

The temperature dependence of the $\text{Cl}_2/\text{GaAs}(110)$ surface product distribution

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The reaction of Cl_2 with $\text{GaAs}(110)$ is studied with soft x-ray photoelectron spectroscopy (SXPS). The temperature dependence of the surface product distribution, in the range of 300–650 K, is derived from SXPS core-level and valence-band spectra and compared to known gas-phase product distributions. It is found that both Ga and As chlorides are formed at room temperature. Following reaction at temperatures above ~ 400 K, no Cl remains on the surface. Instead, for temperatures up to ~ 600 K an As overlayer is formed, whereas reaction at ~ 650 K leads to the stoichiometric removal of Ga and As. These findings indicate that there is a direct correlation between the surface and gas-phase product distributions. © 1996 American Institute of Physics. [S0021-9606(96)03501-5]

I. INTRODUCTION

The reaction of chlorine with $\text{GaAs}(110)$ has received a lot of attention in the past few years and, because of this, quite a bit has been learned about the reaction. It is technologically important to understand the interaction of chlorine in all of its forms (i.e., Cl_2 , Cl_2^+ , Cl, and Cl^+) with GaAs surfaces since chlorine plasmas are commonly used to dry etch GaAs wafers during device manufacture. The (110) face of GaAs is often investigated because it has a single, well-known, easily reproducible reconstruction with a 1:1 Ga:As surface stoichiometry, as compared to the polar (100) and (111) faces which have many possible reconstructions, depending on the stoichiometry of the surface region. It has been shown that Cl_2 chemisorbs dissociatively on $\text{GaAs}(110)$, via a molecular precursor,^{1–3} with Cl atoms bonding at both Ga and As sites.^{4–8} Under some conditions, the (110) surface can be passivated by a monolayer of Cl atoms.⁹ Otherwise, Cl_2 continuously etches GaAs, removing both Ga and As stoichiometrically.^{10–12}

Much of what has been learned about the etching reaction stems from molecular beam scattering investigations of the volatile reaction products.^{1–3,9–13} For example, the gas-phase product distribution depends on the temperature of the surface, T_s . At room temperature, it is composed primarily of GaCl_3 and AsCl_3 , whereas GaCl and elemental As (i.e.,

As_2 or As_4) are the principal products at higher temperatures.^{11,12} The reactivity of the surface increases from room temperature up to ~ 400 K, where it levels off to a nearly constant value.^{10–12} It remains constant up to ~ 550 –600 K, where it again rises.^{10–12} The observed changes in the reactivity and gas-phase reaction product distributions would be better understood with supporting measurements of the temperature dependence of the surface product distribution. Previous studies of the surface product distribution for this system, however, have focused only on the room-temperature reaction.^{4,6,7}

In this experiment, the temperature dependence of the surface product distribution is measured, using soft x-ray photoelectron spectroscopy (SXPS), for the $\text{Cl}_2/\text{GaAs}(110)$ reaction in the temperature range of 300–650 K. It is found that, at low temperatures (~ 300 –400 K), the reaction generates a surface containing primarily Ga and As chlorides. For reactions carried out at temperatures above ~ 400 K, no Cl remains on the surface. Instead, the only surface reaction product is elemental As, up to ~ 650 K. These results correlate well with measurements of the volatile reaction products, indicating a direct correspondence between the gas-phase and surface product distributions.

II. EXPERIMENTAL PROCEDURE

The experimental procedure used in this study is identical to that reported for the investigation of the XeF_2/GaAs

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reaction.^{14–16} GaAs(110) wafers (*p* type, Zn doped, 10^{18} cm⁻³, $\pm 0.5^\circ$, ~ 1 cm \times 2 cm \times 1 mm) were first cleaned by etching in a 1:1:200 solution of HNO₃:H₂O₂:H₂O, then rinsed with de-ionized water and isopropanol before being placed in the ultrahigh vacuum (UHV) chamber. The samples were mounted on Ta foil, which was used for heating the sample. The samples were cleaned in UHV by sputtering with 500 eV Ar⁺ ions, followed by annealing to ~ 825 K.

Measurements of the sample temperature were made with an Ircon infrared pyrometer (model WO8CO2), designed to measure temperatures in the range of ~ 550 – 1050 K. An emissivity of 0.6 was used in determining the temperature, based on a tabulated value of 0.70 for GaAs and a scaling factor of 0.85 to account for absorption by the sapphire window. Since it was not possible to measure the sample temperature during Cl₂ dosing or annealing, a calibration of temperature to heating current was made for each sample prior to its use. For temperatures below ~ 550 K, direct measurement could not be made, so a linear extrapolation of the high-temperature data was used to determine the appropriate heating current. From the reproducibility of the pyrometer temperature-to-current calibration, and from a comparison to subsequent measurements made using a *K*-type thermocouple, the uncertainty in reported temperatures is estimated to be ± 25 K.

Samples were exposed to Cl₂ in a separate stainless steel UHV chamber, which had a typical base pressure of 5×10^{-10} Torr. Sample transfers between the dosing and analysis chambers were carried out entirely under UHV. The dosing chamber was passivated with a large amount of Cl₂ prior to any exposures to reduce the risk of vapor phase transport of metal chlorides to the sample surface. A cold cathode ion gauge was used to monitor the pressure in the dosing chamber in order to avoid the creation of both metal chlorides and radical species from the hot filament of a conventional ion gauge. No evidence of metal contamination was apparent in any of the SXPS spectra collected.

Samples were exposed to Cl₂ in an identical manner throughout the experiment. A clean GaAs(110) wafer was first heated to the appropriate temperature then exposed to 10^{-4} Torr of Cl₂ for 100 s. The wafer was then held at its elevated temperature while the Cl₂ was pumped out. Although this may have led to some changes in the surface product distribution due to additional annealing, it was the only way to avoid reaction at lower temperature. Since the overall Cl₂ pressure was low, even during dosing, this effect should be minimal. Furthermore, the molecular beam scattering experiments to which the present data are compared were run under similar conditions of low Cl₂ pressure. Following chlorination, the sample was quickly transferred to the analysis chamber (base pressure = 1×10^{-10} Torr) to minimize the chance of surface contamination through room-temperature reaction with any residual chlorine. Samples were then allowed to cool to room temperature in the analysis chamber before any SXPS spectra were collected.

The experiments were performed at beamline UV-8a, at the National Synchrotron Light Source located at

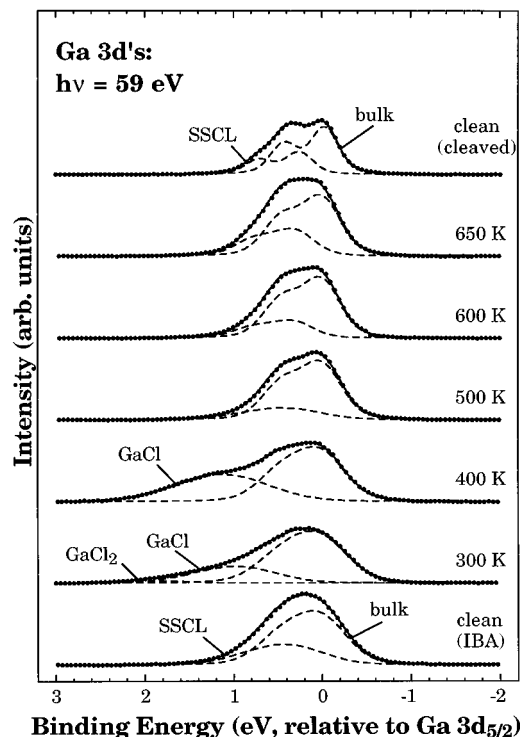


FIG. 1. High-resolution SXPS spectra of the Ga 3d core level, collected from GaAs(110) surfaces exposed to 10^{-4} Torr of Cl₂ for 100 s at temperatures ranging from 300 to 650 K are shown along with numerical fits to the data. The dots are the background-subtracted raw data. The dashed lines show the individual fit components. The solid line is the sum of the fit components. Spectra are scaled so that the bulk Ga 3d components have identical areas, and are labeled with their corresponding reaction temperature. Also shown in the figure are spectra collected from clean surfaces, which contain both bulk and surface shifted core level (SSCL) components. The spectrum labeled “IBA” is from a sputtered and annealed surface. The spectrum labeled “cleaved” is from a cleaved surface (Ref. 6), and was collected with an incident photon energy of 52 eV.

Brookhaven National Laboratory. The SXPS spectra were collected with an angle-integrating ellipsoidal mirror analyzer, which accepts photoelectrons emitted from the sample within an $\sim 85^\circ$ cone centered about the surface normal. The photon energies were selected with a 3 m focal length grazing incidence toroidal grating monochromator. The high-resolution SXPS spectra have a combined energy resolution of ~ 0.2 eV.

III. RESULTS

High-resolution Ga and As 3d core-level spectra are shown in Figs. 1 and 2, along with numerical fits to the data. These spectra were collected from GaAs(110) surfaces reacted with Cl₂ at temperatures ranging from 300 to 650 K. Also included in Figs. 1 and 2 are spectra collected from clean GaAs(110). The photon energies used to collect these spectra were selected so that the measured photoelectrons all have kinetic energies of ~ 35 eV, which maximizes the surface sensitivity of the measurement and ensures that Ga and As 3d spectra reflect the composition of the same part of the near-surface region. The spectra in Figs. 1 and 2 are scaled

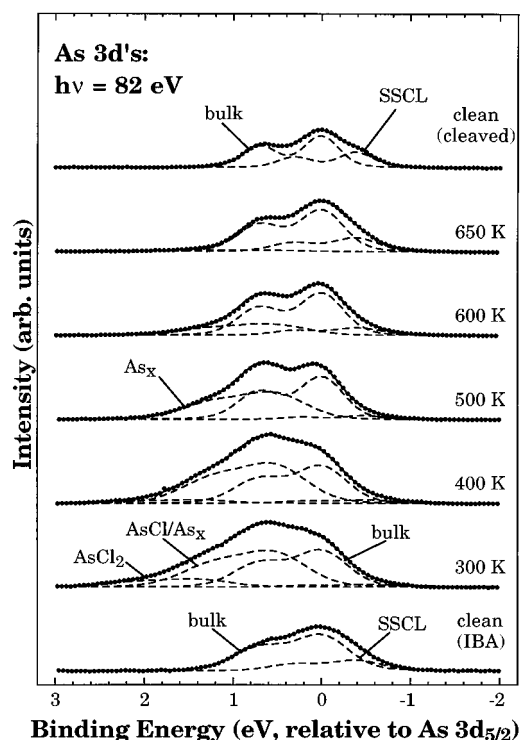


FIG. 2. High-resolution SXPS spectra of the As 3d core level, collected from the same surfaces as in Fig. 1, are shown along with numerical fits to the data. The dots are the background-subtracted raw data. The dashed lines show the individual fit components, and the solid line is the sum of the fit components. Spectra are scaled so that the bulk As 3d components have the same areas, and are labeled with their corresponding reaction temperature. Also shown in the figure are spectra collected from clean surfaces, which contain both bulk and surface shifted core level (SSCL) components. The spectrum labeled "cleaved" was collected with an incident photon energy of 73 eV.

so that the bulk components have identical areas, and they are offset for presentation. Note that the spectra from cleaved GaAs(110), which are included in Figs. 1 and 2 for comparison, are from Ref. 6 and were collected with slightly different photon energies, as noted in the captions.

The Ga and As 3d core-level spectra were numerically fit, after background subtraction, to a sum of Gaussian-broadened Lorentzian spin-orbit split doublets, using a least-squares optimization routine. A smooth background was determined by fitting a third-order polynomial to each side of the peak. The fitting procedure was used to determine the binding energy (BE), area, and Gaussian contribution to the width of each core-level component. The Lorentzian full width at half-maximum was fixed at 0.12 eV in all of the fits. For the Ga 3d core level, the spin-orbit splitting and branching ratio were fixed at 0.45 eV and 0.66, respectively. For As 3d, these parameters were fixed at 0.68 eV and 0.65, respectively. These fitting parameters are consistent with previous studies of the GaAs(110) surface.^{6,14-18}

The shifted components in the Ga 3d core-level spectra of chlorinated GaAs have been previously identified.^{6,7} For clean GaAs(110) surfaces, there is a surface-shifted core-level (SSCL) component 0.28 eV higher in BE than the bulk

component. In the spectra collected from surfaces reacted at the lowest temperatures, the SSCL is replaced by a component shifted ~ 0.9 eV higher in BE that is due to GaCl. In the spectrum collected from the room-temperature reacted surface, there is also a component shifted ~ 1.7 eV higher in BE due to the presence of a small amount of GaCl₂. Note that, for the surfaces reacted at temperatures of ~ 500 K and above, the inclusion of SSCL and bulk components is sufficient to achieve good fits.

The shifted components in the As 3d spectra have been similarly identified.^{6,7,19} For clean GaAs(110) surfaces, there is a SSCL component 0.39 eV lower in BE than the bulk component. In the spectra collected from the chlorinated surfaces, there is a component shifted ~ 0.5 eV higher in BE, which was previously identified as arising from AsCl,^{6,7,19} but which is also consistent with elemental As on the surface.^{14,20-22} For the reasons discussed below, in the spectra collected from samples reacted at lower temperatures this feature arises primarily from AsCl, whereas in the spectra collected from samples reacted at temperatures of ~ 500 K and above this feature arises solely from elemental As. In the spectra collected from the surfaces reacted at the lowest temperatures, there is also a component shifted ~ 1.4 eV to higher BE, due to AsCl₂. Note that, in all of the As 3d spectra, it was necessary to include a SSCL component in order to get good fits to the data. The persistence of the SSCL may either be due to exposed patches of clean surface or to subsurface defects generated in the reaction, as discussed in Refs. 6 and 16.

Valence-band spectra, collected from the same surfaces as the spectra in Figs. 1 and 2, are shown in Fig. 3. The spectra have been scaled to have equal intensity at the same point, 11 eV below the valence-band maximum (VBM), and are offset for presentation. For the clean GaAs(110) surface, there are three noticeable valence-band features, located ~ 1 , 6, and 11 eV below the VBM.^{23,24} The lowest energy feature, which spans from about 0 to 4 eV below the VBM, results from two bands of *p*-like states associated with As, whereas the one located at ~ 6 eV from the VBM is due to a combination of *s*-like Ga and *p*-like As states, and the broad feature centered at ~ 11 eV is from an *s*-like As state.²⁴ Following chlorination at the lowest temperatures, the clean-surface features are less pronounced and a new, broad feature shows up 3–5 eV below the VBM, indicative of chlorine chemisorption.^{6,25-27} However, this chlorine-induced feature is not apparent in spectra collected from surfaces reacted at temperatures of ~ 500 K and above. Instead, in the $T_s = 500$ K spectrum there is a new, sharper feature 2–3 eV below the VBM that is thought to derive from elemental As (see the discussion below). This As-induced feature is still visible in the spectrum collected from the surface reacted at 600 K, but the $T_s = 650$ K spectrum is nearly identical to those collected from clean surfaces.

A simple calculation based on the areas of each of the core-level components provides a quantitative estimate of the amount of each chemical species present on the surface following reaction. If the ratio of the SSCL area to the total core-level area is assumed to represent the signal from half a

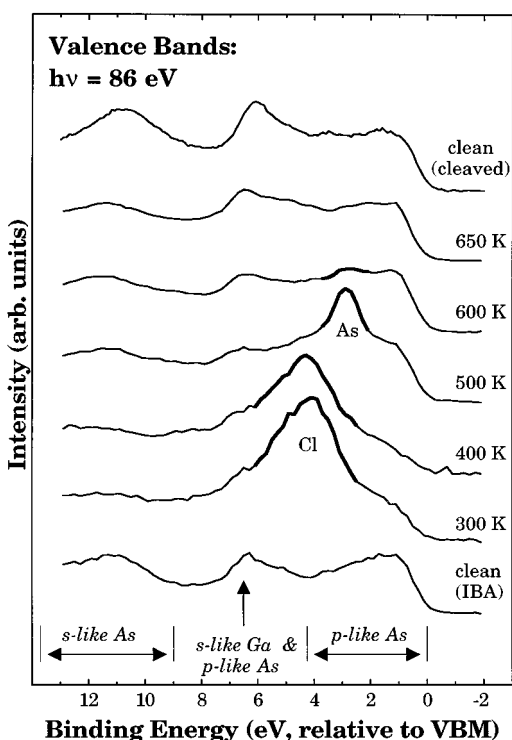


FIG. 3. Valence-band spectra collected from the same surfaces as in Fig. 1 are shown. The spectra are scaled to equal intensity 11 eV below the VBM, and are labeled with their corresponding reaction temperature. Regions of the valence bands are labeled according to the assignment scheme given in Ref. 24. The spectrum labeled “cleaved” was collected with an incident photon energy of 63 eV.

monolayer (ML) of surface atoms (4.6×10^{14} atoms/cm²), then the area ratios of each of the shifted components can be converted into coverages. Neglecting attenuation within the overlayer and chemically induced variations in the photoionization cross sections, the coverage of each species is proportional to its relative contribution to the total core-level intensity. Coverages estimated in this manner are presented in Table I. The coverage of Cl adsorbed on the surface, Cl(a), is found by summing the coverages of each chlorine-containing species, appropriately weighted by the number of Cl atoms in that species, and is also given in Table I.

IV. DISCUSSION

A combination of previous results with those reported here gives a more complete picture of the temperature dependence of Cl₂/GaAs etching reaction. This reaction can be broken down into three temperature regimes: (1) room temperature to ~400 K, where both Ga and As chlorides are formed; (2) ~500–600 K, where Cl is no longer stable on the surface, but elemental As is; and (3) ~650 K and above, where none of the reaction products are stable on the surface.

The room-temperature etching of GaAs(110) with Cl₂ has been investigated previously,^{5–7,11,13} and the results of this study are in accord with these findings. At room temperature, the reaction leads to the formation of both Ga and As chlorides on the surface. The presence of surface GaCl and GaCl₂ is consistent with the chemical shifts seen in photoemission,^{6,7} with Cl atoms located above Ga sites seen with scanning tunneling microscopy (STM),^{5,7} and with the volatile gallium chlorides seen in molecular beam scattering experiments.^{11,13} The 0.5 eV shift to higher BE seen in the As 3d core level following chlorination at room temperature could be due to elemental As, but the presence of an AsCl₂ component, and the stepwise manner in which the two components grow in with exposure (see Ref. 6), strongly suggest that the 0.5 eV shifted component, in large part, arises from AsCl. Furthermore, the volatile As-containing reaction products at room temperature are primarily As chlorides,^{11,13} and STM images of Cl₂/GaAs(110) show Cl atoms situated over As sites.^{5,7} Thus, there may be some contribution to the 0.5 eV shifted component from elemental As, but since neither As₂ nor As₄ contribute significantly to the gas-phase product distribution at room temperature,^{11,13} adsorbed elemental As is, at best, a spectator in the room-temperature etching reaction.

The surface product distribution changes as the surface temperature is raised. Following reaction at 400 K, there is no GaCl₂ and only a trace of AsCl₂ present. The removal of higher chlorides correlates with the observed increase in surface reactivity between 300 and 400 K,^{10–12} and is likely due to an increase in the volatility of chlorine compounds at these temperatures.

For reaction temperatures above ~400 K, however, the chemical composition of the surface is greatly modified, as

TABLE I. Estimated coverage of each surface species, in ML, where 1 ML is defined as the total number of Ga and As atoms in the outermost layer of an ideal, stoichiometric GaAs(110) surface. The Cl coverage is estimated by assuming that all of the AsCl/As_x contribution is from AsCl, and thus represents an upper limit.

	Coverage of each surface species (ML)							
	As				Ga			Cl
	SSCL	AsCl/As _x	As _x	AsCl ₂	SSCL	GaCl	GaCl ₂	Cl(a)
Clean	0.50	0.50
300 K	0.06	1.00	...	0.22	...	0.41	0.04	1.93
400 K	0.08	1.10	...	0.09	...	0.64	...	1.92
500 K	0.10	...	0.94	...	0.31
600 K	0.26	...	0.50	...	0.37
650 K	0.52	...	0.07	...	0.50

seen by the lack of any chemically shifted components in the Ga 3*d* spectra and by the loss of the broad Cl-induced feature in the valence bands. Although the Ga 3*d* spectra collected from samples reacted at temperatures of ~ 500 K are nearly identical to those from the clean surfaces, the valence-band and As 3*d* spectra still show signs of some reaction product on the surface. The valence-band spectrum collected from the sample reacted at ~ 500 K lacks the broad Cl-induced feature, and instead has a much sharper feature 2–3 eV below the VBM. The disappearance of the Cl-induced feature indicates that there is little or no Cl on the surface. However, there is still a prominent high BE feature in the As 3*d* spectra. Since it is unlikely that this component has a significant contribution from As bound to Cl, it is therefore attributed to elemental As. The appearance of the new feature 2–3 eV below the VBM, in the region of *p*-like As states, is consistent with features evident in valence-band spectra when As is evaporated on GaAs(110).^{20,28} Note that the formation of elemental As at elevated temperatures has also been observed in the analogous XeF₂/GaAs etching reaction.¹⁴ From Table I it is seen that there is ~ 1 ML of As on the surface following reaction at ~ 500 K. This is consistent with experimental²⁰ and theoretical²⁹ observations that up to 1 ML of As is stable on GaAs(110). The formation of an As overlayer in reactions carried out between ~ 400 and 600 K may be responsible for the constancy of the surface reactivity in this temperature range.^{10–12}

Following reaction at ~ 650 K, the surface is free of any reaction products and essentially has the stoichiometry of a clean surface (see Table I). Thus, whereas chlorination at temperatures of ~ 500 –600 K leads to the preferential removal of Ga and the buildup of elemental As on the surface, reaction at ~ 650 K leads to the stoichiometric removal of both Ga and As. The results of molecular beam scattering experiments indicate that As is removed primarily as As dimers and tetramers at ~ 650 –700 K, whereas Ga is removed primarily as GaCl.^{11–13} This is also supported by thermal desorption experiments, which found that only GaCl and As desorb from chlorinated GaAs(110), with As desorbing at ~ 700 K.^{30,31}

Hence, chlorination at ~ 650 –700 K may provide an adequate means of cleaning GaAs wafers, as long as this process does not cause the surface to roughen. Although SXPS is not the best technique for determining the degree of surface disorder, some information can be gleaned from the widths of the line shapes since disorder increases these widths. The components of the core-level spectra collected from the surface reacted at 650 K have less Gaussian broadening than the components of the spectra collected from a sputtered and annealed clean surface (see Figs. 1 and 2). The widths of the bulk and SSCL components are, in fact, comparable to those of the cleaved surface. Thus, this sharpening up of the core level components indicates that chlorination at ~ 650 K produces a more well-ordered surface than does sputtering and annealing.

V. CONCLUSIONS

The chemical composition of the surface as a function of temperature follows the general trends seen in the gas phase product distributions. Namely, gallium and arsenic chlorides are formed at lower temperatures, with gallium chlorides and elemental arsenic formed at higher temperatures. Since gallium chlorides and elemental arsenic are the thermodynamically predicted products for the reaction conditions typical to these experiments,^{32,33} it is expected that raising the surface temperature, and thereby overcoming any low-temperature kinetic limitations, leads to their formation. The increase in reactivity between 300 and 400 K, seen with molecular beam scattering, correlates with an apparent increase in the volatility of chlorine compounds at this temperature. The further increase in reactivity at ~ 550 –600 K marks the onset of the stoichiometric removal of Ga and As. Thus, a direct correlation is found between the surface and gas-phase product distributions.

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