Electron states at semiconductor interfaces: the intrinsic and extrinsic charge neutrality levels

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A discussion is presented on the semiconductor interface barrier formation. Schottky barriers and heterojunction band offsets are analysed by means of the intrinsic and extrinsic charge neutrality levels. These levels are shown to be controlled by the interface geometry and its local chemistry. The chemical properties and the charge neutrality levels of different Schottky barriers are presented. Heterojunction band offsets are also analysed and are shown to depend on the electronegativity of the metal intralayers deposited at the interface: more electronegative metal atoms tend to reduce the heterojunction band offsets.

1. Introduction

Interface semiconductor growth and the barrier heights of different semiconductor junctions (Hiraki et al. 1989; Perfetti et al. 1992) are at the heart of the modern microelectronic technology. To understand those issues, the fundamental research in surface physics has to address different problems such as the chemical and thermodynamical parameters modifying the epitaxial semiconductor growth (Tsukada & Kawazu 1992), and the link between the chemical properties of the interface and the electronic and transport properties of a junction.

One of the important results that has been stressed in the past few years, in the field of semiconductor interface barrier formation (Perfetti et al. 1992), is that the junction barrier height is crucially dependent on the chemical properties of the last layers forming the physical interface. This links the semiconductor growth studies and the semiconductor interface formation, with the transport properties of a junction (Rhoderick & Williams 1988).

A great deal of the recent work in the field of semiconductor interfaces (Stiles et al. 1987; Spicer 1989; Prietsch et al. 1988) has been addressed to understanding how the different barrier heights depend on the very early stages of deposition of the first few atoms (either metal or semiconductor-like) deposited on the semiconductor. This research has tried to relate the barrier heights of thick deposited overlayers to the ones found in the very low coverage limit of deposition. Interface semiconductor barrier heights have been analysed by using physical models related to the induced density of interface states (IDIS model) (Tejedor et al. 1977; Tersoff 1984), or to the electron levels associated with defects (defect model (DM)) (Spicer et al. 1979). The IDIS model seems to explain the main trends in the semiconductor barrier behaviour, with the DM introducing some modulation depending on the quality of the ideal interface (Mönch 1990). In the IDIS model, the main concept is related to the new density of electron states induced near the semiconductor energy gap by the proximity of another crystal. In the case of a metal–semiconductor junction, these
states are mainly due to the tails of the metal wavefunctions that penetrate into the semiconductor energy gap; part of these states are compensated by a reduction in the density of states of the semiconductor valence band (Flores & Tejedor 1987). Then a semiconductor charge neutrality level (CNL) can be defined, this level playing the role of an effective Fermi energy for the semiconductor. The CNL is controlling how the charge is transferred between the semiconductor and any other crystal deposited on it. In general, the CNL is going to pin strongly the interface Fermi energy for a metal–semiconductor interface. For a heterojunction (Tejedor & Flores 1978; Tersoff 1985), the band offset can be obtained by aligning the CNLS of both semiconductors, a misalignment causing a charge flow between the two semiconductors that tend to restore the initial situation.

It is important to stress that the semiconductor CNL is not independent of the interface. Thus, for Schottky junctions the CNL is only an intrinsic semiconductor property if a jellium model is taken for the metal (Flores & Ortega 1992a). The CNL is found to fluctuate around the intrinsic value when the different chemical bonds at the interface are considered in detail. This has led to the introduction of the extrinsic CNL, which fluctuates around the intrinsic one depending on the interface geometry and the particular metal forming the junction (Flores et al. 1989).

For heterojunctions, the semiconductor intrinsic CNLS determine the interface band offset for ideal junctions, i.e. if the semiconductors have a small mismatch and the same crystal structure (Tejedor et al. 1977; Tersoff 1984). For more complex geometries (Niles & Margaritondo 1986; Perfetti et al. 1986; Perfetti 1987; Niles et al. 1988), with metal or semiconductor intralayers, the semiconductors CNLS can be modified (Pérez et al. 1990; Muñoz et al. 1989; Pérez 1992), this effect inducing a band-offset change.

The aim of this paper is to present a summary of different results for Schottky interfaces and heterojunctions, showing how the semiconductor CNL is a function of the local chemistry and the geometry appearing at the interface. This modification offers a way for controlling semiconductor barriers by introducing the appropriate atomic species at the interface.

Our discussion will be split into two parts: in §2 we present our results for Schottky interfaces, while in §3 we consider the heterojunction case. Finally, we present our conclusions in §4.

2. Schottky barriers

Our discussion in this section will be concentrated on the case of different metals deposited on GaAs(110). Our aim is first of all, to understand how the Schottky barrier formation depends on the metal coverage and, then, to relate the barrier heights to the extrinsic CNL. This analysis will provide the basis for relating the interface chemistry with the Schottky barrier height.

(a) Semiconductor-like interfaces: Al and Sb on GaAs(110)

First of all, we consider the Al and Sb cases (Ortega et al. 1992b; Flores et al. 1992) that present a semiconductor-like interface behaviour when deposited at low coverages on GaAs. Figure 1 shows the GaAs(110) surface and the different sites that have been considered for analysing the chemisorption properties of the metal atoms deposited on the semiconductor. In our approach we have analysed, for Al and Sb, either half a metal monolayer ($\theta = \frac{1}{2}$ means a metal atom per semiconductor unit cell) or a metal monolayer deposited on the semiconductor on different surface sites. We
Figure 1. GaAs(110) surface. The different sites (A to E) used for analysing the properties of the adsorbed metal atoms are shown.

Figure 2. Local density of states for an Al monolayer deposited on GaAs(110). $E = 0$ corresponds to the GaAs valence band top.

Figure 3. Local density of states for an In half a monolayer deposited on GaAs(110); long-bridge position.

have analysed these cases by using a parameter-free self-consistent LCAO method, that provides a convenient way of obtaining the local metal–semiconductor interaction (details about the method have been published elsewhere (García-Vidal et al. 1991)).

The main results that are worth commenting here are the following ones. (i) For $\theta = \frac{1}{2}$ the Al (or Sb) atoms tend to be bonded to both semiconductor surface atoms Ga and As, occupying the midpoint of the long bridge position (site C in figure 1). With this geometry the Al (or Sb) atoms form bond with two atoms of the semiconductor maximizing their chemisorption energy (we emphasize that we have neglected in our calculations the substrate reconstruction). (ii) For $\theta = 1$, with two metal atoms per unit cell, we find that the Al (or Sb) atoms tend to be located simultaneously at the A and B sites (see figure 1), on the semiconductor dangling bonds. It is easy to understand that this new geometry is favoured by the number of bonds that each atom is forming at the surface. Thus an Al atom is bonded either to Ga or As, and to two other Al atoms. The three Al bonds are saturated by the valence electrons of each atom. For Sb, things are similar because two other electrons (out of five) are
forming a lone pair not bonded to any other species. (iii) Our calculations (Ortega et al. 1992b; Flores et al. 1992) yield a chemisorption energy per unit cell for the Al (or Sb) monolayer, larger than twice the one calculated for half a monolayer. This shows that the Al (or Sb) atoms should attract each other strongly, tending to form islands at the interface. (iv) From the point of view of the Schottky barrier formation, the monolayer structure gives a semiconductor-like density of states at the interface. Figure 2 shows the density of states calculated for Al on GaAs(110), and $\theta = 1$. The important point to notice about these results is the energy gap structure: for this density of states the interface Fermi level would be unpinned in the semiconductor energy gap. (v) Our conclusion is that the Schottky barrier for Al and Sb on GaAs(110) can be only formed after the metal atoms start being deposited on a second layer.

(b) Metal-like interfaces: the Kondo-like peak; extrinsic CNLS

Let us consider, in this section, the alkali metals (Ortega & Flores 1989; Ortega et al. 1992a) and the In cases (Ortega et al. 1993). These metals present a similar behaviour to each other from the point of view of the Schottky barrier formation. At variation with the semiconductor-like interfaces we shall see how these metals, at low coverages, present a metal-like structure modulated by strong correlation effects.

We have analysed the In case (Ortega et al. 1993) for $\theta = \frac{1}{2}$ and 1. For half a monolayer we have found that the In atoms are bonded to GaAs(110) occupying site C of figure 1. As found previously for Al, the In atoms form bonds with Ga and As maximizing in this way the chemisorption energy. Things appear differently for $\theta = 1$: this is basically due to the In size. Our calculations show that one In atom per unit cell defines the maximum amount of metal that can be adsorbed on the semiconductor surface. More metal atoms can only be adsorbed by forming a second metal layer.

These results have important implications for the Schottky barrier formation. First of all, when In is deposited on GaAs(110), we can expect to find the In atoms located on site C (figure 1); moreover, the saturation coverage for the first layer is $\theta = \frac{1}{2}$. For this case we have one In atom and three electrons per unit cell and, consequently, a metal-like interface structure is expected to appear. Figure 3 shows the local density of states for In on GaAs(110) for $\theta = \frac{1}{2}$: the important point to notice about this density of states is that the Fermi level is pinned by a half-occupied surface band appearing in the semiconductor energy gap. This result suggests that the Schottky barrier is already formed for an In deposition of $\theta = \frac{1}{2}$; we have checked that the second monolayer only broadens the narrow linewidth of the surface state found for $\theta = \frac{1}{2}$, yielding practically the same position for the interface Fermi level.

Alkali metals present a similar behaviour from the point of view of the Schottky barrier formation (Ortega & Flores 1989; Ortega et al. 1992a). For low coverages, we find that the alkali atoms tend to be bonded to Ga. When the deposition is increased, we also find that the maximum metal coverage for the first layer is $\theta = \frac{1}{2}$; extra alkali metal atoms can only be deposited on a second layer. Then, the Schottky barrier formation can be expected to evolve like in the In case: for $\theta = \frac{1}{2}$, with one electron per unit cell, we find a half occupied surface band (similar to the one shown in figure 3 for In) pinning strongly the Fermi level. Any further metal deposition will broaden the metal induced surface states, pinning the Fermi energy practically at the same position as found for $\theta = \frac{1}{2}$.

Electron correlation effects can change partly the previous picture. The point to realize is that the induced surface band (for In or the alkali metals) is rather narrow
(ca. 0.6 eV) compared with the intrasite Coulomb interaction associated with its Wannier wavefunction (ca. 1.2 eV). Then correlation effects (Flores & Ortega 1992b; Plummer & Dowben 1993; DiNardo et al. 1990) tend to split the initial surface band into three pieces: the central one is the Kondo-like peak still pinning the Fermi level; there also appear two other peaks roughly located symmetrically with respect to the central peak: these peaks can be described as the ionization (donor) and the affinity (acceptor) levels associated with the surface band. The Kondo-like peak is found to have a width of 25 meV or less; this means that unless the interface is kept at very low temperature, that peak should be washed out. Anyway, what is important regarding the present discussion is that the Fermi energy should still be pinned by the density of states appearing around the Kondo-like peak of the surface band found in the one-electron calculations (like the one shown in figure 3 for In). We conclude that the Fermi level and the barrier height could be calculated using a one-electron picture, disregarding many-body effects.

After having discussed how the Schottky barrier for the alkali metals and In is almost completely formed after the deposition of the first metal layer, we turn our attention to the analysis of the extrinsic and intrinsic CNLS for these metals. This is conveniently performed if we analyse the CNL for different geometries and metals in the following way. First assume to have a metal atom deposited on one of the sites shown in figure 1; then, for \( \vartheta = \frac{1}{4} \), relax the metal layer moving the metal atoms in the direction perpendicular to the semiconductor surface. For each position, we calculate the induced density of interface states and determine the Fermi level and the semiconductor CNL (the total semiconductor charge located below the CNL yields a global neutral condition). In all our calculations, we find that \( E_F \) and the CNL, \( \phi^0 \), differ by less than 0.05 eV. So, from now on we disregard this difference and show in figure 4, for different metals, \( \phi^0 \) (or \( E_F \) measured from the valence band top) as a function of the distance between the metal layer and the last semiconductor layer. For each metal, we show different curves corresponding to the different sites chosen for the metal atoms. The crucial point to notice is that, in all the cases, \( \phi^0 \) evolves as a function of the metal–semiconductor distance and shows a different behaviour depending on the adsorption site. This allows us to define the extrinsic CNL as the particular CNL found in each calculation. In general, we define the intrinsic CNL as the average of all the extrinsic values calculated at a fixed metal–semiconductor distance, for all the possible sites the metal atom can be adsorbed on. The reason for this is that the jellium model can be viewed as the average of all the different sites the metal atom can occupy.

In figure 4, we have also drawn the intrinsic CNL defined as the average of the extrinsic values calculated for different sites (notice that we are only using a finite number of sites for obtaining the mean CNL). The conclusion that can be drawn from figure 4 are the following: (i) In all the cases the intrinsic CNL is around 0.7 eV ± 0.10 eV (remember this is GaAs(110)). (ii) The extrinsic CNLS present important fluctuations around the intrinsic one. These fluctuations are smaller for K, the most electropositive metal atom, and increase in the direction of electronegativity, in particular, for In, the extrinsic CNLS can change between 1.2 eV and 0.2 eV. (iii) The values calculated for the points having the maximum chemisorption energy energies are the following:

\[
\phi^0(K) = 0.70 \text{ eV}, \quad \phi^0(\text{Na}) = 0.66 \text{ eV}, \quad \phi^0(\text{Li}) = 0.60 \text{ eV}, \quad \phi^0(\text{In}) = 0.49 \text{ eV},
\]

showing a trend that yields a Fermi energy closer to the valence band top for more
electronegative atoms. (iv) Finally, we also find that the extrinsic CNLS calculated for the maximum chemisorption energy are very close to the intrinsic levels, for very electropositive atoms. This is due to the small fluctuations the extrinsic CNLS have in these atoms, around the intrinsic one.

3. Heterojunctions

Previous discussion has shown how for Schottky junctions the barrier height is crucially dependent on the interface chemical properties. Basically, these properties modify the extrinsic CNL of the semiconductor, yielding a corresponding shift of the interface Fermi energy.

Heterojunctions show a similar behaviour. For ideal interfaces, the band offsets can be obtained with a good accuracy by using the intrinsic CNLS. Table 1 shows different semiconductor band offsets, $\Delta E_F$, calculated using the IDIS model; these values are compared with other theoretical calculations (Van de Walle & Martin 1987) and the experimental evidence (Margaritondo & Perfetti 1987). The results shown in table 1 have been obtained using the following CNLS: for GaAs, the mean value given above ($\phi^0$(GaAs) = 0.70 eV), for GaP the value given in Gozzo et al. (1992), for ZnSe the value of Pérez et al. (1990), and for the other semiconductors the values given by Tersoff (1984). Let us stress the good agreement obtained between the results given by the IDIS model and the ones obtained by LDA calculations. Where the agreement is worse, the IDIS model has a better predictive power.

Figure 5. Energy levels for (a) an ideal ZnSe–Ge(110) interface and (b) a ZnSe–Al–Ge(110) interface. The intrinsic, $\phi_i^{\text{Ge}}$, and the extrinsic, $\phi_x^{\text{Ge}}$, CNLS (dashed lines) are shown as well as the band offsets, $\Delta E_x$, and the Fermi energy, $E_F$ (line across the junction).

Table 1. Band offsets for different semiconductors

<table>
<thead>
<tr>
<th></th>
<th>IDIS</th>
<th>LDA</th>
<th>EXP</th>
</tr>
</thead>
<tbody>
<tr>
<td>GaP–Si</td>
<td>0.77</td>
<td>0.61</td>
<td>0.80</td>
</tr>
<tr>
<td>GaAs–Ge</td>
<td>0.52</td>
<td>0.63</td>
<td>0.35–0.55</td>
</tr>
<tr>
<td>AlAs–Ge</td>
<td>0.87</td>
<td>1.05</td>
<td>0.95</td>
</tr>
<tr>
<td>AlAs–GaAs</td>
<td>0.35</td>
<td>0.37</td>
<td>0.38</td>
</tr>
<tr>
<td>ZnSe–Ge</td>
<td>1.64</td>
<td>2.17 (1.58)</td>
<td>1.40</td>
</tr>
<tr>
<td>ZnSe–GaAs</td>
<td>1.12</td>
<td>1.59 (0.99)</td>
<td>1.10</td>
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</table>

Non-ideal interfaces need a different analysis. We shall show by considering the ZnSe–Al–Ge(110) interface (Pérez et al. 1990) how band offsets can be changed by using appropriate intralayers. In particular, we shall show how the band offset changes are related to the modifications introduced in the CNLS of each semiconductor. As discussed previously for the Schottky barriers, metal intralayers (depending on the properties of the interface) can modify the CNLS; for each particular case, one should calculate the extrinsic CNLS and obtain the band offset by their alignment.

(a) ZnSe–Al–Ge(110) interface

We shall discuss this interface following Pérez et al. (1990). In this work, the ZnSe–Ge(110) band offset was analysed as a function of the number of Al intralayers deposited at the interface. The calculation was performed by using a semi-empirical LCAO method that allowed for the charge transfer across the different interfaces. By calculating the total density of states, the semiconductor CNLS as well as the interface Fermi level were obtained. Figure 5 shows a diagram of the different energies involved in the problem. Figure 5a corresponds to the ideal interface with a band
offset of 1.70 eV, and the following intrinsic CNLS, $\phi^0(\text{Ge}) = 0.18$ eV and $\phi^0(\text{ZnSe}) = 1.82$ eV. As mentioned above for Schottky barriers, $\Delta E_V = 1.70$ eV is not exactly $\phi^0(\text{ZnSe}) - \phi^0(\text{Ge}) = 1.64$ eV, but the difference is small. Figure 5b shows the interface with an Al intralayer. We have drawn the extrinsic CNLS and the Fermi energy that appears inside the finite density of states induced in the energy gap by the metal intralayer.

Table 2 gives $E_V$, $\Delta E_V$ and the extrinsic CNLS, $\phi^0(\text{ZnSe})$ and $\phi^0(\text{Ge})$, as a function of the number of Al layers. First point to notice is the following: in all cases, $\Delta E_V$ is given, within 0.05 eV, by the differences between the extrinsic CNLS $\phi^0(\text{ZnSe}) - \phi^0(\text{Ge})$; this shows that the interface electrostatic dipole is adjusted to give the alignment of both extrinsic CNLS. The small difference between these levels allows for the charge transfer that creates that electrostatic dipole. Table 2 also shows how the extrinsic CNLS depend on the number of Al layers; for more than two Al layers we recover the Schottky-barrier limit for each particular interface, with both extrinsic CNLS having reached its metal–semiconductor limit.

The main effect of the intralayer already appears for one and two Al monolayers: table 2 shows how the extrinsic CNLS are modified for these coverages yielding a value of $\Delta E_V$ larger than the one obtained for the ideal heterojunction. It is also interesting to realize that the Fermi energy is very close to the CNLS of both semiconductors although a little higher in energy due to the charge transfer from the metal to the semiconductors.

The main conclusion we can draw from these results are the following. (i) First, the band offset is saturated for two Al monolayers; for a further deposition, no band-offset evolution is induced at the interface. (ii) The band-offset changes are mainly due to the modifications in the extrinsic CNLS of the two semiconductors.

One should keep in mind that the extrinsic CNLS are a function of the interface geometry and the bonding between the metal and the semiconductors. The results of table 2 were obtained using a specific geometry: we should stress here that different results were obtained by changing the interface chemical bonding (Pérez et al. 1990). This shows that the intralayer effect could be only accurately obtained by determining (experimentally or theoretically) the interface geometry. Then, one should calculate for this geometry the semiconductor CNLS and the corresponding band offset.

**Table 2.** $E_V$, $\Delta E_V$, and the CNLS of ZnSe and Ge as a function of the number of Al layers deposited at the interface.

(The values are referred to the ZnSe valence band top.)

<table>
<thead>
<tr>
<th>no. of Al layers</th>
<th>$E_V$</th>
<th>$\Delta E_V$</th>
<th>$\phi^0(\text{ZnSe})$</th>
<th>$\phi^0(\text{Ge})$</th>
</tr>
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<tbody>
<tr>
<td>-</td>
<td>1.70</td>
<td>1.82</td>
<td>1.88</td>
<td></td>
</tr>
<tr>
<td>1</td>
<td>2.67</td>
<td>2.11</td>
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<td>2</td>
<td>2.61</td>
<td>2.24</td>
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<td>3</td>
<td>2.58</td>
<td>2.20</td>
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<td>4</td>
<td>2.57</td>
<td>2.19</td>
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<tr>
<td>5</td>
<td>2.58</td>
<td>2.20</td>
<td>2.56</td>
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</table>

(b) General considerations for metal intralayers in heterojunctions: band-offset modifications

The previous section has shown how an Al intralayer modifies the ZnSe–Ge(110) band offset. Can we draw more general conclusions to predict the effect of different metal intralayers on the heterojunction band offset? The aim of this section is to make a few comments on this direction, linking the Schottky-barrier formation with the effect of metal intralayers on the heterojunction band offsets.

The first point to notice is obvious. For a thick metal intralayer, the heterojunction band offset is determined by the independent behaviour of the two Schottky barriers formed at the two metal–semiconductor interfaces. Our previous comments on the ZnSe–Al–Ge heterojunction show that that limit is recovered for a two monolayers thick film. The case of a monolayer is found to be intermediate between the ideal heterojunction case and the thick intralayer limit. Then, the analysis of the thick intralayer case can be expected to give the main trends in the effect of the metal intralayers.

Regarding the Schottky barriers, it is worth commenting how the metal atom electropositivity changes the extrinsic CNL. For the cases discussed above, we have found that for the alkali metals and the In interfaces, the extrinsic CNLS for the most stable geometries decrease with the atom electronegativity. Thus for K we find the largest CNL while for In we find the lowest one. Basically, the change of this extrinsic CNL, $\phi^0$, with the metal atom electronegativity, $\chi$, can be described as a typical semiconductor index:

$$ S = -d\phi^0/d\chi \approx -dE_F/d\chi. $$

(1)

Based on the experimental evidence (Rhoderick & Williams 1988), we infer that $S$ increases with the semiconductor ionicity. This suggests that when a metal intralayer is deposited in a heterojunction, the larger is the metal electronegativity, the larger is the modification in the CNLS of both semiconductors. As the ionic semiconductors have the larger energy gaps and the larger changes in the CNLS, we deduce that metal intralayers of more electronegative atoms tend to reduce the heterojunction band offset.

This result has been checked by specific calculations. Thus, for the GaP–Si(110) interface, the results of Muñoz et al. (1989) show that for Cs, Al and H intralayers, the band offsets decrease with the atom electronegativity ($\Delta E_V$ is found to be the largest for the Cs case and the lowest for H). This is also in agreement with the experimental results of Perfetti (1987).

In conclusion, the heterojunction band offsets can be modified appropriately by choosing the electronegativity of the atoms deposited at the interface. More electropositive atoms yield larger barriers.

4. Conclusions

In this paper, a short review of the most important results obtained in our group for different semiconductor interfaces is presented. Our calculations show that the interface semiconductor barriers depend crucially on the interface chemistry and its atomic geometry. In particular, we have found that the local interface geometry of the metal–semiconductor junction determine the extrinsic CNL of the semiconductor and its Schottky barrier. We have also found that the extrinsic CNLS of a given
semiconductor fluctuates around its intrinsic CNL as a function of the metal electronegativity: more electronegative metal atoms yield larger fluctuations. In particular, only for very electropositive metals the interface Fermi level is pinned near the intrinsic CNL. This shows that, in general, Schottky barriers can only be obtained by specific calculations, determining the interface geometry and the particular extrinsic CNL. We have performed this task for a few metal atoms. Calculations yield extrinsic CNLS for the interface geometry having the maximum chemisorption energies that are smaller for more electronegative atoms.

Heterojunction band offsets can be analysed following the same ideas. First of all, band offsets of ideal heterojunctions can be obtained with good accuracy by aligning the intrinsic CNLS of both semiconductors. The effect of metal intralayers on the heterojunction band offsets can also be described by means of the extrinsic CNLS of both semiconductors. Like in Schottky barriers, the metal intralayers modify the semiconductor extrinsic CNL: their alignment defines the new band offset. Our analysis shows that the heterojunction band offset depends on the metal electronegativity: more electronegative atoms tend to reduce the heterojunction band offset. This result offers a way of controlling the heterojunction band offsets.

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