

## Effect of an Al interlayer on the GaAs/Ge(100) heterojunction formation

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The chemistry, structure, and growth kinetics of epitaxial GaAs/Ge heterojunctions are controllably modified using an Al interlayer of one to two monolayers in thickness. Photoemission spectroscopy is used to investigate the interface formation with and without the Al interlayer. Although the valence-band and core-level spectra indicate dramatic changes of the chemistry and structure caused by the Al (deposited on the substrates both at room temperature and at 340°C) at the interface, the band-structure lineup is not affected. The Fermi level in the gaps is influenced by both the presence of the Al interlayer and the deposition temperature. The Fermi level moves toward the valence band by 0.15 and 0.3 eV (relative to the GaAs  $c(4\times 4)$ /Ge degenerate  $n$ -type interface) for room-temperature and 340°C deposition, respectively. The Fermi-level position is simply related to the amount of As diffusion into the Ge layer and its role as an  $n$ -type dopant. The results support the conclusion that the band offset is primarily an intrinsic (bulk) property which is insensitive to interfacial charge distribution or chemistry to within  $\pm 0.05$  eV.

### INTRODUCTION

Over the past two decades, band offsets at heterojunction interfaces have been studied extensively.<sup>1-9</sup> A study of a large ensemble of interfaces has shown that the local microscopic effects at the interface contribute to the band offsets by at most 0.15–0.2 eV.<sup>5</sup> If it were truly this large and controllable, such a contribution could provide a tuning capability for the band offsets of up to  $8kT$  at room temperature. It is therefore essential to examine in detail the magnitude of band-discontinuity variations and to understand the origin of possible interfacial contributions.

The effect of interfacial properties such as the overlayer morphology, the substrate-surface orientation, and the chemistry and structure of the substrate-surface layer on the band offsets has been investigated for prototypical lattice-matched heterojunctions such as Ge/GaAs, Ge/ZnSe, GaAs/ZnSe, and Si/GaP.<sup>1-9</sup> An extensive study of GaAs/Ge(100) interfaces grown by molecular-beam epitaxy showed that the band offsets are insensitive to such factors; this study concluded that the contributions from changing the stoichiometry and the structure of the substrate surface layers, and from introducing an overpressure of As<sub>4</sub> during the interface formation were within the experimental uncertainty, which was  $\pm 0.05$  eV.<sup>8</sup>

The ability to adjust the band offsets by intentionally introducing extrinsic species at the interface remains a question of great interest. The observed insensitivity of the band offsets in the GaAs/Ge system demonstrated that the tunability of the band offsets cannot be achieved by varying the intrinsic constituents of the semiconductor forming the heterojunction.<sup>8</sup> This led us to study the effect of an interlayer on the GaAs/Ge heterojunction. The modulation of the interfacial chemistry and interdiffusion by introducing an adatom interlayer has been reported to affect Schottky-barrier formation and semiconductor oxidation processes.<sup>10-13</sup> The strong bonding of Al-As, the

maintenance of the GaAs and Ge lattice constant by the formation of AlAs, and the doping nature of Al in Ge led us to choose Al as an interlayer. The strong reactivity of Al with As should result in a Ga-rich and perhaps AlAs-stabilized interface. If Al atoms diffuse outwards, they should act as an acceptor in Ge. In addition, AlAs is lattice matched to  $\pm 0.1\%$  to both Ge and GaAs and thus could form a chemically foreign but structurally continuous interface. Therefore, whether Al diffuses outwards or stabilizes the As at the interface, it should change the chemistry and the charge redistribution on both sides of the junction compared to that of the GaAs/Ge junction alone. These changes will also demonstrate the sensitivity of the band offsets of unintentional local contaminations present in the monolayer range of thickness at the interface.

### EXPERIMENT

Buffer layers of the order of 0.5  $\mu\text{m}$  were grown by molecular-beam epitaxy (MBE) on a sputter-annealed GaAs(100) substrate. The substrate preparation procedure and growth parameters are described elsewhere.<sup>8</sup> The ordering of the surface layer was determined *in situ* by low-energy electron diffraction. The  $c(4\times 4)$  surface reconstruction of the GaAs(100) was used in our study. While the exact stoichiometry of GaAs  $c(4\times 4)$  is currently of considerable controversy, it is characterized by at least a full monolayer with perhaps an additional one-quarter of a monolayer of  $a$ -As on this As-capped surface. Both the Al and Ge deposition rates were calibrated using a quartz-crystal thickness monitor. The Al interlayer was evaporated from a boron nitride cell at the rate of 1 Å/min. A thickness equivalent to a monolayer of Al was deposited on the substrate at room temperature and at 340°C. Epitaxial Ge overlayers were deposited at the rate of 2 Å/min on a 340°C substrate for both types of Al interlayer. The pressure during the Al and Ge deposition was maintained in the  $(5-9)\times 10^{-10}$  Torr range. To en-

sure minimum oxidation of the Al-covered surface, it was quickly investigated (within an hour of preparation at  $< 2 \times 10^{-10}$  Torr) and covered with a monolayer of Ge.

A grasshopper monochromator (4° beam line at the Stanford Synchrotron Radiation Laboratory) provided soft-x-ray photons to probe the MBE-grown GaAs surface and subsequent heterojunction interface. A PHI double-pass cylindrical mirror analyzer was used to obtain valence-band and core-level spectra of the GaAs, Al, and Ge. The overall experimental resolution was 0.2 eV for the core levels and 0.45 eV for the valence band.

## RESULTS

Figure 1 shows the valence-band spectra for the clean MBE-grown GaAs(100)  $c(4 \times 4)$  surface and for the Al-covered surfaces at different substrate temperatures. The effect of the substrate temperature during deposition is noticeable in the upper two curves. For the room-temperature deposition, the valence band shows a contribution from the Al metallic states which fill the heteropolar gap at 4.3 eV below the valence-band edge. For the 340°C deposition, the first peak below the valence-band edge narrows, indicating the loss of As, and the second peak broadens, indicating the formation of Al-As bonds. While the room-temperature deposition results in the formation of small Al clusters, the 340°C deposition results in the formation of Al-Ga-As alloys. In neither case is an O(2*p*) resonance observed at  $\sim 6.4$  eV below the top of the valence band.<sup>14</sup> Because of the large cross section for O on Al at these photon energies, we estimate that possible O contamination of the heated surface prior to heterojunction growth is less than 1% of a monolayer.

The attenuation of the Ga(3*d*) intensity with the Al

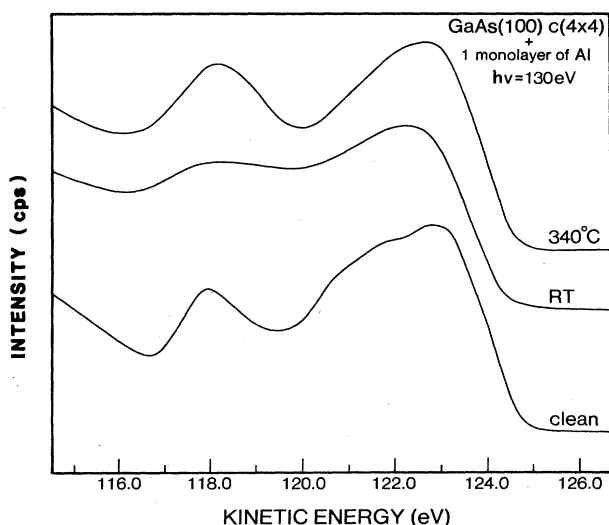


FIG. 1. Valence-band energy distribution curves taken with 130 eV photon energy for the clean and Al-covered GaAs(100)  $c(4 \times 4)$  surface. One monolayer of Al was deposited on the substrates both at room-temperature and at 340°C. The upper two curves show the distinct differences between the two deposition temperatures. While the room-temperature deposition indicates the formation of Al clusters, the 340°C deposition temperature results in the formation of Al-Ga-As alloys.

overlayer shows a temperature dependence. The sharp decrease in the Ga(3*d*) signal after the deposition of one monolayer of Al at room temperature suggests the formation of Al clusters. Assuming an escape depth of 5 Å, we estimated the clusters to cover two-thirds of the surface. The slight change in the Ga(3*d*) signal when the deposition temperature is increased to 340°C suggests the formation of Al-Ga-As alloys. Figure 2 shows a plot of the normalized core intensities as a function of Ge coverage on a semilog scale. For all cases, the Ga(3*d*) curve follows a straight line. The slope of the line for the GaAs/Ge interface with the Al interlayer is slightly larger than that for the GaAs/Ge interface alone, i.e., 4.5 versus 4.0 Å. This is probably due to the interaction between Al and GaAs. Nevertheless, the Ga(3*d*) curves suggest the confinement of the Ga atomic species to the interfacial region or minimal diffusion of Ga into the Ge overlayer. In contrast, the As(3*d*) curve indicates a diffusion of the As atomic species into the Ge overlayer. Notice that the Al(2*p*) curve is temperature dependent. The curve for the

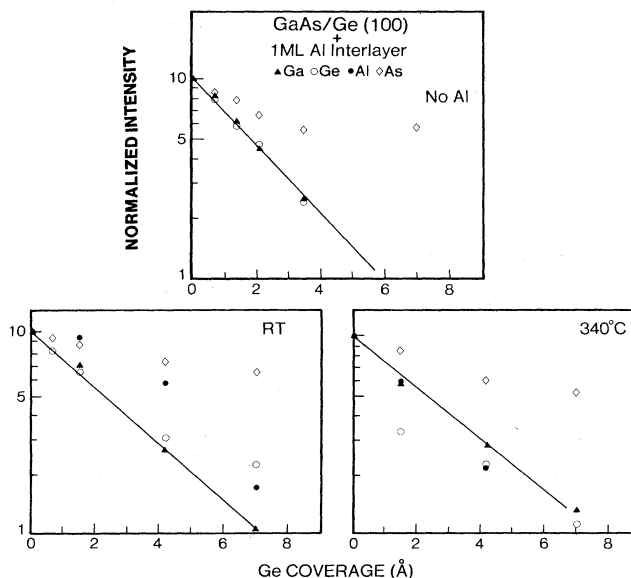


FIG. 2. Evolution of the Ga, As, and Al core intensities are plotted on a semilog scale as a function of Ge coverage. For the Ga(3*d*) and As(3*d*) peaks, the core intensity is normalized to the starting value after the Al deposition; for the Al(2*p*) peak, the core intensity is referred to the freshly evaporated Al layer. Notice that the Ga(3*d*) attenuation rate is similar for the three interfaces studied here. This suggests that the Ga atomic species are confined to the interfacial region regardless of the Al interlayer and the Al deposition temperature. On the other hand, the As atomic species diffuses out through the Ge thin film. The Al(2*p*) peak attenuates at different rates depending on the Al deposition temperature. The room-temperature Al deposition case suggests the formation of Al clusters and minimal interdiffusion, while the 340°C Al deposition indicates that Al diffuses into GaAs with formation of Al-Ga-As alloys. The Ge(3*d*) core intensity is referred to the thick Ge layer. To allow direct comparison of the overlayer with the attenuating substrate species, the Ge(3*d*) data is plotted as 1—the normalized intensity vs coverage. ML denotes monolayer.

room-temperature Al deposition supports the formation of Al clusters with Ge filling between the clusters first. Also, the curve suggests minimal diffusion of Al into the Ge thin film, while the curve for the 340°C Al deposition suggests that Al has reacted with and diffused into the GaAs surface.

The influence of the Al interlayer on the evolution of the As, Ge, and Ga atomic species is best understood by plotting their core-level intensities relative to the Al(2*p*) intensity as a function of Ge coverage. Figure 3 shows a plot of the intensity ratios  $I_{Al}/I_{Ga}$ ,  $I_{Al}/I_{Ge}$ , and  $I_{Al}/I_{As}$  for the GaAs(100)  $c(4 \times 4)$  starting surface with the Al interlayer deposited at the two different temperatures. For the room-temperature deposition,  $I_{Al}/I_{Ga}$  and  $I_{Al}/I_{As}$  increase, while for the 340°C deposition temperature, they decrease with increasing Ge coverage. The room-temperature deposition case can be explained by assuming that Ge first fills the area between the Al clusters. An alternate explanation is the diffusion of Al into the Ge layer. However, the latter explanation is less probable, as suggested by Fig. 2. Apparently, the 340°C deposition temperature causes the anion-rich GaAs(100) surface to turn into a cation-rich-like surface. The interface formation in this case is similar to that of a Ga-rich GaAs(100) starting surface.<sup>9</sup>

The Fermi-level position in the gaps is influenced by the presence of the Al interlayer and the deposition temperature. For the GaAs  $c(4 \times 4)$ /Ge(100) interface alone, the Fermi level is found to be degenerate with the Ge conduction-band minimum.<sup>8,9</sup> For the same system with an Al interlayer deposited on a room-temperature substrate, the Fermi level moves by 0.15 eV towards the valence band while, when the deposition temperature is increased to 340°C, it moves by 0.3 eV in the same direction. In fact, the Fermi-level position for the latter case is

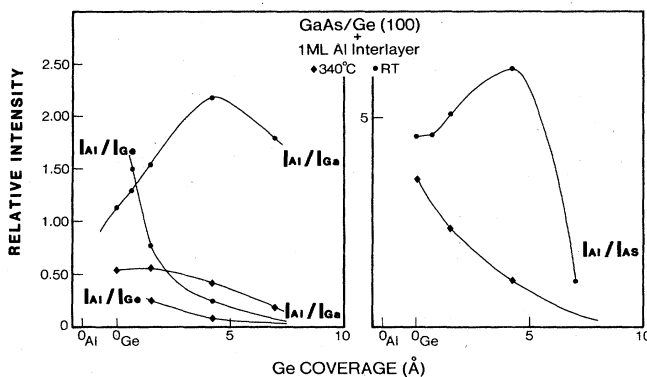


FIG. 3. Intensity ratios  $I_{Al}/I_{Ga}$ ,  $I_{Al}/I_{Ge}$ , and  $I_{Al}/I_{As}$ , as obtained from the Al(2*p*), As(3*d*), Ga(3*d*), and Ge(3*d*) peak areas, are plotted as a function of increasing Ge coverage. The  $I_{Al}/I_{Ga}$  and  $I_{Al}/I_{As}$  ratios increase with increasing Ge coverage for room-temperature deposition. This can be explained by the formation of Al clusters and/or the diffusion outwards of Al. The decrease in  $I_{Al}/I_{Ga}$  and  $I_{Al}/I_{As}$  for the 340°C deposition suggests that Al has reacted with the GaAs substrate forming Al-Ga-As alloys. The evolution of the  $I_{Al}/I_{Ge}$  ratio with increasing Ge coverage for both deposition temperatures is consistent with the above conclusions.

similar, within  $\pm 0.05$  eV, to that for GaAs/Ge(100) with a Ga-rich starting surface. This again suggests that the Al in this case changes the anion-rich surface to a cation-rich surface and reduces the As *n*-type doping of the growing Ge overlayer. The change in the Fermi-level position for room-temperature deposition suggests that either the Al acts as a chemical trap for As thus lowering the As incorporation in the Ge thin film, or the outdiffused Al acts as a *p*-type dopant, thus partially compensating the As *n*-type doping effect in the Ge layer. These observations are consistent with the conclusions in the previous paragraphs.

To measure the band offset, we basically need to determine the interfacial position of the valence-band edge of both semiconductors. The valence-band edge on the substrate side is determined by measuring the Ga(3*d*) peak position. The Ga(3*d*) shift with increasing coverage of Ge saturates after 5 Å of Ge. Up to such coverage, the Ga(3*d*) line shape does not change; broadening of the line shape occurs at a coverage above 7 Å. On the overlayer side, the Ge(3*d*) position is used. Because the overlayer is thin, we expect it to be fully depleted. Therefore, the Ge(3*d*) peak position is expected to track the valence-band edge once the electronic band structure is fully developed. Our observation, which is supported by theoretical calculation, indicates that the valence-band electronic structure is fully developed after a 5 Å overlayer thickness. Hence, the distance in energy between the Ga(3*d*) and the Ge(3*d*) position is related to the valence-band offset by subtracting a constant number. This constant number is obtained from the difference between the Ga(3*d*) and Ge(3*d*) binding energies relative to their respective valence-band edges as measured in the bulk semiconductors. Therefore, changes in this measured difference from one system to the other are indicative of changes in the valence-band offset. The nature of our interlayer experiment made it difficult to apply the method in Ref. 5. In prior studies,<sup>15</sup> however, we found that these two methods yield the same value of  $\Delta E_v$  to within 0.07 eV.<sup>8</sup>

Table I lists the measured difference between the Ga(3*d*) and the Ge(3*d*) binding energies for three interfaces. The difference is the same for the three interfaces within our experimental uncertainty of  $\pm 0.05$  eV. As in earlier work, the uncertainty is based on reproducibility among systematic experiments on the same system. The constancy of the band offset for the GaAs/Ge interface in spite of the Al interlayer is surprising. Although the chemical structure of the interfacial region is clearly affected by the presence of the Al interlayer, the band offset

TABLE I. The measured energy separation between the Ga(3*d*) and Ge(3*d*) peak positions for the interfaces we studied. Notice that the energy separation is the same for the three interfaces within our experimental uncertainty of  $\pm 0.05$  eV.

GaAs(100) $c(4 \times 4)$ /Ge Interlayer	$\Delta E_B^{Ga-Ge}$ (eV)
No Al	10.30
Al deposited at room temperature	10.27
Al deposited at 340°C	10.28

is not. This insensitivity in  $\Delta E_p$  is measured for Al interlayer thickness up to two monolayers (the maximum Al coverage we studied).

## DISCUSSION

The starting GaAs(100)  $c(4 \times 4)$  surface and its interface with the Al overlayer is very complex. The literature on the GaAs/Al interface presents a wide variety of explanations for the chemistry and the paths through which the ultimate interface structure evolves.<sup>16–18</sup> This confusion stems from our lack of complete understanding of the geometry of the GaAs(100)  $c(4 \times 4)$  surface reconstruction, which presents an additional difficulty in determining a detailed description of the Al overlayer. The least knowledge of our As<sub>4</sub>-prepared GaAs(100)  $c(4 \times 4)$  surface is of an ordered As monolayer plus  $0.25 \pm 0.1$  monolayers of amorphous As on the free surface.<sup>9</sup>

Although we did not intend to study the GaAs/Al interface, our results illuminate some of the key issues for that interface. The type of reactions that occur between the deposited Al overlayer and the substrate are temperature dependent, as noted by other authors.<sup>16–18</sup> Our experimental observations suggest a nucleation of Al when deposited at room temperature.<sup>17</sup> The sharp decrease in the Ga(3*d*) signal and the broadening of the As(3*d*) line shape suggest that no interdiffusion occurs in this case. The evolution of the core levels when Al is deposited at 340°C is distinctively different. The As(3*d*) line shape narrows and the Al(2*p*)-As(3*d*) energy difference indicates the formation of an Al-rich Al-Ga-As alloy.<sup>19,20</sup> Also, the slight change in the Ga(3*d*) signal indicates a diffusion of Al into the substrate. For both deposition temperatures, the change in the Ga(3*d*) linewidth precludes the formation of free metallic Ga upon Al deposition. In fact, this is expected because the GaAs(100)  $c(4 \times 4)$  surface exhibits an excess of a monolayer of surface As. Also, the chemical shift in the Ga(3*d*) position due to the formation of Al-Ga-As alloys is very small.<sup>19,20</sup>

The evolution of the interface formation with increasing Ge coverage provides further insight into the GaAs/Al interface. The interface formation for the two deposition temperatures is distinctively different. Important to note is that in all cases the Ge overlayer was deposited at 340°C. Therefore, both room-temperature- and 340°C-deposited Al interlayers were annealed 10–15 min before the deposition of the Ge overlayer. Apparently, such annealing is not sufficient to change the GaAs/Al interfacial chemistry at room-temperature Al deposition to resemble that at 340°C Al deposition.

Contrary to the case of the GaAs(110):Al/Ge interface,<sup>13</sup> the  $I_{\text{Ga}}/I_{\text{As}}$  ratio is independent of the Al interlayer and its deposition temperature. For both deposition temperatures, As segregates to the free Ge surface in spite of the strong bonding of Al-As. Based on the  $I_{\text{As}}/I_{\text{Ge}}$  intensity ratios, the amount of segregated As is the same, independent of the Al interlayer and the deposition temperature.<sup>2,6</sup> As for the intrinsic GaAs/Ge heterojunction, the interface formation process is dominated by surface GeAs<sub>x</sub> growth, which for the (100) orientation always exhibits the same surface reconstruction, two domain

$2 \times 1$ .<sup>21</sup>

The increase in the  $I_{\text{Al}}/I_{\text{Ga}}$  and  $I_{\text{Al}}/I_{\text{As}}$  ratios with increasing coverage of Ge in Fig. 3 for the room-temperature deposition indicates the presence of Al clusters from the initial monolayer. The Al is reacting with the topmost GaAs layer in this case and does not diffuse into the GaAs. This also is supported by the evolution of the Al(2*p*) intensity as function of Ge coverage in Fig. 2. In contrast, for the 340°C deposition temperature, the  $I_{\text{Al}}/I_{\text{Ga}}$  and  $I_{\text{Al}}/I_{\text{As}}$  ratios decrease with increasing Ge coverage as shown in Fig. 3. The Al in this case has completely reacted and interdiffused with the GaAs surface. For both Al-deposition temperatures, the interface exhibits a small percent of Ga in some bonding configuration different than that of GaAs, as indicated by the broadening of the Ga(3*d*) line shape at Ge coverages above 6 Å. Both the broadening of the Ga(3*d*) line shape at thick Ge coverages and the presence of a monolayer of As at the free Ge surface regardless of the Al-deposition temperature suggest that even if the Al monolayer completely reacts it does not form a spatially uniform Al-Ga-As layer. These observations indicate that the Ga-As bond is easier to break than the Al-As bond, as one would expect.

The Al interlayer does not only influence the interfacial chemistry, it also influences the Fermi-level position in the gaps. For room-temperature deposition, the Fermi level moves by 0.15 eV toward the valence band, while for 340°C deposition, it moves by 0.3 eV. The presence of the Al interlayer does not pin the Fermi level as was observed for the GaAs(110)/Al interface.<sup>22</sup> Clearly, the deposition-induced defects<sup>22</sup> created during the GaAs(100)/Al interface formation do not occur in sufficient density to cause a complete pinning of the Fermi level. This observation is consistent with previous results on the GaAs/Ge(100) interface.<sup>9</sup>

The main issue that concerns us here is the band offsets and how they are affected by the changes in the interfacial chemistry and charge distribution. Figure 4 shows a schematic of the energy-band diagram near the interface. The Fermi-level position is indicated for the intrinsic GaAs  $c(4 \times 4)$ /Ge(100) interface and for that interface with an interlayer of roughly a monolayer of Al at two deposition temperatures. In addition to the observed variations in the Fermi-level position in the gaps, the interface is found to contain As, Al, Ga, and Ge in various concentrations that depend on the deposition temperature. Surprisingly, the band offsets are stable to all these variations. These observations suggest that the interfacial band lineup is a property of the semiconductors forming the heterojunction. Further, they suggest that the interfacial dipoles are offset by some mechanism to achieve that equilibrium band lineup.

## SUMMARY

We have used synchrotron radiation to investigate the effect of a monolayer of Al interlayer on the GaAs/Ge(100) heterojunction formation. Valence-band and core-level spectra show that the interfacial chemistry and structure are affected by the presence of the Al inter-

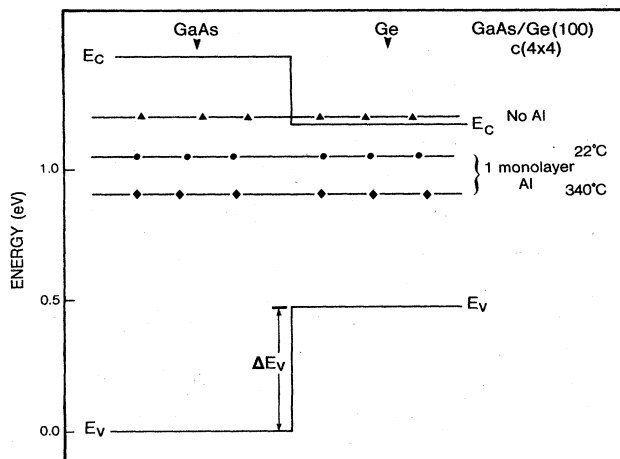


FIG. 4. Schematic of the band-energy diagram near the interface is shown. The Fermi-level position in the gaps is also shown for the three cases studied here. The result for the clean GaAs(100)  $c(4 \times 4)$  surface is consistent with previously reported results. The Fermi-level position is not only influenced by the presence of the Al monolayer, but also by the deposition temperature. The Fermi-level position moves by 0.15 and 0.3 eV toward the valence band for both Al deposition temperatures, room temperature, and 340°C, respectively. This is consistent with the formation of clusters for the room-temperature deposition and the formation of Al-Ga-As alloys for the 340°C temperature. Importantly, the band offset is the same regardless of the presence of the one monolayer of Al interlayer and independent of the Fermi-level position in the gaps.

layer. Our study shows that Al forms clusters when deposited at room temperature, while it forms spatially nonuniform Al-Ga-As alloys when deposited at 340°C.

Both the presence of the Al interlayer and the deposition temperature influence the Fermi-level position in the gaps. We have measured the *same* value for the band offset, within  $\pm 0.05$  eV, regardless of the presence of the Al interlayer and its deposition temperature. The constancy of the band offset suggests that the band-structure lineup for epitaxial heterojunctions is primarily an intrinsic property of the two semiconductors. Any interfacial contributions are small perturbations within the experimental uncertainty of  $\pm 0.05$  eV. It would be interesting to know whether this observation is intrinsic to the GaAs/Ge system or to heterojunctions in general. Further systematic experiments on other heterojunctions are necessary to answer this question.<sup>23</sup>

#### ACKNOWLEDGMENTS

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CdS/Si heterojunctions. Their conclusions are consistent with ours (private communication). However, generalizations of these conclusions for all heterojunctions are not favorable at this stage.