequent fits to the Ga 3*d* level, the Lorentzian full width at half maximum (FWHM) was constrained to be 120 meV for all the components.[12] Additionally, the spin-orbit splitting, the spin-orbit–branching ratio, and the surface core-level shift were constrained to be 0.450 eV, 0.655, and 0.280 eV, respectively.[12] However, the Gaussian widths were allowed to vary subject to the constraint that the Gaussian broadening of the bulk and surface components was the same (the Gaussian broadening includes contributions from the instrumental broadening, the phonon broadening, and the effect of a nonhomogeneous surface potential). The position of the reacted features was also left floating. Obviously, these constraints are not unique, and although they can be justified on more rigorous physical grounds, our justification for using them is essentially pragmatic in nature; they provide a consistent series of fits over the entire exposure range that has been studied with the minimum of assumptions.

Upon further exposure to XeF_2, the Ga 3*d* core level develops a second reacted component shifted by $\simeq 1.8$ eV from the Ga 3*d* bulk component. Although the intensity of this component is weak at XeF_2 exposures of 60 L, it is quite noticeable at 110 L (Fig. 1).

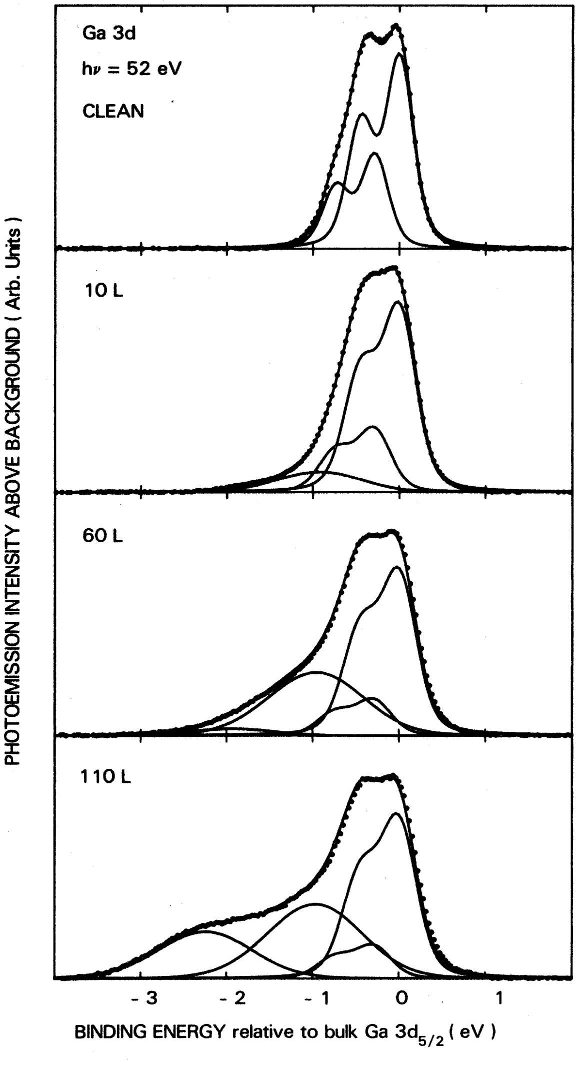

FIG. 1. Initial stages of fluorination. Ga 3*d* photoemission from the clean surface, under surface-sensitive conditions, and from the same surface dosed with increasing amounts of XeF_2. The dots represent the raw data after a smooth background has been subtracted. The other components, in order of increasing binding energy, are the Ga 3*d* bulk emission, the Ga 3*d* surface component, and the first and second chemically shifted components, respectively. The solid line through the points is the sum of the individual components. Notice that the intensity of the Ga 3*d* surface component decreases in a systematic fashion with increasing exposure. Notice also that the second chemically shifted component and the surface component coexist over a wide exposure range.

One of the surprising consequences of this analysis (Fig. 1) is the slow rate at which the Ga 3*d* surface component is attenuated by XeF_2 exposure. Although the second reacted component is already visible at XeF_2 exposures of 60 L, the surface component is still present. It may be argued that the intensity of the surface component is subject to a high degree of uncertainty because it is not resolved in the raw data. However, the shape of the Ga 3*d* core level, particularly near the maximum, is highly sensitive to the intensity and position of the surface component. By paying particular attention to the distribution of residuals around the maximum, the intensity of the surface component can be extracted fairly reliably. One piece of evidence which supports this assertion (see below) is the systematic manner in which the surface component is attenuated with increasing XeF_2 exposure. Furthermore, the As 3*d* surface component initially falls off at the same rate with increasing XeF_2 exposure.

The final stages of fluorination are illustrated in Fig. 2. As before, the Ga 3*d* core level is fitted with a bulk component, a surface component, and two reacted components. By 160 L the intensity of the second chemically shifted component is greater than the intensity of the first. However, the Ga 3*d* surface component is still present up to XeF_2 exposures of 260 L. Note that above 110 L the position and Gaussian width of the second chemically shifted component is entirely determined by the high-binding-energy tail of the Ga 3*d* emission. By 1260 L the Ga 3*d* surface component has finally disappeared, leaving only the bulk component and the two chemically shifted components.

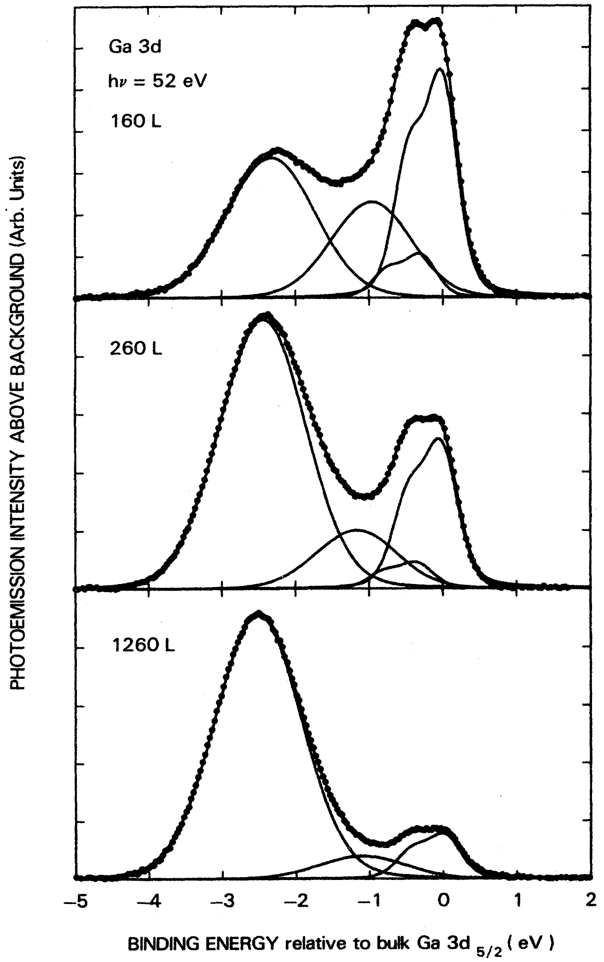

FIG. 2. Final stages of fluorination illustrating the growth of the second reacted component. Note that the surface component is still present up to XeF_2 exposures of 260 L.

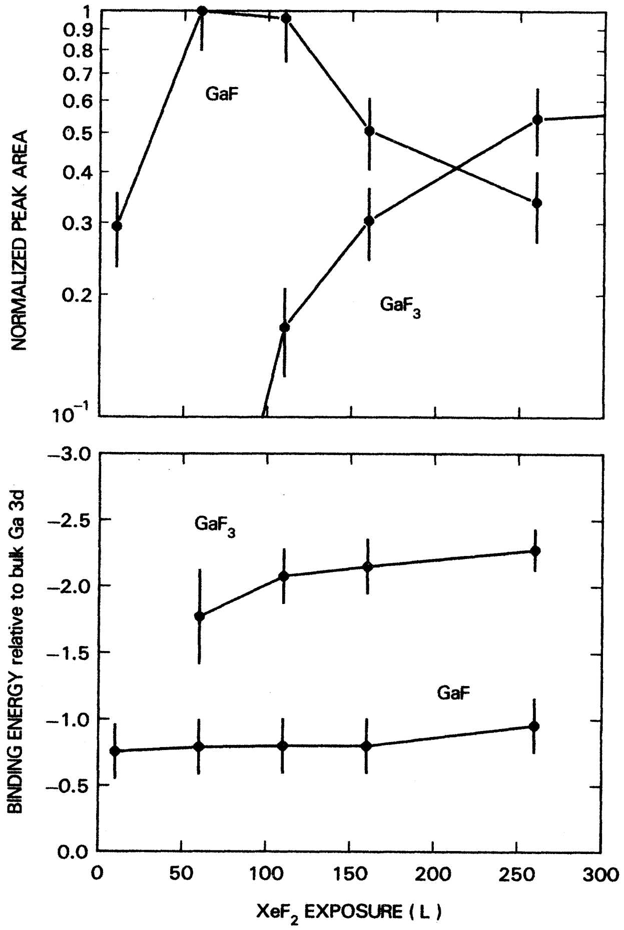

FIG. 3. Normalized peak areas of the two reacted components as a function of XeF_2 exposure (upper graph). Both components are normalized to the maximum peak area. For the first component this occurs at 60 L, whereas for the second the maximum peak area is found at the largest exposure studied (1260 L). Also shown is the binding energy of the two reacted components relative to the bulk Ga 3*d* component (lower graph).

The main findings of the least-squares analysis of the core-level line shapes are illustrated in Fig. 3. Both the position of the reacted components, relative to the bulk Ga 3*d* component, and the normalized area of the reacted components are presented as a function of the XeF_2 exposure. For reasons that will be discussed more fully later, these have been labeled monofluorides and trifluorides, respectively. As can be seen from the lower of the two graphs, both of the reacted features exhibit a gradual increase in binding energy with increasing XeF_2 exposure. This behavior has been observed before in a photoemission study of molecular O adsorption on GaAs(110).[4] The upper graph demonstrates that the intensity of the first reacted component originally increases, passes through a maximum, and then decreases. The intensity of the second reacted component rises sharply as the intensity of the first component is falling, suggesting that the formation of the second component obscures the first. Note that only two reacted components have been used to fit the Ga 3*d* line shape. We found that the inclusion of a third reacted component, situated between the two main reacted components, did not improve the quality of the fits. Although our analysis does not rule out the possibility that a third component is present, our results suggest that, if present, its intensity is small compared with the two reacted components that we have used. Note also that the Gaussian width of the reacted features exceeds twice the width of the bulk Ga 3*d* component. If the Gaussian width were determined only by the instrumental broadening, then the increased width of the reacted components over the bulk component would clearly be unphysical. However, it has been observed that molecular O (Refs. 4 and 5) also produces broad, chemically shifted components in the Ga and As 3*d* levels, and this may arise from an adsorption-induced modification of the surface potential in the neighborhood of the adsorbate. Miller and Chiang[5] have also argued that the width of the reacted component may arise from adsorbates in different adsorption sites. As mentioned above, we attempted to fit a larger number of reacted components, with a substantially narrower Gaussian linewidth, to both the Ga and As 3*d* core levels. However, we found that the latter choice of trial functions did not produce a satisfactory distribution of residuals.

We also note, in passing, that the effect of fluorination on the structural order of the surface could be inferred from the angular distribution of the photoelectrons measured using the ellipsoidal mirror analyzer[9] in display mode. For example, valence-band features on the clean surface, measured with kinetic energies in the range 25–50 eV, produced angular distributions which possessed the expected onefold symmetry about the GaAs(110) mirror plane. These were completely destroyed by XeF_2 exposures of 60–100 L.

The As 3*d* core level has been analyzed in a similar fashion. In some respects its exposure-dependent behavior is simpler than that of the Ga 3*d* core level because there appears to be only one chemically shifted component. The spin-orbit splitting, the spin-orbit–branching ratio, the surface core-level shift, and the Lorentzian linewidth (FWHM) were constrained to be 0.680 eV, 0.646, 0.370 eV, and 120 meV, respectively.[12] As before, the Gaussian linewidth was allowed to vary subject to the constraint that the Gaussian linewidth of the surface and bulk components was the same. The position and width of the reacted component were allowed to vary.

The exposure-dependent behavior of the As 3*d* core level is illustrated in both Figs. 4 and 5. Once again, it is necessary to introduce another component into the As 3*d* core level at XeF_2 exposures of 10 L to describe the effect of F adsorption. Further exposures of XeF_2 cause this component to grow, but by 150 L the intensity of this feature, relative to the bulk component, has saturated. As with the reacted components on the Ga 3*d* core level, the reacted component also increases its binding energy slightly with respect to the position of the bulk component upon XeF_2 exposure. Figures 4 and 5 also illustrate the gradual increase in the Gaussian broadening of the surface and bulk levels, also seen on the Ga 3*d* core

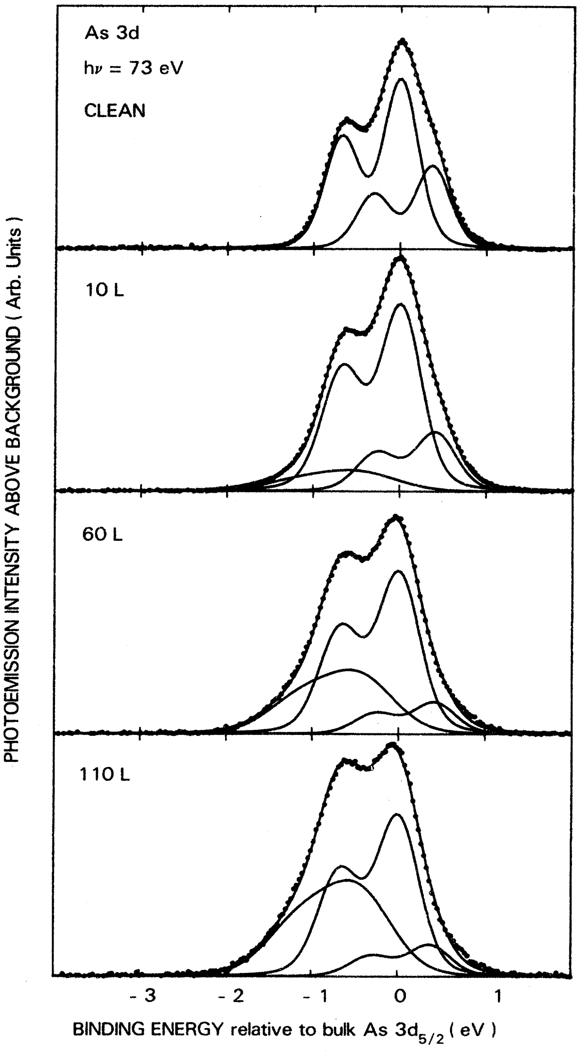

FIG. 4. Initial stages of fluorination. As 3*d* photoemission from the clean GaAs(110) surface and from the same surface exposed to increasing amounts of F. The photon energy was chosen such that the photoelectron escape depth was the same as Figs. 1 and 2. Therefore a direct comparison between Fig. 1 and this figure and Fig. 2 and Fig. 5 is possible. The components, in order of increasing binding energy, are the As 3*d* surface component, the bulk component, and the chemically shifted reacted component.

level, with increasing XeF_2 exposure. The exposure-dependent behavior of the As 3*d* core level is summarized in Fig. 6. The intensity of the first As 3*d* reacted component follows the exposure-dependent behavior of the first reacted component in the Ga 3*d* core level. It also displays an initial growth, followed by a gradual decrease. This suggests that the creation of the second Ga reacted component may also obscure the As reacted component.

IV. FLUORINE-INDUCED SURFACE BAND BENDING

The adsorption of atomic F on cleaved GaAs(110) surfaces produced room-temperature band-bending behavior characteristic of a highly electronegative species; the induced Fermi-level movement within the gap is substantially larger for *n*-type GaAs(110) than for *p*-type GaAs(110). On *p*-type surfaces, 100-L exposures of XeF_2 moved the Fermi level from just above the valence-band maximum (E_v) to $\simeq 0.30$ eV above E_v. Further XeF_2 exposure moved the Fermi level slightly higher, saturating at $E_F - E_V \simeq 0.4$ eV (1260 L). Likewise, the adsorption of XeF_2 on *n*-type GaAs(110) (1260 L) caused the Fermi level to move from just below the conduction-band minimum to within 0.45–0.60 eV of E_v.

This behavior is similar to the room-temperature adsorption of Cl on GaAs(110) (Ref. 2) and the low-temperature (100 K) adsorption of O on GaAs(110) (Ref. 13). In both cases the adsorbate produces much larger band-bending shifts on the *n*-type GaAs(110) surface than on *p*-type GaAs(110), and this behavior is considerably more pronounced when the substrate is cooled.[13] Several studies of metal adsorption on GaAs(110) surfaces, at low temperature, have demonstrated that the opposite is the case.[14,15] The deposition of metal adsorbates on GaAs(110), at low temperature, produces much larger

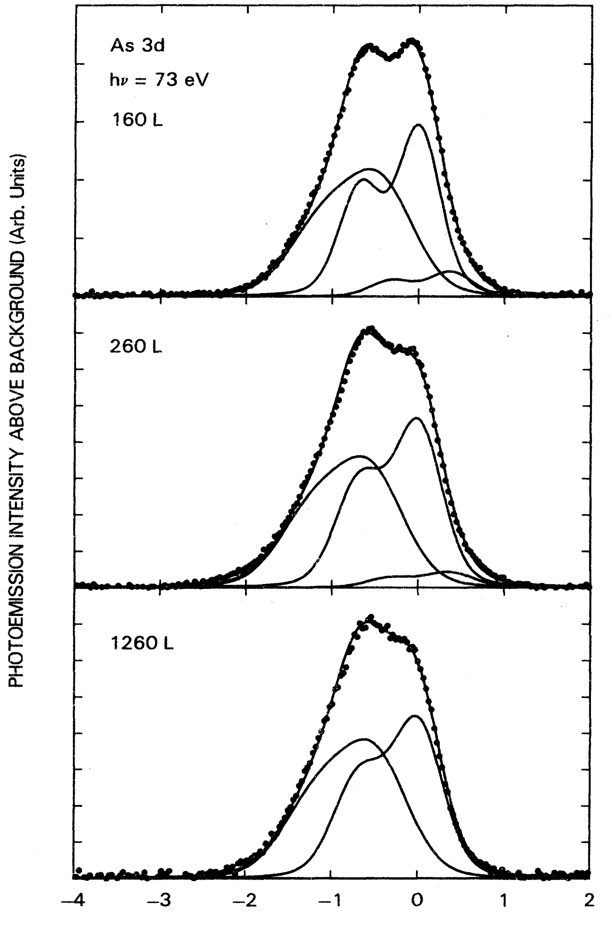

FIG. 5. Final stages of fluorination. Notice that the main changes in the As 3*d* core level above XeF_2 exposures of 160 L is produced by the gradual broadening of the bulk component and the disappearance of the surface component. The ratio of the reacted component to the bulk component does not change appreciably above 160 L. The exposure-dependent behavior of the As 3*d* reacted component is similar to the behavior of the first reacted component on the Ga 3*d* level, but quite different from the behavior of the second.

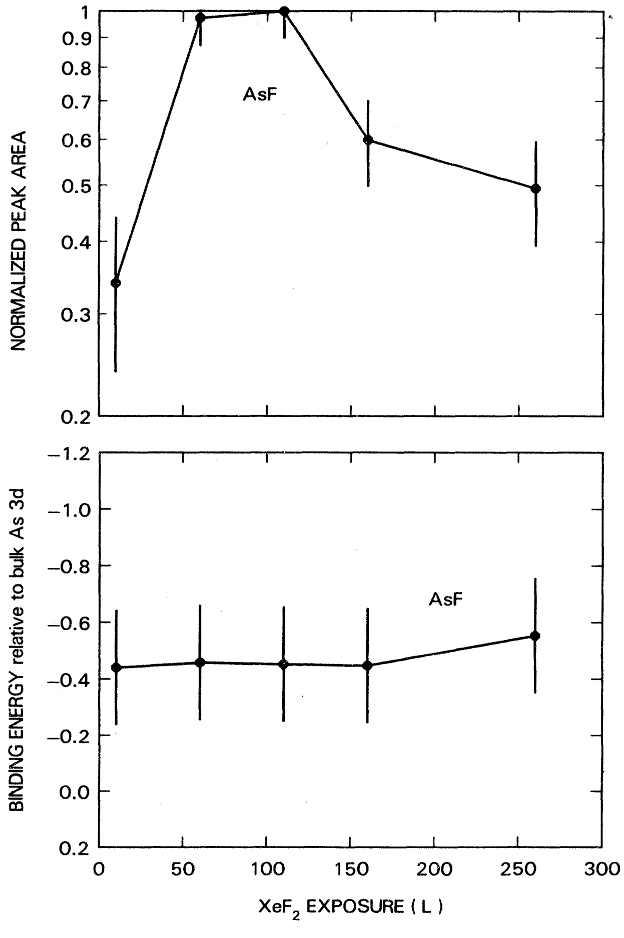

FIG. 6. Area of the reacted As component (top graph) normalized to the maximum peak area (110 L). The reacted component in the As 3*d* core level also increases in binding energy (bottom graph) with increasing XeF_2 exposure.

band-bending shifts on *p*-type GaAs(110) than on *n*-type GaAs(110). These observations have led to a model of adsorbate-induced band bending where the localized adsorbates themselves act as donors or acceptors.[14-17] For example, most of the electropositive metals are expected to produce donor levels within the gap at low coverage and produce much larger band-bending shifts on *p*-type GaAs(110) surfaces. The electronegative adsorbates, such as Cl, F, and O, are expected to form acceptor levels at low coverage and this, in turn, will produce much larger band-bending shifts on *n*-type GaAs(110) surfaces. For the *n*-type GaAs(110)-O system the depletion region around the room-temperature adsorption site has been imaged directly with scanning tunneling microscopy.[18] Consistent with the picture outlined above, a corresponding depletion region has not been observed around the adsorbed O atom on *p*-type material.[18]

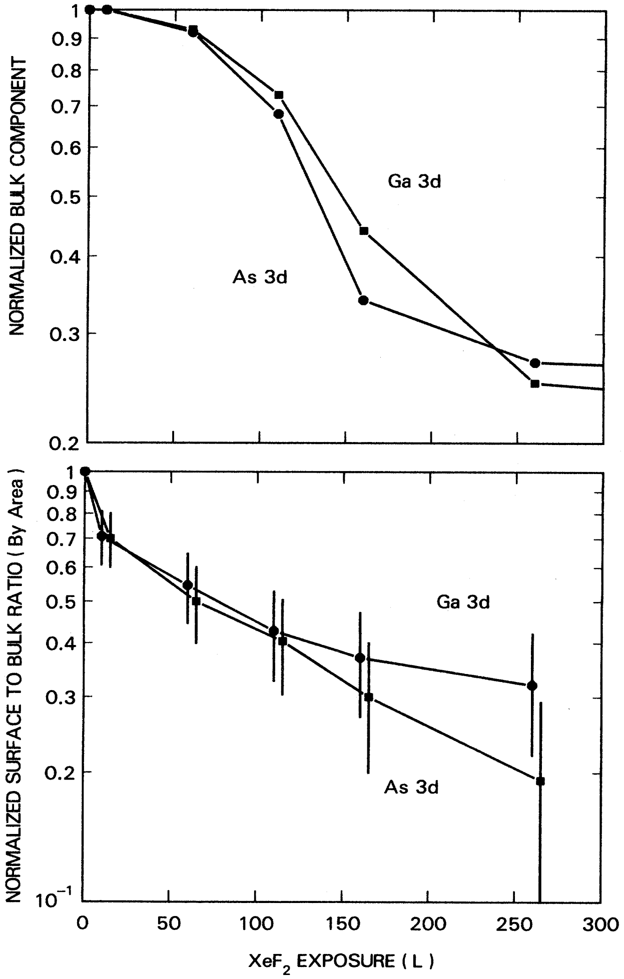

FIG. 7. Peak areas of the Ga and As 3*d* bulk components vs XeF_2 exposure (top graph) normalized to the intensity of the bulk components on the clean GaAs(110) surface. The area of the Ga and the As 3*d* surface components divided by the area of their respective bulk components (bottom graph). For clarity, the surface-to-bulk ratios of the As 3*d* core level has been displaced by 5 L to higher exposure.

V. DISCUSSION

In the preceding sections it has been demonstrated that the room-temperature adsorption of atomic F on GaAs(110) is surprisingly different from the room-temperature adsorption of either molecular Cl or molecular O. While the interaction between Cl and GaAs(110) has been shown to be predominantly an interaction between Cl and surface As,[1-3] we have demonstrated that F bonds to both the surface cations and the surface anions. For example, at low exposures to XeF_2, a chemically shifted feature is observed in both the Ga and As core-level spectra. Moreover, the rate at which the surface core-level component is attenuated is approximately equal for the cation and the anion. This behavior is consistent with simple bond-strength considerations. For example, using the diatomic bond strength as a figure of merit, we find that both the Ga—F ($\simeq 6.0$ eV) and the As—F ($\simeq 4.3$ eV) bond strengths are substantially larger than the Ga—As bond strength ($\simeq 2.2$ eV).[20] We have also demonstrated, again in contrast to Cl,[1-3] that for the range of exposures investigated here there is no evidence of an inherent saturation coverage. Indeed, the only point of obvious similarity between F and Cl is that they both move the Fermi level close to the valence-band maximum of GaAs. However, we note that, in their study of molecular Cl adsorption on GaAs(110), Troost *et al.*[2] found that the onset of the main Cl uptake coincided with the disappearance of the 20-eV transition in the low-energy electron-energy-loss spectra. The characteristic feature at 20 eV arises from transitions from the Ga 3*d* level to empty surface states.[21] This finding is inconsistent with the simple picture of Cl adsorption on GaAs(110), in which the Cl only bonds to the surface anions. Troost *et al.* also noted[2] that bond-strength considerations do not support the preferential bonding of Cl to surface anions. The diatomic bond strengths of the Ga—Cl and As—Cl bonds are both relatively large and of a comparable magnitude ($\simeq 4.99$ and 4.64 eV, respectively[20]).

The core-level photoemission results we have presented lead to a fairly simple picture for the adsorption and subsequent reaction of F on GaAs(110). We have demonstrated that upon initial XeF_2 exposure both the Ga and the As 3*d* core levels develop high-binding-energy components shifted by less than 1 eV from the bulk peak position. We have also shown that the anion and cation surface components are attenuated at approximately the same rate with increasing XeF_2 exposure. We attribute this behavior to the formation of surface Ga and As monofluorides at comparable rates. Both monofluoride components reach their maximum intensity in the exposure range between 50 and 110 L. However, it is important to note that the Ga and As surface components persists far beyond this, indicating that the initial stages of the F-substrate reaction is spatially inhomogeneous. Moreover, at higher exposures there is substantial intensity in the second chemically shifted Ga feature before the surface component has completely vanished, suggesting that the final stages of the F-substrate reaction are also spatially inhomogeneous. Initial-state charge-transfer considerations, comparison with binding energies of bulk Ga compounds, and the fact that the second reacted component in the Ga 3*d* core-level emission continuously evolves into a bulklike phase all strongly indicate that the second reacted component is associated with the formation of GaF_3. GaF_3 and AsF_3 are, of course, the expected major products in the reaction between GaAs and F. At room temperature this reaction appears to be a frustrated etching reaction. While GaAs is consumed, only As may be volatilized from the surface, owing to the low vapor pressure of GaF_3. Although the GaF_3 reaction product dominates the Ga spectra beyond exposures of 100 L, comparatively little is happening to the As 3*d* core-level emission, other than a gradual diminution of the surface component. However, it should not be inferred from this that the As remains chemically inert. In fact, the near constancy of the bulk Ga-to-As ratio, throughout the entire range of XeF_2 exposures, suggests that As must be removed from the surface, most probably as AsF_3.

The reaction of F with the stoichiometric GaAs(110) surface is different in many respects to the reaction of F with Si(111). First, our results suggest that the reaction is spatially inhomogeneous. The etching reaction appears to proceed while parts of the surface remained uncovered. No evidence for unreacted regions has been found, so far, in studies of Si(111).[8,19] Second, while thick layers of reaction intermediate subfluorides (SiF, SiF_2, and SiF_3) are always observed on Si(111), no intermediate fluoride species (e.g., GaF_2 or AsF_2) are observed on GaAs(110). These observations have kinetic consequences. In an interrupted-reaction experiment such as this, the intensity of a reaction intermediate should be inversely proportional to its reactivity. Thus, in the Si-F etching reaction, the rate-limiting nature of the SiF_3+F reaction step is manifested by a large buildup of SiF_3 groups on the substrate and the formation of a thick layer of reaction intermediates. Conversely, the absence of detectable reaction intermediates on GaAs(110), except for the surface monofluoride species, suggests that it is the initial disruption of the subsurface GaAs lattice that is rate limiting. Perhaps the disruption resulting from greater-than-surface fluorination raises the energy of the surrounding Ga and As atoms so that further reaction to completion is extremely facile.

VI. CONCLUDING REMARKS

The adsorption of atomic F on GaAs(110) has been studied using high-resolution core-level photoelectron spectroscopy. Least-squares analysis of both the As and Ga 3*d* core levels suggests that the initial stage of F adsorption involves F bonding both to surface As and Ga. This produces a chemically shifted component in both the As and Ga 3*d* core levels which saturates upon increasing XeF_2 exposure. By XeF_2 exposures of 60 L a second chemically shifted component is observed in the Ga 3*d* core level and its intensity does not appear to saturate with increasing exposure. The binding-energy difference between the second reacted component and the bulk Ga 3*d* component is approximately 3 times that of the first reacted component, suggesting that the former arises from a trifluoride reaction product. The fact that no corresponding feature is observed on the As 3*d* component is consistent with the basic thermodynamic considerations. At room temperature, GaF_3 is a solid, whereas AsF_3 is gaseous.[20]

One of the more surprising results of the least-squares core-level line-shape analysis is the persistence of the Ga and As 3*d* surface components upon XeF_2 exposure. Although the suggestion[4] that the oxidation of GaAs(110) was spatially inhomogeneous did meet with substantial resistance,[6,7] our results strongly suggest that the fluorination of GaAs(110) is spatially inhomogeneous. The evidence for this comes from the fact that the Ga 3*d* surface component is present over a wide coverage range with the second reacted feature, which we have assigned to GaF_3. Although it may be argued that the surface component is not actually resolved in our raw data, we have emphasized that the area around the maximum of the Ga core level is very sensitive to the position and intensity of the surface component. Furthermore, the low-binding-energy side of the As 3*d* core level is broadened by the presence of the surface component up to XeF_2 exposures of 260 L. As an independent check, it would be interesting to study this system with low-energy electron loss. One may expect the surface-exciton losses[21] to exhibit the same degree of persistence as the surface core-level components. Finally, we have noted that the adsorption of F on GaAs(110) is significantly different from the adsorption of Cl on GaAs(110); the reasons for this are not entirely clear.

ACKNOWLEDGMENTS

The assistance of J. Yurkas, A. Marx, and C. Costas is acknowledged. Research was carried out at the Brookhaven National Laboratory, which is sponsored by the U.S. Department of Energy, Division of Materials Science and Division of Chemical Sciences.

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