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STUDY OF THE ELECTRONIC SURFACE STATE
OF III-V COMPOUNDS

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SEMI-ANNUAL TECHNICAL PROGRESS REPORT

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15 March 1974

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SUMMARY

A model for the surface states of the clean GaAs (110) face based on experimental and theoretical work is developed and presented in Section B under Results and Discussions. Additional results of Cs on the clean (110) GaAs face are given in Section C, and brief histories of each GaAs cleavage experiment are given in Section D. Section E mentions work to obtain quantitative information on surface roughness. Sections F and G, respectively, outline work on GaSb and Cesium Oxides.

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INTRODUCTION

The overall aim of this work is to provide basic knowledge of surfaces and interfaces necessary for the development of practical photocathodes in the 1-2 μ range. One phase of the program has started with developing understanding of the surface states on the clean III-V materials. It will progress through understanding of the effect of cesium and Cs-cesium oxide layers on the surface states. Another phase of the program will be aimed at understanding the cesium oxides themselves and the striking dependence of these oxides on small variations of cesium. Finally, the knowledge gained in these studies will be applied to aid in the engineering of practical photocathodes in the 1-2 μ spectral region.

RESULTS AND DISCUSSIONS

A. GaAs Studies

Based on work supported by this contract and that supported in the theoretical group of Professor Walter Harrison under NSF support, we believe that a comprehensive model has been obtained of the surface states on the cleavage (110) face of GaAs. The experimental and theoretical results leading to this model, as well as the model itself, are given in Section B, which is a preprint of an article which we wish to publish as soon as possible. In sections C and D, more details are given of the GaAs experiments. Other work is outlined in sections E thru G.

B. Surface State Band on the GaAs (110) Face (See attached copy of manuscript written by P. E. Gregory, W. E. Spicer, S. Ciraci, and W. A. Harrison)

C. Cesium GaAs Work

Much of the information about surface states in the past has come from Schottky barrier studies. A metal-semiconductor contact is studied, and the position of the Fermi level at the interface is found. Meade¹ has reported from such studies that the Fermi level is pinned at the surface of GaAs crystals, and this pinning implies the existence of surface states in the energy gap. Heine², however, has pointed out that the surface states found in Schottky barrier experiments may be due to the presence of the metal on the surface. The surface states in that case are really the tails of the metallic wave function penetrating a short distance into the semiconductor energy gap.

Since Meade's results could be interpreted as being in conflict with our results, we felt that it was important to investigate the effects of a metal on the GaAs surface. Cesium metal is easily applied to the surface, and has been used for years to lower the work function as an aid to photoemission studies and to produce practical photocathodes. Thus we chose to study Cs on GaAs.

We have applied Cs to each of the three GaAs samples studied (see Table I in manuscript described in Section B above) The work must still be considered to be preliminary, however. Much more work needs to be done.

Cesium was applied to the sample using commercial cesium channels manufactured by RCA. When GaAs is cesiated to produce maximum yield, believed to be one monolayer of Cs on the surface, the EDC's are dominated by a very large peak at low electron energies, and the structure near the higher energies becomes somewhat difficult to

identify. Thus we applied cesium in several stages so that structure could be followed as a function of cesium coverage.

Figure C1 shows sample 18n clean and lightly cesiated (the two curves are not normalized relative to each other). The curves are plotted relative to the Fermi level. As can be seen, structure in the two curves aligns very well. The Fermi level is pinned in the same place on the cesiated sample as it is on the clean sample.

Figure C2 shows sample 19p clean and lightly cesiated. Note that Figure C2 is plotted relative to the valence band maximum instead of to the Fermi level. The p-type sample behaves very differently from the n-type sample of Figure C1. When the peaks are aligned for the clean and cesiated p-type sample, we see that the Fermi levels are no longer aligned. The clean sample has no band bending, but the cesium apparently causes the bands to bend down by $\sim .4$ eV. Additional cesiation (not shown) causes about .1 eV additional band bending.

This result, although preliminary, appears to be in agreement with the theory of Heine² and reconciles our results with those of Meade¹ and Uebbing and Bell³. An additional difference between the n- and p-type cesiated GaAs is that the n-type GaAs exhibits the same peaks for clean and cesiated GaAs above -5 eV, but two new peaks appear on the cesiated p-type data, the peaks labeled A and B on Figure C2.

We believe peak A, which can be seen at all photon energies measured, is due to filled extrinsic surface states induced from the cesium on the surface. The origin of peak B is not understood yet. These measurements have not been repeated on another cleave, and it would appear that it is important to confirm the results by more measurements.

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D. Brief History of GaAs Cleaves and Experiments

Samples: 14n GaAs = $6 \times 10^{14} \text{ cm}^{-3}$ n-type GaAs

18n GaAs = $1.7 \times 10^{18} \text{ cm}^{-3}$ n-type GaAs

19p GaAs = $3 \times 10^{19} \text{ cm}^{-3}$ p-type GaAs

(Pressures quoted are from Redhead gauge, unless otherwise stated)

Sample 14n GaAs, Cleave #1 pressure $\sim 1.1 \times 10^{-10}$ Torr: Left sample at about 1.4×10^{-10} Torr for one month, plus several hours over 5×10^{-9} Torr, with no effects on EDCs.

Sample 14n GaAs, Cleave #2 pressure 2 to 4×10^{-11} Torr: Cleaved sample at -180°C , took data at -95°C . Found EDC shape changed when cold. Visible light on cold sample produced EDCs of same shape as room temperature. Exposed sample to oxygen. Exposures of 1L, 3L, 10L, 40L, 100L, 400L, 10^3L , 10^4L , 10^5L , 10^6L with no change in EDCs. Exposure of 10^8L produced some changes. Additional exposures of $\sim 1.5 \times 10^9\text{L}$ and 10^{12}L produced few changes beyond those already seen.

The oxygen exposures lasted over a period of about 22 days. Pressure usually dropped to $\sim 2 \times 10^{-11}$ T overnight except after the largest exposures. Some data was taken at -95°C on cleave during O_2 exposures. No differences were found from clean behavior at low temperature. A complete set of Cu data was taken for 10^6L and 10^8L .

Sample 14n GaAs, Cleave #3 deliberate "bad cleave" with notched crystal: Pressure at cleave = 1.4×10^{-11} (dropped to 8×10^{-12} next day). EDCs were smeared out somewhat. 10^7L oxygen exposure restored the usual structure (see Fig. D1). 10^3L exposure produced no further changes. No differences in surface states found between good and "bad" cleaves.

Sample 14n GaAs, Cleave #4, cleavage chip was $\approx .05$ " thick rather than .14 previously used, pressure $\sim 9.2 \times 10^{-11}$ Torr: Some of this pressure may have been As pressure due to As loose in the system from attempt to heat clean previous cleave. This cleave changed with time. Bulk structure moved about .3 eV closer to the Fermi level. After the cleave the Fermi level was ~ 1 eV above the VBM. Two days later, it was .7 eV above the VBM, the position it normally occupies. The Fermi level was at an intermediate position between the initial and final position the day after the cleave. Pressures were: at cleave, 9.2×10^{-11} ; next day, 4.5×10^{-11} ; second day, 2.4×10^{-11} ; third day, 1.8×10^{-11} . Found that shining visible light on sample at room temperature caused structure to move $\sim .15$ eV down from E_F . (No effect was found when visible light was shown on sample 14n GaAs, Cleave #2). It was also found that at 10.2 eV, changing the U.V. intensity could move the curve. Cesiumed this cleave and found no shift in position of Fermi level.

Sample 18n GaAs, Cleave #1, pressure = 5×10^{-11} . Very bad cleave. Part of face was uncleaved (20%). The EDCs were somewhat broadened with a background of scattered electrons. By covering half of LiF window could take EDCs from cleaved half only. EDCs looked smeared like deliberate "bad cleave" on sample 14n. However, 100L and 10^3L oxygen exposure did not bring back the structure. Heating to 200°C in vacuum and while exposing to 10^3L O_2 also had no effect. This cleave was much rougher looking than the deliberate "bad cleave." EDCs could be taken at LN_2 temperature (sample 14n charged up too much to permit EDCs that cold). Small (.05 eV or less) shift downward of EDCs when illuminated with visible light. Effect possibly slightly larger on cold sample.

Sample 18n GaAs, Cleave #2, pressure = 5.3×10^{-11} : First EDC after cleave was taken at 7.7 eV, then at other lower energies, in attempt to find any fast disappearing surface state near VBM. Previous cleaves EDCs were first taken at 10.2 eV. Result was negative. Made very light cesiation (work function dropped by $\sim .5$ eV). Made moderate cesiation (work function dropped by ~ 1.5 eV). Evaporated clean Cu so Fermi level could be located. Made heavy cesiation. Cu beads were exhausted so E_F could not be located, structure very weak. Heat cleaned sample to 465°C . Cesium still stuck on surface, but structure became somewhat sharper. A representative sample of EDCs from sample 18n, cleave #2, are shown in Figures D2 and D3.

Sample 19p GaAs, Cleave #1, pressure = 1.8×10^{-10} : Four days after cleave, Redhead gauge read 3×10^{-10} . Moderately cesiated sample. Saw E_F move .4 eV, two new peaks. Two days later, Redhead gauge read 2.4×10^{-10} . Cesium sample fairly heavily. E_F moved somewhat. Still saw emission up to E_F . Ten days after cleave pressure = 2.6×10^{-10} . Oxidized sample to lower the yield. Exposure was ~7L. Structure near E_F harder to see, probably still there. Data still being analysed. At end of experiment, ion gauge said 9×10^{-10} .

E. Determination of Surface Roughness

The construction of an optical system to determine the surface roughness of cleaved crystals is near completion. It is based on the work of Henzler¹ and Ibach et al². The motivation behind this project is that surface states may well be affected by the condition of the surface, and we want to have a means of judging the surface roughness.

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F. GaSb Studies

An experiment to study the (110) face of GaSb is underway. The object is to try to find surface states on GaSb, and the procedure is similar to that used in the study of GaAs.

G. Studies of Cs and Cesium Oxides

A preliminary experiment has been completed in an attempt to determine the best method of depositing cesium on a wide band gap substrate (quartz). The object was to study photoemission from Cs and Cs oxides without being concerned about photoemission from the substrate. The method used in this experiment was cooling a quartz substrate while filling the vacuum chamber with cesium from commercial cesium channels. Quartz was chosen as the substrate because of its wide bandgap and inertness. However, it also is a poor conductor of heat and apparently its front surface remained relatively warm even though its back side was cooled to near liquid nitrogen temperature. The cesium channels, in addition, are incapable of putting out large amounts of cesium. We, therefore, were unable to get the necessary several monolayers of cesium to stick. For our next experiment we shall use cesium ampoules and a metal substrate. We should be able to cool the substrate down and the ampoules should provide enough cesium for a study of cesium and its oxides. However, here we will be faced with the problem of photoemission from the metal substrate.

CONCLUSIONS

The clean GaAs cleavage (110) surface is found to have an empty band of surface states extending to the middle of the bulk band gap. These states are associated primarily with the Ga surface atoms. The filled states, associated with the As surface atoms, are located below the valence band maximum. Cs on the p⁺ GaAs sample is found to induce new filled surface states in the band gap region which pin the Fermi level. The pinning point moves away from the valence band maximum with increased Cs coverage. For the maximum Cs coverage, the Fermi level is pinned 0.5 eV above the valence band maximum.

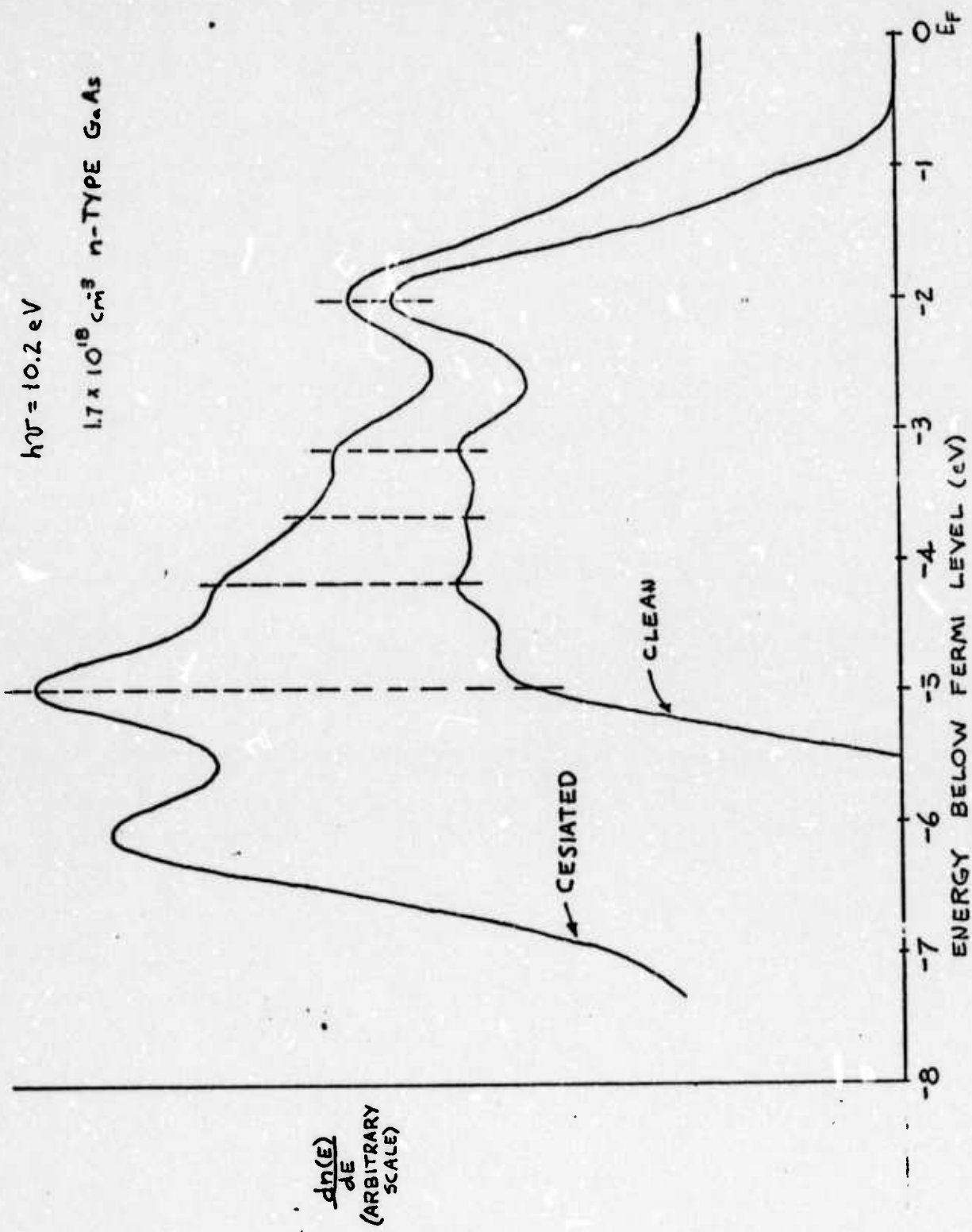


FIGURE C1

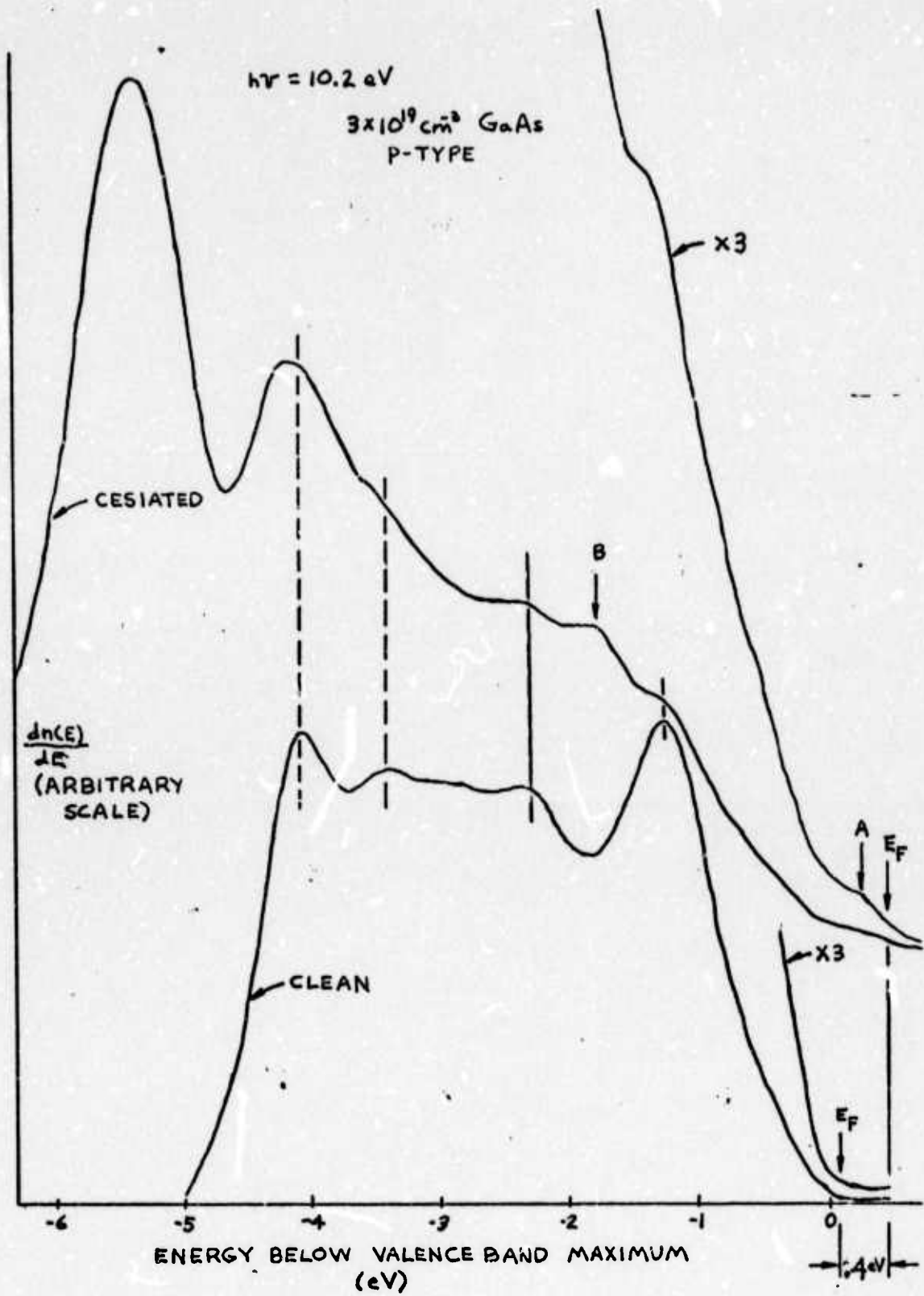


FIGURE C2

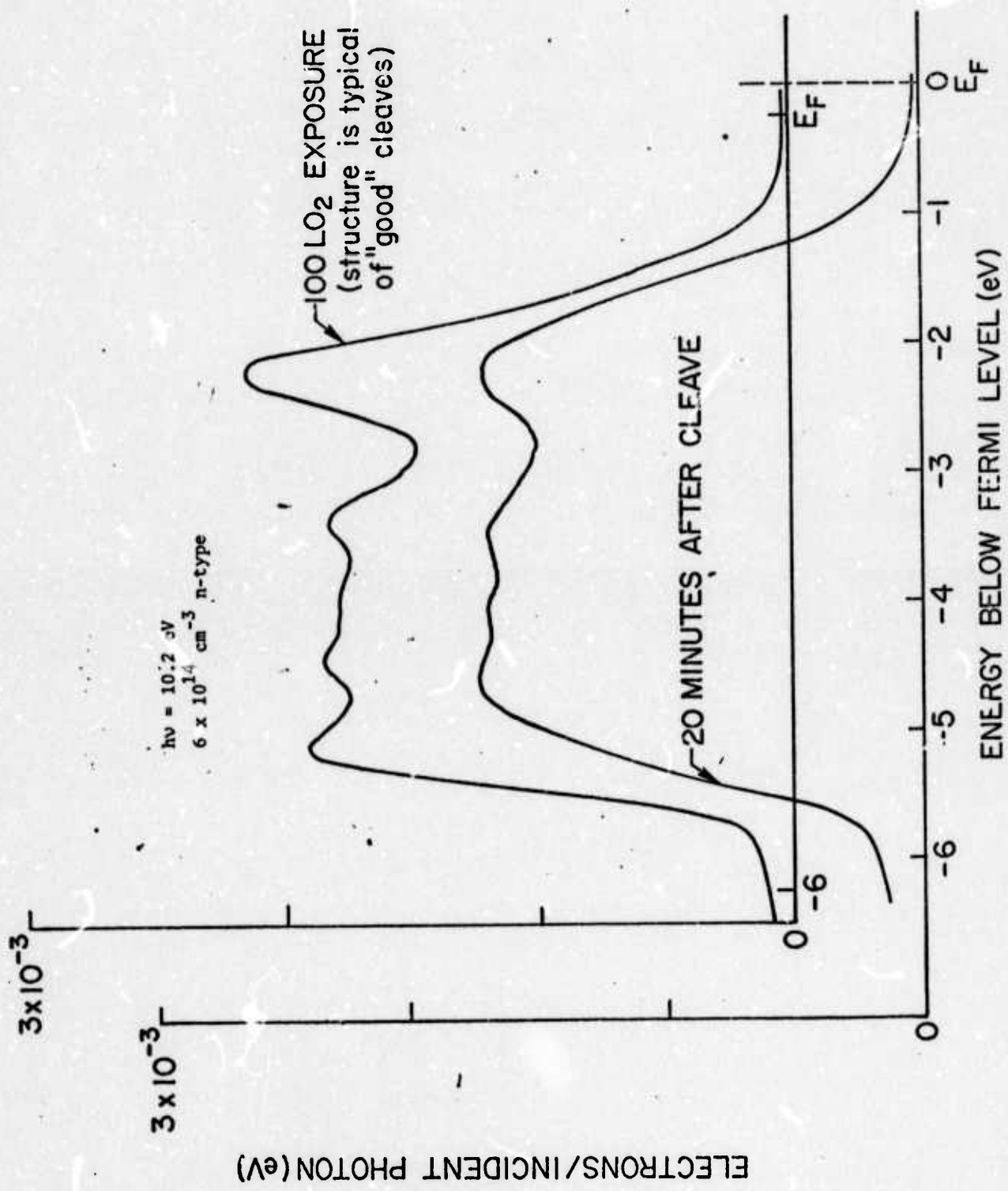


FIGURE D1

1.7x10¹⁸ n-TYPE GaAs

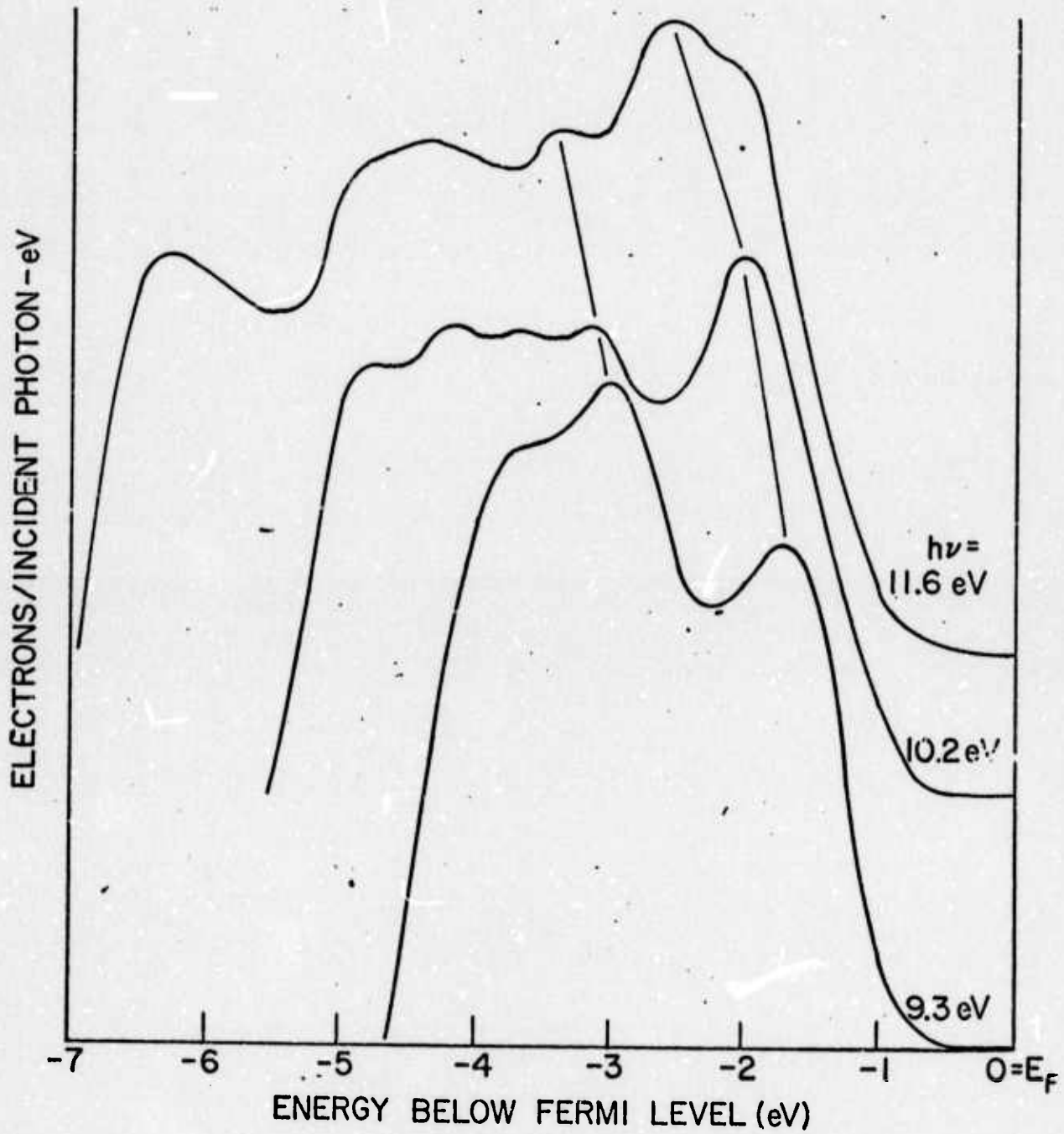


FIGURE D2

1.7×10^{18} n-TYPE GaAs

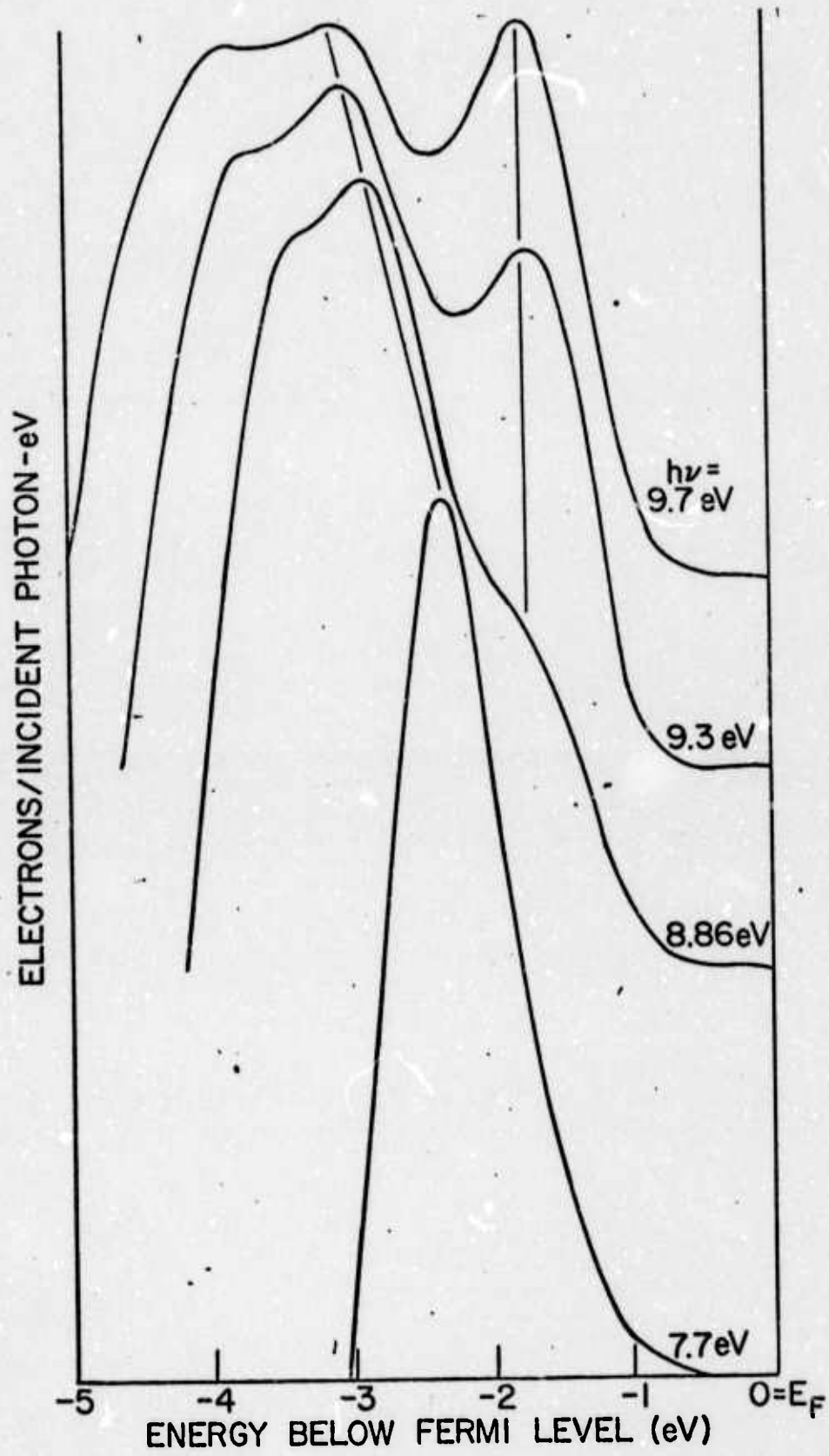


FIGURE D3

SURFACE STATE BAND ON THE GaAs (110) FACE

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ABSTRACT: Careful photoemission studies of surface states on the cleavage GaAs (110) detect no filled states in the band gap. However, empty states pin the surface Fermi level on n-type GaAs at mid-band gap. Filled states are placed below the valence band maximum and empty surface states in the upper half of the band gap. Calculations, using the Bond Orbital Model, agree with these results and associate the empty and filled bands with Ga and As, respectively.

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Recently through experimental^{1,2,3} and theoretical⁴ studies, major advances have been made in the understanding of Si surface states. Through photoemission studies, a good measure of the density of filled states^{1,2,3} and their dependence³ on crystal face has been obtained. However, contradictory results have been reported concerning the clean GaAs cleavage (110) plane. Eastman and Grobman² (EG) have reported filled surface states lying in the bottom half of the band gap; whereas, Van Larr and Scheer⁵ (VLS) and Dinan, Glabraith, and Fischer⁶ (DGF) found no filled states there. Here we report: (1) very careful experimental work which shows the bottom half of the valence band to be free of surface states (i.e., the filled states lie below the valence band maximum) and empty surface extending down to the middle of the band gap, in agreement with DGF; and (2) theoretical work which explains the large band gap between the filled and empty surface states and

relates this to the electronic population of surface atoms. Brief comments are made concerning the effects of oxygen and of Cs on the surface states and surface atoms.

The surface of GaAs has been studied by ultraviolet photoemission spectroscopy, using a technique similar to that used by Wagner and Spicer¹ to study Si surface states. The cleaved (110) surfaces were usually very smooth, but two roughly cleaved surfaces (i.e., surface containing many steps) were purposely made. The samples studied are described in Table 1. Energy distribution curves (EDCs) are given in Figs. 1a and b. In most cases, the pressure was below 5×10^{-11} Torr, and measurements begun within 15 to 30 minutes after the cleave.

For GaAs as Si, three methods were used to identify surface states. In one, the cleaved surface was exposed to oxygen and examined for structure near the band gap which is preferentially affected. In contrast to Si, where strong changes due to removal of surface states were found, oxidation did not preferentially remove any states in the band edge (see Fig. 1b). This suggests the absence of normally filled surface states. Another test, is to look for surface state pinning as a function of bulk doping. As Fig. 1a illustrates, no pinning was found on p⁺⁺ GaAs. This indicates the absence of normally filled (donor) surface states. For the n-type samples, the Fermi level was pinned at midgap independent of doping. This indicates normally empty (acceptor) surface states extending down to midgap.

The third test is to identify structure in the EDCs which cannot be explained in terms of bulk band structure⁷. EDCs were taken every few tenths of an eV from 5.6 to 11.8 eV and no such structure was observed. The position in energy of all observed structure was found to depend on $h\nu$ as expected, if due to bulk band structure. No structure was found; such as that in Si, whose energy was independent of $h\nu$. Note in Fig. 1a that for the n-type samples, there is no emission originating from near the Fermi level and that for p-type, the Fermi level comes at the valence band maximum. This gives further evidence against normally filled (donor) states

lying in the band gap.

Our GaAs data is consistent with the model shown in Fig. 2. For sake of comparison, the Si (111) surface states have also been indicated. Note that the GaAs surface states are split into a normally filled and empty band separated by an appreciable band gap. Within our experimental accuracy (± 0.1 eV), we have obtained no evidence that the filled band extends into the band gap.

Since our results differ from those of EG, we have taken extreme care in measurements and have studied a number of samples⁸. Our results are in good agreement with DGF and, in concluding that the filled band lies below the valence band gap, agree with VLS.

The Bond Orbital Model⁹ for the electronic structure of tetrahedrally coordinated solids gives us a simple approximate way of estimating the bands, and surface bands for these materials. We begin with silicon and construct energies for individual sp^3 hybrid on each atom as illustrated in Fig. 3a. We then form bond orbitals lowered in energy by $(1-S)V_2$ and antibonding orbitals higher by $(1+S)V_2$ in each bond with $V_2 = 2.2$ eV in silicon. The surface, or dangling, hybrids do not, of course, form bonds. The bonding states then broaden into valence bands of width $4V_1$, with $V_1 = 1.4$ eV for silicon. The formation of conduction bands is not so simple but we take the conduction band edge above the top of the valence band by the observed gap of 1.14 eV. This leaves the dangling bond states, which broaden (due to coupling between dangling bonds through the back-bonds) into two bands, degenerate at certain symmetry directions of the surface Brillouin Zone¹⁰. (Surface reconstruction would split the occupied and unoccupied surface bands, but we will not consider that here.)

The electronic structure of GaAs is similarly constructed but begins with two types of hybrids, one for Ga and one for As, differing in energy by $2V_3$ with $V_3 = 1.2$ eV. The resulting bands, one completely in the bulk valence band and occupied, and the other one empty and in the energy gap, are as shown in Fig. 3b. As polarity increases in going to II-VI and I-VII compounds, V_3 will increase and therefore the gap between surface states could increase further in such materials.

In Fig. 4, we indicate in real space the approximate bulk and surface configurations deduced from this work. In the bulk, the Ga and As atoms have a rearrangement of charge because of the formation of the bond. At the surface, one of the four covalent bonds per atom is broken and three remain. As a result, the net charge relaxes towards zero; i.e., a surface As has approximately five and Ga approximately three valence electrons. Each type of surface atom contributes three electrons to the covalent bond. This leaves each surface As with two excess electrons which form the filled surface band lying below the top of the valence band. All three Ga electrons are taken up in the covalent bond leaving none to populate surface states; thus, the empty surface band is formed primarily from the Ga states.

Brief mention will be made of studies of the oxidation of GaAs, since this bears on the above mentioned model. A more complete report will be given elsewhere. In studying n-type GaAs, we found a very striking result: within ± 0.1 eV, the pinning of the Fermi level by the empty surface states was unchanged by oxidation (see Fig. 1b). This is in strong contrast to Si where the surface state pinning is destroyed by oxidation¹. We suggest that this result is due to oxygen combining with the two surface As electrons, reducing the energy of those electrons and removing the filled band while leaving the surface Ga atoms, and thus the empty surface band largely unaffected. We would, thus, expect the oxygen atoms to be located roughly over the As atoms and a stable monolayer to be formed with 50% atomic coverage of the GaAs corresponding to oxidation of only the As surface atoms.

One also needs to relate the surface states reported here for clean GaAs to the Schottky barrier work on GaAs metal interfaces where surface state pinning about 0.5 eV above the top of the valence band is typically found.¹¹ We have done preliminary work with Cs on p⁺⁺ GaAs and find that the Cs produces surface states in the band within 0.5 eV of the valence band maximum. In particular, we find that these states pin the surface Fermi level about 0.5 eV above the valence band maximum in agreement with Uebbing and Bell.¹² We believe that extensions of this work will have important implications for practical "negative electron affinity" photocathodes.¹³

TABLE I

<u>Sample</u>	<u>Doping</u>	<u>Bulk Fermi level Position above V_m(eV)</u>	<u>No. of Cleaves Studied</u>
18 n	1.7 x 10 ¹⁸ cm ⁻³ n-type	1.4	2
14 n	6 x 10 ¹⁴ cm ⁻³ n-type	1.2	4
19 p	3 x 10 ¹⁹ cm ⁻³ p-type	0	1

References

Useful discussions with Dean Eastman, Traugott Fischer, H. Lüth, and Jack Rowe are gratefully acknowledged.

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† Work supported in part by NSF

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FIGURE CAPTIONS

- FIG. 1a EDC for $h\nu = 10.2$ eV for n- and p-type GaAs samples. The zero of energy is taken at the valence band maximum. E_f indicates the Fermi level position.
- 1b Effect of oxygen exposure on n-type GaAs. Note that the Fermi level (E_f) is unaffected by the oxidation.
- FIG. 2 Models for surface states on the cleavage faces of Si and GaAs.
- FIG. 3 Diagram showing the formation of bulk (bold lines) and (110) surface (dashed lines) band structure of tetrahedrally coordinated semiconductors.
- a) Silicon Energy parameters $V_1 = 1.4$ eV, $V_2 = 2.2$ eV, $V_3 = 0$
- b) GaAs Energy parameters $V_1 = 1.7$ eV, $V_2 = 2.1$ eV, $V_3 = 1.2$ eV
- FIG. 4 GaAs surface and bulk atoms. At the surface, Ga has approximately three valence electrons and As approximately five. The filled surface band is associated principally with the As surface atoms; the empty surface band principally with the Ga surface atoms.

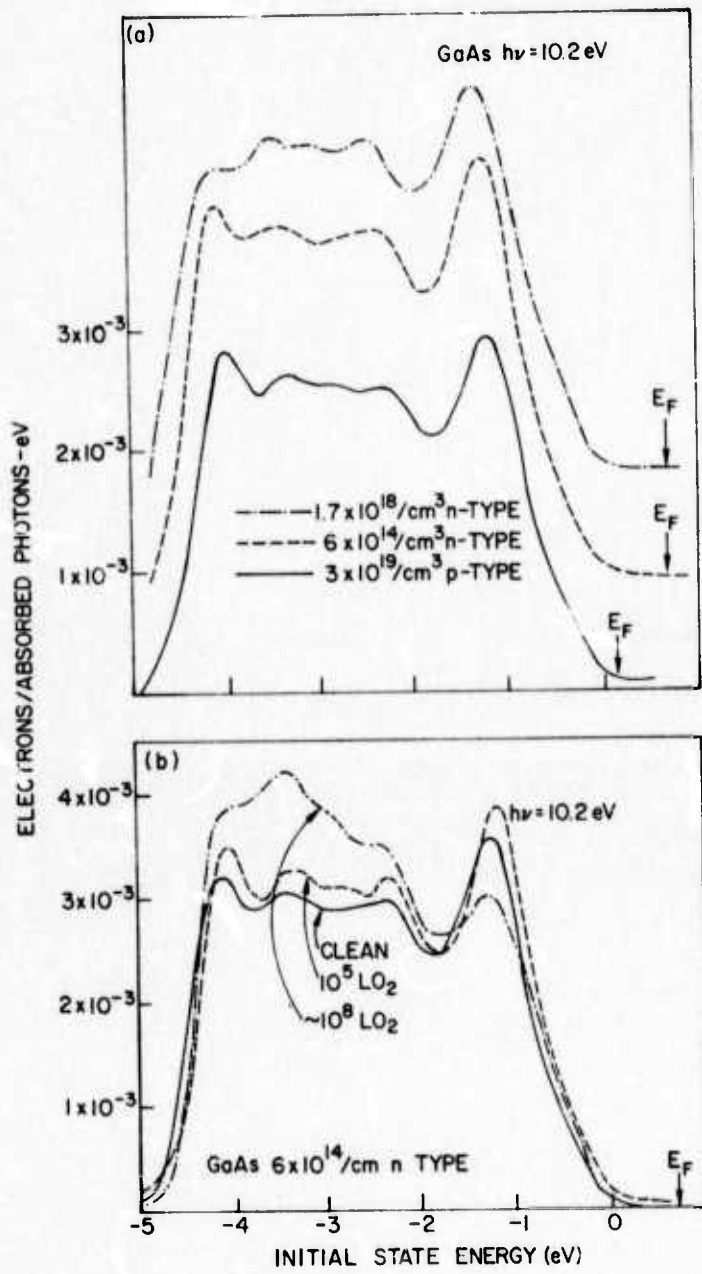


Fig. 1

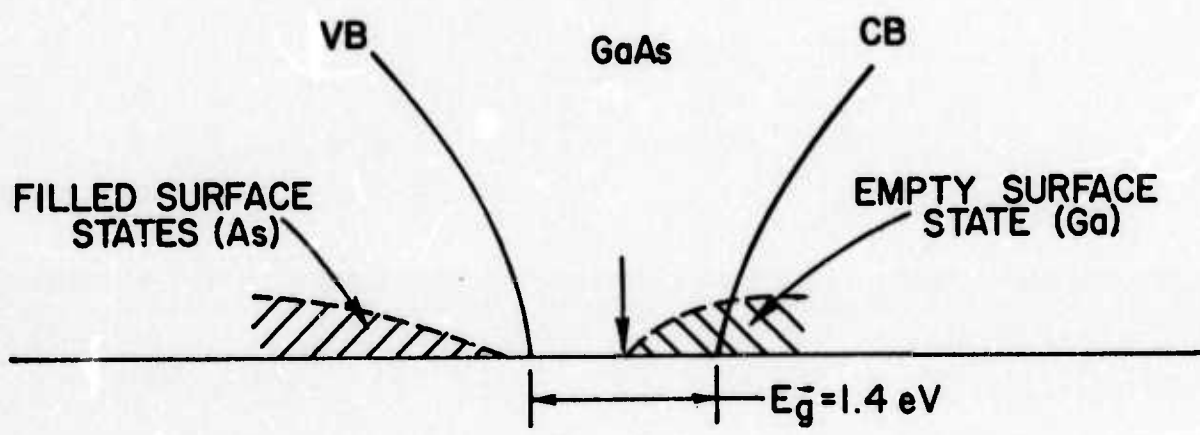
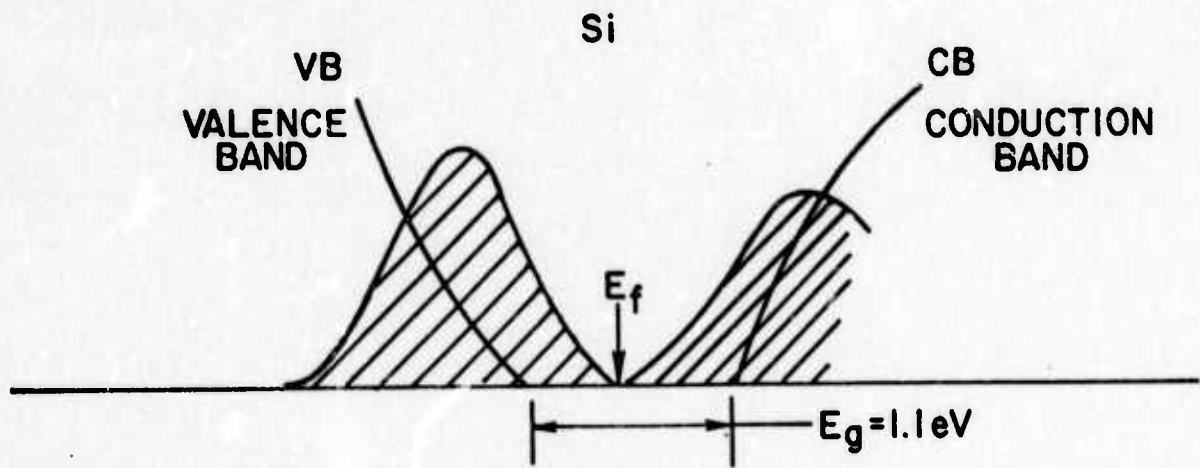


Fig. 2

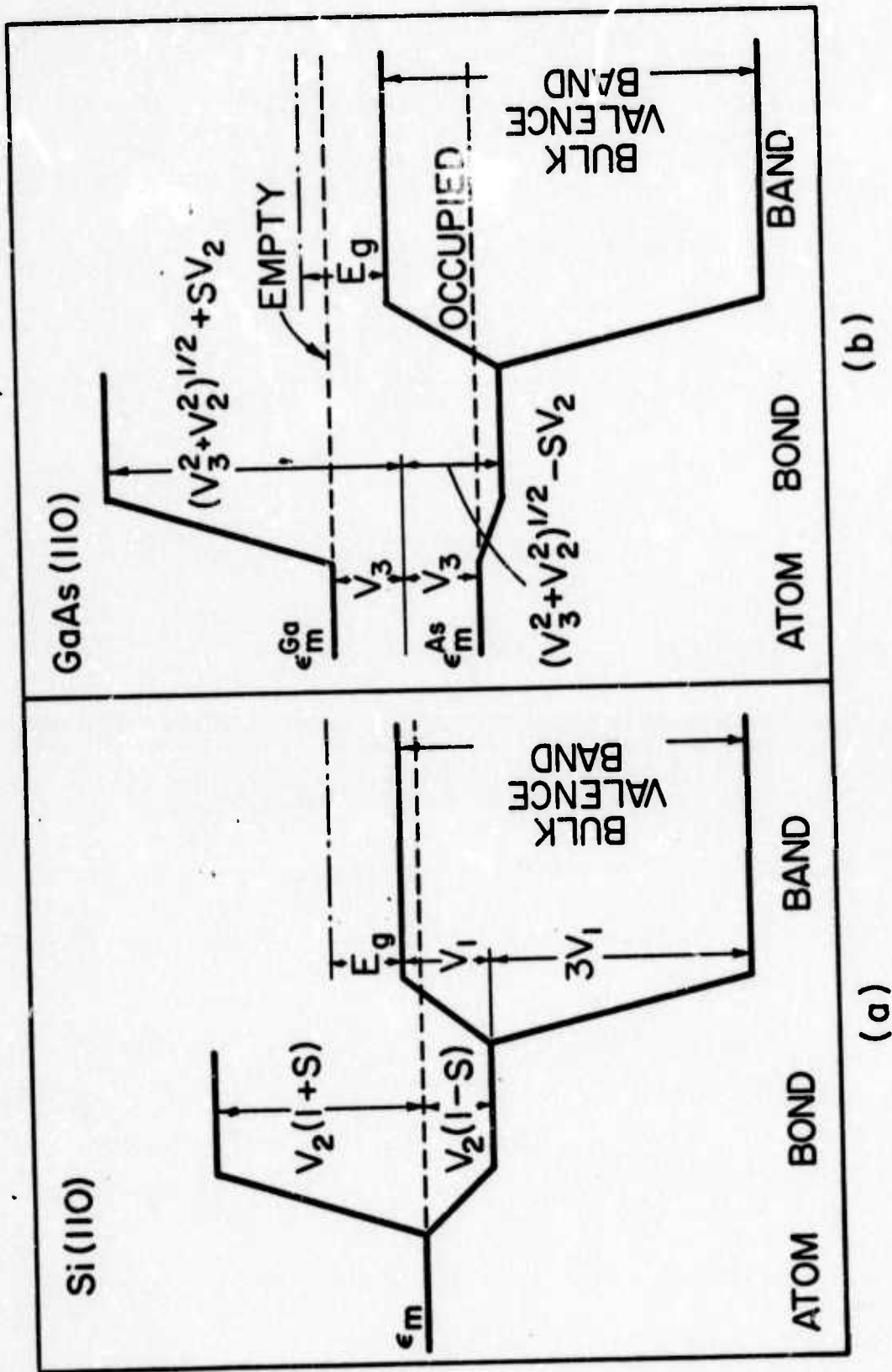


Fig. 3

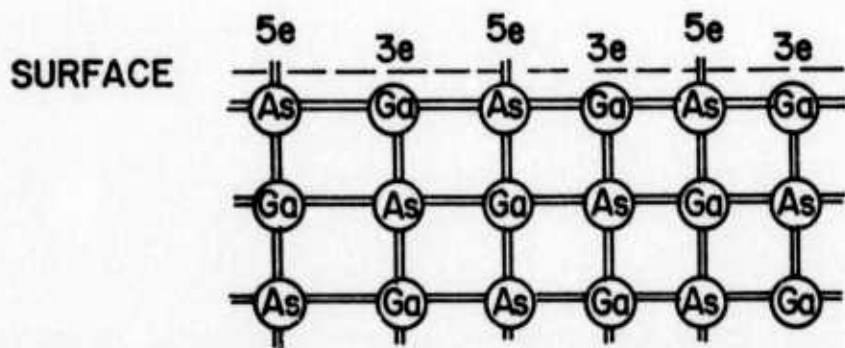


Fig. 4