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TECHNICAL REPORT-10

## SPECIES-SPECIFIC DENSITIES OF STATES OF Ga AND As IN THE CHEMISORPTION OF H<sub>2</sub>O ON GaAs(110)

by

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SPECIES-SPECIFIC DENSITIES OF STATES OF Ga AND As IN THE CHEMISURPTION OF H<sub>2</sub>O ON GaAs(110)\*

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Abstract

Auger line shape measurements for Ga and As have been made for the adsorption of  $H_2O$  on cleaved GaAs(110) to determine the species-specific densities of states at the Ga and As sites. The adsorption kintics for  $H_2O$  has been compared to that for  $O_2$ . It is found that  $H_2O$  adsorbs two or three orders of magnitude faster than  $O_2$ . The Auger lineshapes at corresponding coverages are similar, indicating that the changes in the local charge distribution around Ga and As caused by  $H_2O$  adsorption is similar to that caused by  $O_2$  adsorption.

## I. Introduction

Because of the potential usefulness of stable insulating layers on III-V compound semiconductors, the adsorption of oxygen on GaAs surfaces has been the focus of considerable effort.(1-20) Interpretation of results for oxygen chemisorption, such as a determination of the equilibrium bonding site or the adsorption mechanism, may, however, be unreliable if competing adsorption processes occur that are not readily distinguishable. The adsorption of H<sub>2</sub>O is one possibility. If H<sub>2</sub>O adsorbs more readily than oxygen, changes in electron-spectroscopic measurements ascribed to oxygen could be caused by the presence of small amounts of  $H_2U$  in the ambient gas. In particular, differences occurring in the initial stages of adsorption, as are observed for oxygen chemisorption on GaAs(110), may then be ascribable to the adsorption of  $H_20$ . We summarize here results of experiments addressing these questions. In these experiments the kinetics of  $H_20$  and  $0_2$  adsorption on cleaved GaAs(110) were measured and the species-specific densities of states of Ga and As for H<sub>2</sub>O adsorption recorded and compared to our previous results for O adsorption.<sup>(21)</sup>

The local charge distribution at the Ga and As surface sites can be monitored as a function of coverage on the GaAs(110) surface by measuring for each species of interest an Auger transition that involves the valence band. Because of the dipole-dipole nature of the Auger process, the probability of finding the "up" electron in an Auger transition a distance R from the initial core hole falls off as  $1/R^6$ .<sup>(22)</sup> From these measurements Auger lineshapes can be extracted that can be interpreted as a fingerprint of the species-specific valence band density of states around the particular species of interest. We have made such measurements for H<sub>2</sub>U adsorption on GaAs(110) and followed changes in Ga and As lineshapes with water vapor exposure. Adsorption kinetics measurements are made by determining the coverage as a function of exposure. The KLL oxygen line is used to monitor the coverage.

II. Experimental

The experiments were conducted in a stainless steel UHV system with a base pressure of  $< 1 \times 10^{-10}$  torr on n-type Te-doped GaAs crystals. Samples were cleaved in vacuum, using a knife-edge and anvil technique. Water vapor from a stainless steel container of deionized water was admitted to the vacuum chamber through a variable leak valve. Prior to admission to the main chamber, the water was degassed by pumping on it with a turbomolecular pump. The chamber pressure was monitored with an ion gauge. Exposures between 10 Langmuirs and  $10^6$  L were made.

The signal-averaged Ga and As  $M_1M_{45}V$  Auger lines were deconvoluted with an elastically backscattered-electron spectrum and with a Lorentzian representative of the core levels involved in the CCV transitions. The backscattered-electron spectrum serves to model the electron energy loss and instrument broadening functions. This procedure has been described in detail elsewhere.<sup>(23)</sup> These Auger transitions occur at kinetic energies close to the minimum of the electron mean free path ( $\lambda$ ~7Å), providing very good surface sensitivity.

III. Results and Discussion

When the GaAs(110) surface is exposed to water vapor, rapid adsorption occurs. This adsorption manifests itself as an increase in the O KLL Auger signal. It is not known what the adsorbing species is. The magnitude of the O KLL line was used as a measure of the coverage. Coverages were determined by considering the oxygen KLL signal to be due to OH (or O) sitting on top of GaAs(110) with no rearrangement of OH(or O), Ga, or As perpendicular to the surface. Calculated concentrations using this model were compared to measured concentrations using the 0 KVV and the Ga and As LMM Auger peak heights with the appropriate sensitivity factors<sup>(24)</sup> and escape depths.<sup>(25)</sup> Figure 1 shows a comparison of kinetics measurements for H<sub>2</sub>O and O<sub>2</sub>. For H<sub>2</sub>O the exposure required to reach a given coverage is always at least one to two orders of magnitude smaller than that for oxygen. At the lowest coverages it is smaller by a factor of  $10^3$  or more. The implication is that a ratio of H<sub>2</sub>O and O<sub>2</sub> pressures of at least  $10^{-4}$  must be maintained in order to be certain that there is no influence of H<sub>2</sub>O on any of the measurements. In our O<sub>2</sub> adsorption measurements care was taken that this ratio way maintained.<sup>(21)</sup>

The influence of the  $H_2O$  adsorption on the local charge distribution at the Ga and As sites was measured by following the Ga and As  $\mathrm{M_{1}M_{45}V}$  lines. These undergo changes as the coverage increases. Figure 2 shows the  $M_1M_{45}V_{7}$ derived Ga and As-specific densities of states as a function of coverage. Coverage here refers to the amount of oxygen as derived from the O KLL line. The Ga-specific density of states undergoes substantial changes. At  $\theta = 0.3$ the intensity of peaks A and C have increased relative to peak B, and peak B has broadened. At  $\theta$  = 0.5 the intensity at the bottom of the band has increased slightly. For increasing coverages, the intensity at the bottom of the band continues to increase, and at  $\theta$  = 0.65 this feature appears as a distinguishable peak at about -9.5 eV. The As-specific density of states also undergoes changes with increasing coverage. For coverages up to about 1 monolayer those changes consist of a shifting intensity from the top of the band to the middle of the band, such that region E no longer forms a valley between peaks D and F. At  $\theta$  = 1 there is a large increase in intensity in the center of the As-specific DOS.

To aid the interpretation of these results a comparison is made with those for GaAs exposed to oxygen<sup>(21)</sup>, shown in Figure 3 for the Ga-specific DOS and in Fig. 4 for the As-specific DOS. The Ga spectra are quite similar to each other for similar "oxygen" coverages, as are the As spectra for This indicates that the charge distributions around Ga and around As  $\theta < 1$ . are the same at similar "oxygen" coverages, whether this coverage is due to exposure to oxygen or to water vapor. There are two possible interpretations. One is that OH is bonded to the surface atoms, but that the line shapes are not sensitive to the presence of the H. This clearly depends on how much the H affects the distribution of charge. We are presently investigating H adsorption on GaAs to test this. A more plausible explanation is that  $H_2O$  acts merely to catalyze the chemisorption of O on the surface by lowering reaction barriers, in the same manner as wet oxidations of Si, and that H<sub>2</sub> leaves the surface during the reaction. We have not yet attempted mass spectrometer and flash desorption measurements to verify this.

We have interpreted<sup>(21)</sup> the spectra shown for 0 adsorption as follows. We believe that there are two competitive mechanisms active, the adsorption onto structural defects in the substrate and the adsorption onto "good" GaAs surface areas. Although both processes occur simultaneously, the former is much more rapid and thus dominates the adsorption process at low coverages. We believe that adsorption onto structural defects involves Ga atoms and causes pinning of the Fermi level and that the adsorption onto the "good" GaAs(110) surface occurs in patches and involves both Ga and As atoms. The charge configuration around the Ga atoms in this slower stage is similar to that in Ga<sub>2</sub>O<sub>3</sub>. The present results imply that the situation is similar for H<sub>2</sub>U. However, if a typical amount of water vapor is present in a vacuum system during oxygen exposures, the rapid adsorption kinetics implies that

much of the initial adsorption will be caused by  $H_2O$ . This in turn implies that the initial rate of adsorption of oxygen may be even slower than believed, (2,4,8,10,11,20) if  $H_2O$  is present in even small quantities. If water does contribute in the early stages, H may also participate in bonding at defect sites and thus cause Fermi level pinning.

A more detailed examination of Fig. 1 shows that the change in coverage with exposure is faster for  $H_20$  than for  $0_2$  at all exposures, but that the curve has a somewhat different shape than that for  $0_2$ . In particular, the initial adsorption is more rapid, and adsorption continues for  $H_20$  where it begins to saturate for  $0_2$ . Coverages greater than one monolayer are readily achievable with  $H_20$ , but require larger exposures of  $0_2$  than we were capable of making, implying some activation barrier for the latter that either does not exist for the former. As shown in Fig. 4, this differening behavior is also evident in the line shapes. Above  $\theta = 1$ , the As line shape changes dramatically and no longer corresponds to that for  $0_2$  exposures. The cause of this effect is unknown.

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In conclusion, we have summarized measurements of changes in the localized charge distribution around Ga and As atoms for the chemisorption of  $H_20$ . We have shown that these changes are very similar to those caused by oxygen chemisorption for coverages below one monolayer, but that differences occur above one monolayer. We have also demonstrated that the adsorption kinetics are two to three orders of magnitude more rapid for  $H_20$  than for  $0_2$ . Thus even small ambient-gas contamination of  $H_20$  may affect oxygen adsorption kinetics measurements. Although we have not found difference in the localized charge distribution due to  $H_20$ , the presence of  $H_20$  contamination may affect the interpretation of other electron spectroscopic measurements made for oxygen chemisorption on GaAs(110). In particular

mechanisms for Ferni level pinning and theinitial stages of adsorption proposed on the basis of results obtained at low oxygen exposures must be viewed with caution.

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## Figure Captions

Figure 1. "Oxygen" coverage on cleaved GaAs(110) resulting from exposures to oxygen (dashed line) and water vapor (solid line). The coverage is determined from the 0 KLL line, as described in the text.

Figure 2.  $M_1M_{45}V$ -derived Ga and As species-specific densities of states for different "oxygen" coverages, resulting from exposure to water vapor. The Ga and As spectra are between kinetic energies of 115 and 135 eV and between 135 and 160 eV, respectively. The total area under each spectrum is normalized to that of the clean surface. The "oxygen" coverage is listed in fractions of a monolayer. The lower scale is determined from rigidly lining up the spectra for the clean surface with the major peaks in an XPS valence band spectrum of the clean surface.

Figure 3. Ga-specific densities of states for different "oxygen" coverages resulting from exposures to water vapor (solid curve) and to oxygen (dashed curve). The areas under each spectrum are normalized to that of the clean surface.

Figure 4. As-specific densities of states for different "oxygen" coverages resulting from exposures to water vapor (solid curve) and to oxygen (dashed curve). The areas under each spectrum are normalized to that of the clean surface.









