ADVANCED MATERIALS INTERFACES

Supporting Information

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Tapered Cross-Section Photoelectron Spectroscopy Provides Insights into the Buried Interfaces of III-V Semiconductor Devices

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Keywords: Tapered Cross-Section, Photoelectron Spectroscopy, III-V semiconductors, buried interfaces, energetic alignment

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1. Tapered cross-section preparation and characterization

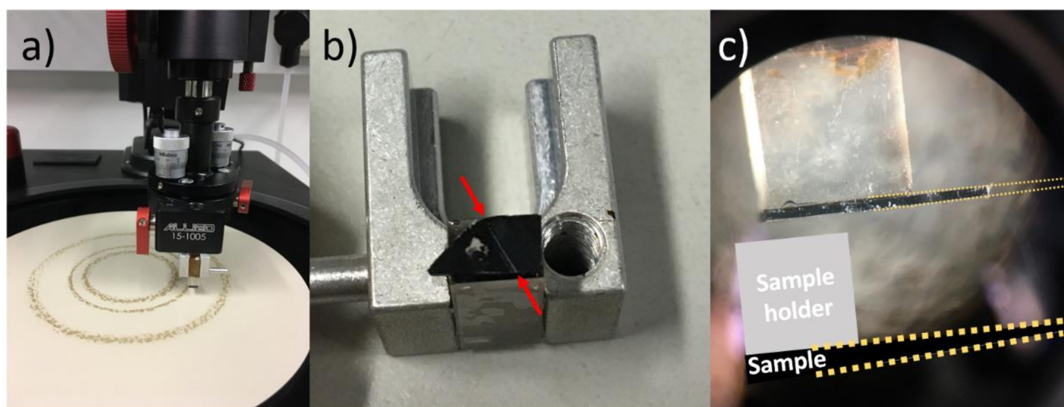

Figure S1: Images of the experimental polishing procedure. a) The sample is glued on a sample holder that is then attached to the MULTIPREP^{TM} polishing system. b) After polishing, a clear demarcation (see red arrows) of the exposed interface between Ge and GaInP can be seen; on the left side of the interface an accumulation of matter can be distinguished. c) With an optical microscope, the tapered geometry can be observed on top and it is sketched on the bottom, the yellow dots are guides to the eye showing a small tapering angle, *i.e.,* the angle between the TCS and the bottom of the sample.

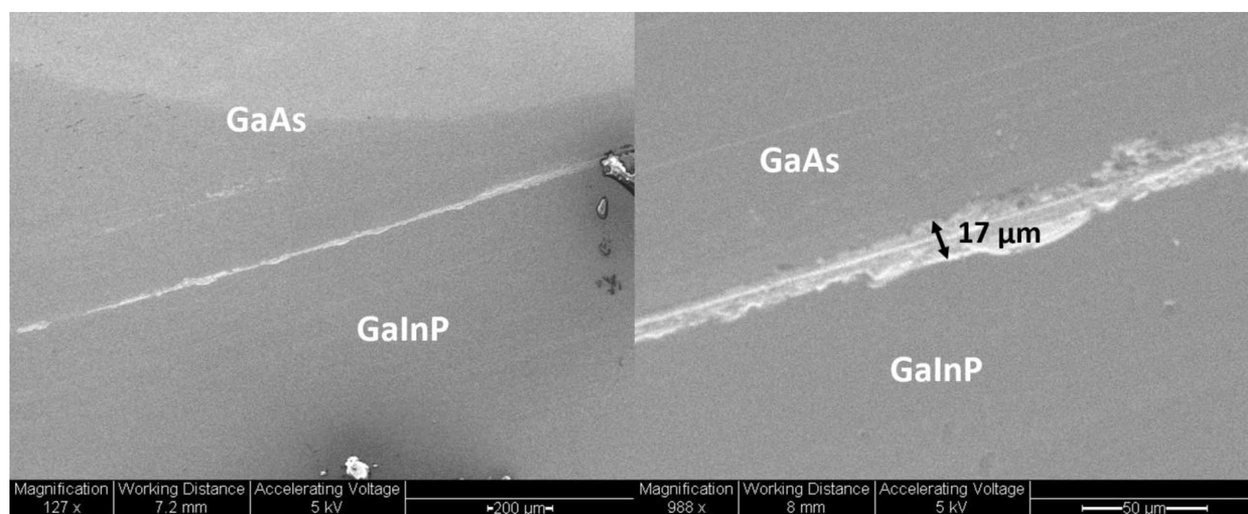

Figure S2: Additional SEM images of GaAs|GaInP.

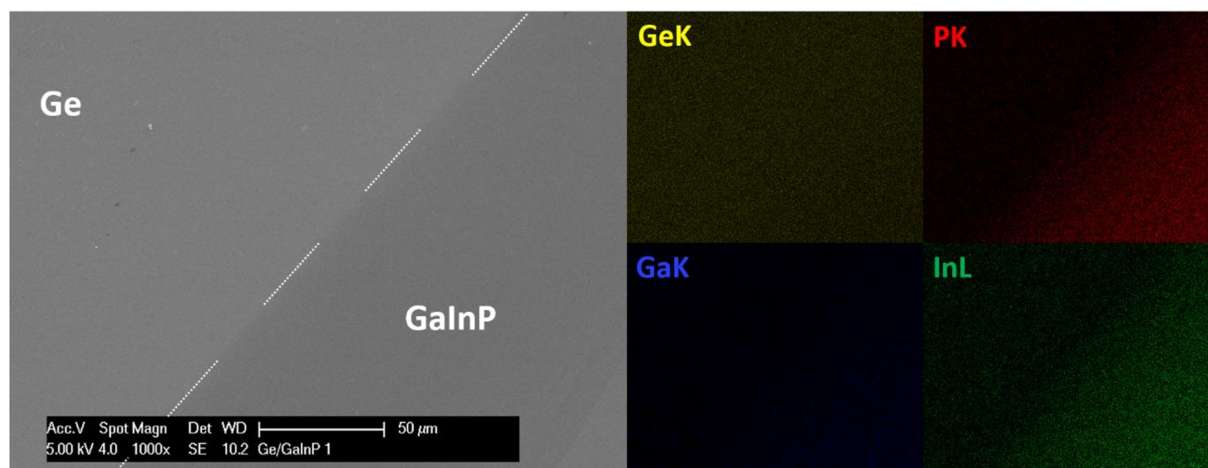

Figure S3: SEM images of Ge|GaInP and EDX mapping of GeK, PK, GaK, and InL. Ge is observed everywhere because the depth of analysis is high enough to detect the substrate under the GaInP layer but Ga, In and P signals are more intense in the right bottom corner. Dotted lines are put as a guide of the eye to distinguish the Ge|GaInP interface.

2. Getting insights with the TCS-PES and its limits

Case study of Ge|GaInP:

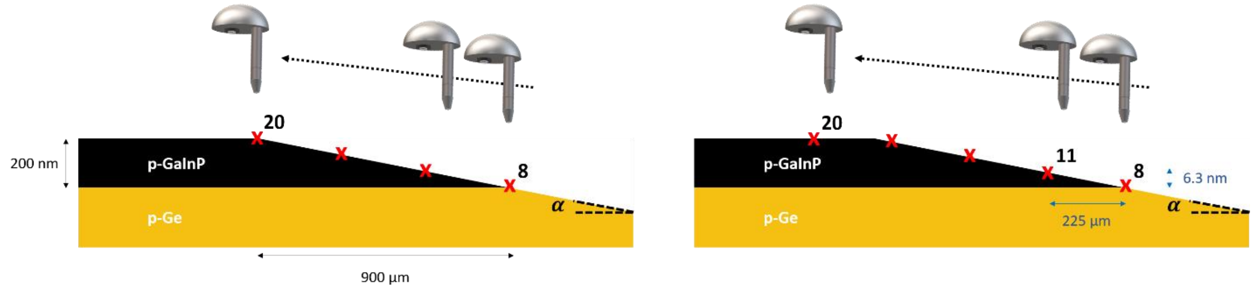

Figure S4: Scheme of the TCS of the Ge|GaInP sample. A 200 nm GaInP layer was deposited on top of a Ge substrate. The TCS-PES was conducted with a step width of 75 µm. It revealed an interface at position 8. Two methods are compared to calculate the TCS angle α.

The tapering angle can be calculated using the thickness of the different layers (*i.e.* normal part width) that have been prepared and the estimated tapered width based on XPS measurements. The following equation to calculate the tapering angle α

$$\alpha = \text{tapering angle} = \tan^{-1}\left(\frac{\text{normal part width}}{\text{tapered width}}\right)$$

1st method (left): we consider that the TCS goes from position 1 to position 20 without measuring a part of the sample that is not polished (as depicted on the right). Then, position 20 corresponds to the end of the TCS and there is 200 nm height difference between position 20 and the buried interface, according to the MOCVD preparation. According to the step width of 75 µm, 900 µm separates position 8 from position 20.

$$\tan^{-1}\left(\frac{200\ \text{nm}}{900.10^3\ \text{nm}}\right) \approx 0.013\ °$$

2nd method (right): we consider that the TCS stops before position 20 therefore, we need to use another metric. The Ge3d core level can just be discerned at position 11. We, therefore, consider that at position 11 the thickness of the GaInP film corresponds to three times the IMFP (λ). Below 3λ, 95% of the electrons photogenerated from the Ge3d orbital are damped. This 3λ equals 6.3 nm. According to the step width of 75 µm, 225 µm separates position 8 from position 11.

$$\tan^{-1}\left(\frac{6.3\ \text{nm}}{225.10^3\ \text{nm}}\right) \approx 0.002\ °$$

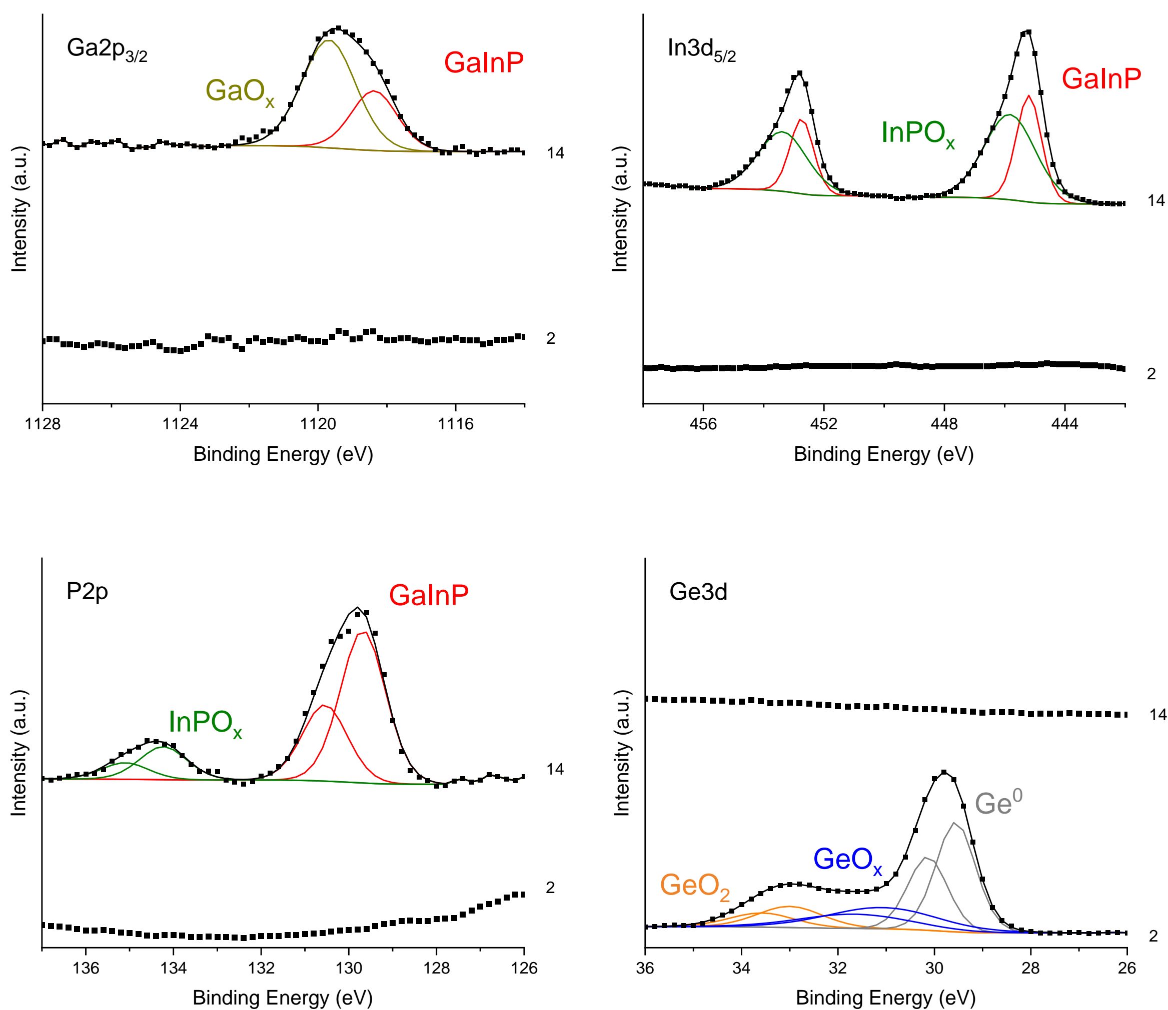

Figure S5: $Ga2p_{3/2}$, In3d, P2p, and Ge3d core level spectra recorded at positions 2 and 14 to show the fitting methodology and the peak identification that has been used for the BE shifts depicted in **Figures 4, S6, and S7**.

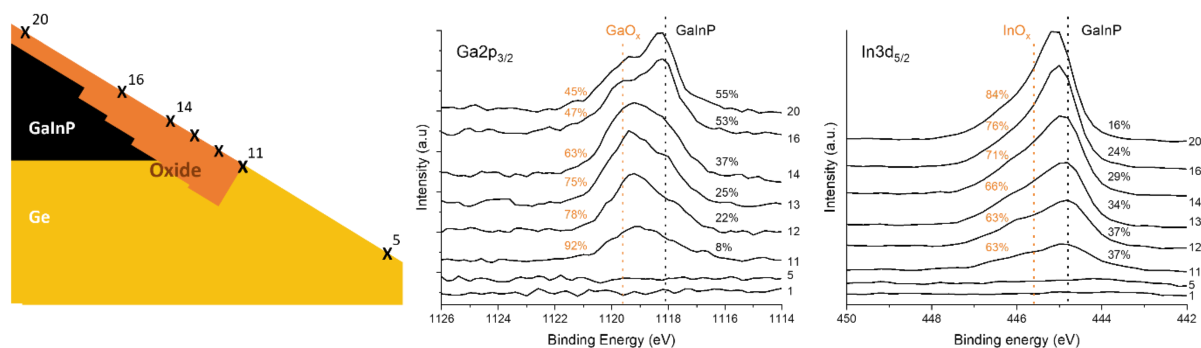

Figure S6: $Ga2p_{3/2}$ and $In3d_{5/2}$ core level spectra were recorded at different positions that are indicated on the scheme of the tapered cross-section for the "Ge|GaInP n°2 sample" (See **Table S1** for more details). Core level spectra were recorded at different positions with a step width of 150 µm. In addition to the GaInP (black) and Ge ((yellow) layers, oxide compounds (orange) have been detected.

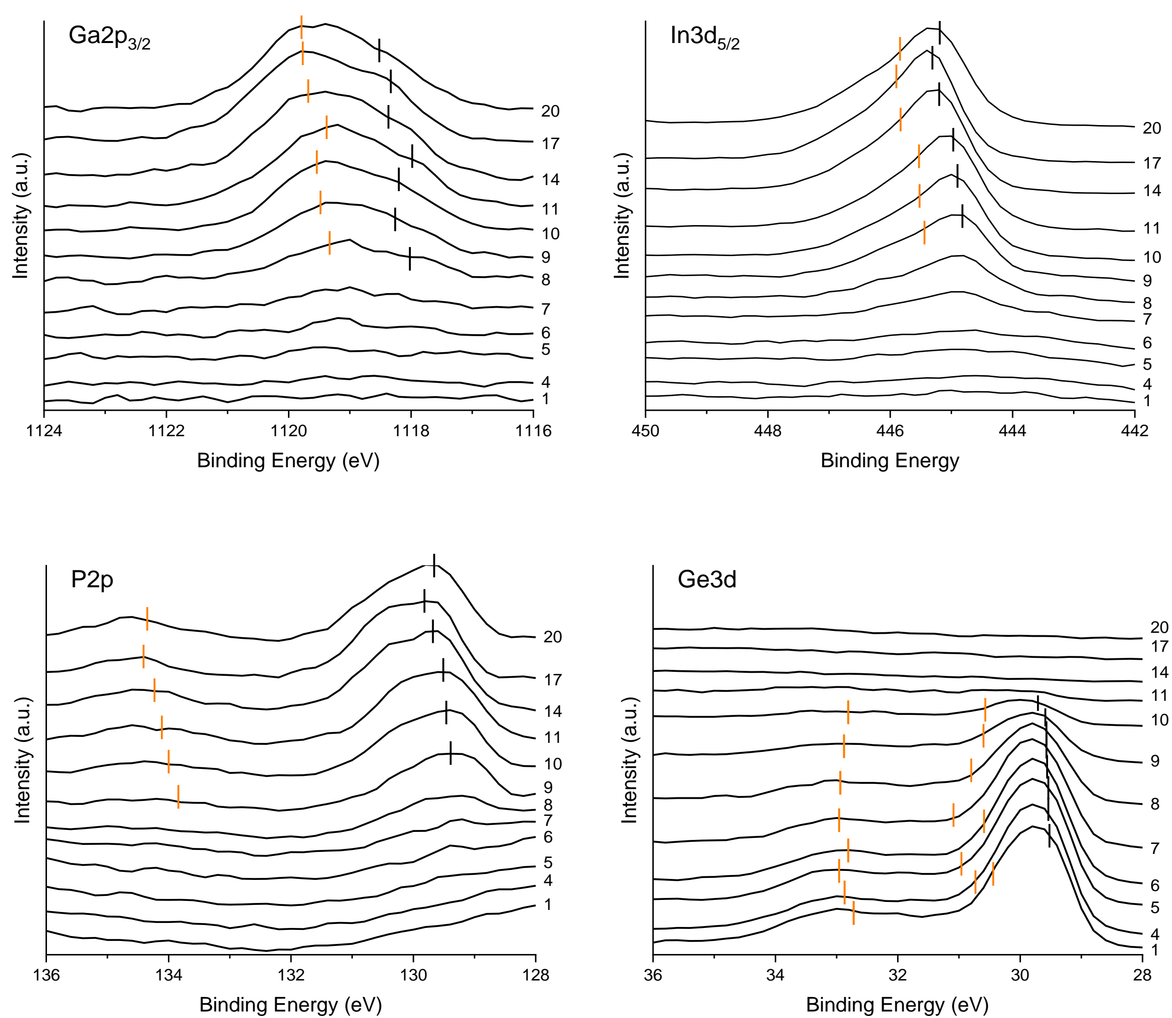

Figure S7: $Ga2p_{3/2}$, $In3d_{5/2}$, P2p, and Ge3d core level spectra were recorded at different positions with a step width of 75 µm. The spectra were then fitted to distinguish the GaInP and the Ge^0 states from the more oxidized species. The vertical lines correspond to the peak maxima of the GaInP and Ge^0 states (black line) and the more oxidized states (orange line). The black lines were then used to plot **Figure 3** of the main article.

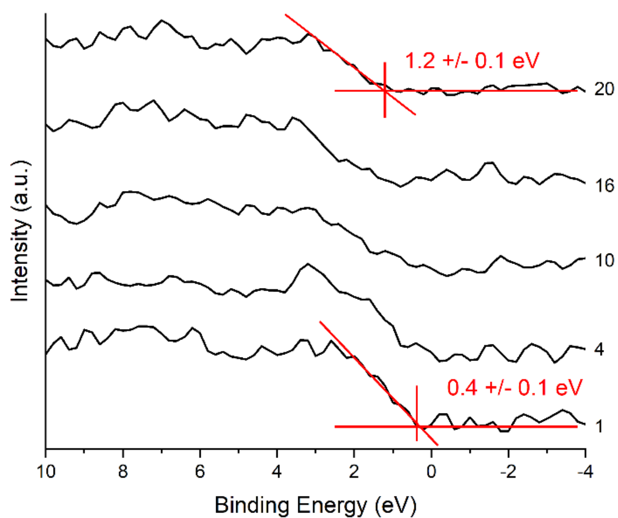

Figure S8: VB edge measured by XPS along the TCS of Ge|GaInP. With, as a reminder, position 1 corresponds to pure Ge, position 20 corresponds to pure GaInP, and the interface is evidenced at position 8. It is unclear if due to the high kinetic energy the oxidation states at the surface of the TCS alter the VB edge.

Case study of n-GaAs|n-GaInP,

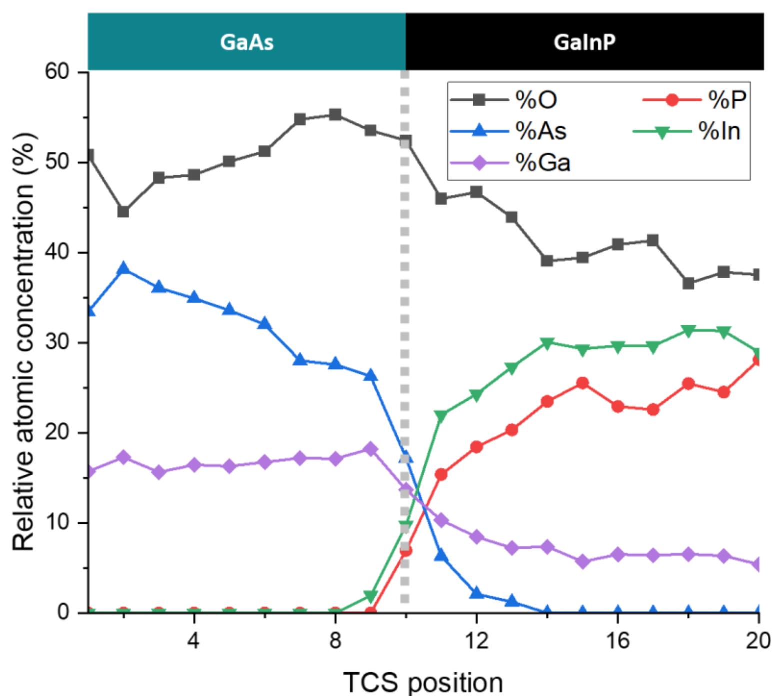

Figure S9: Relative atomic surface concentration of different elements (O, P, As, In, Ga) present across the tapered cross-section GaInP|GaAs. These concentrations were determined from the individual core level spectra across the tapered cross-section. The full spectra were integrated without distinction between the different states.

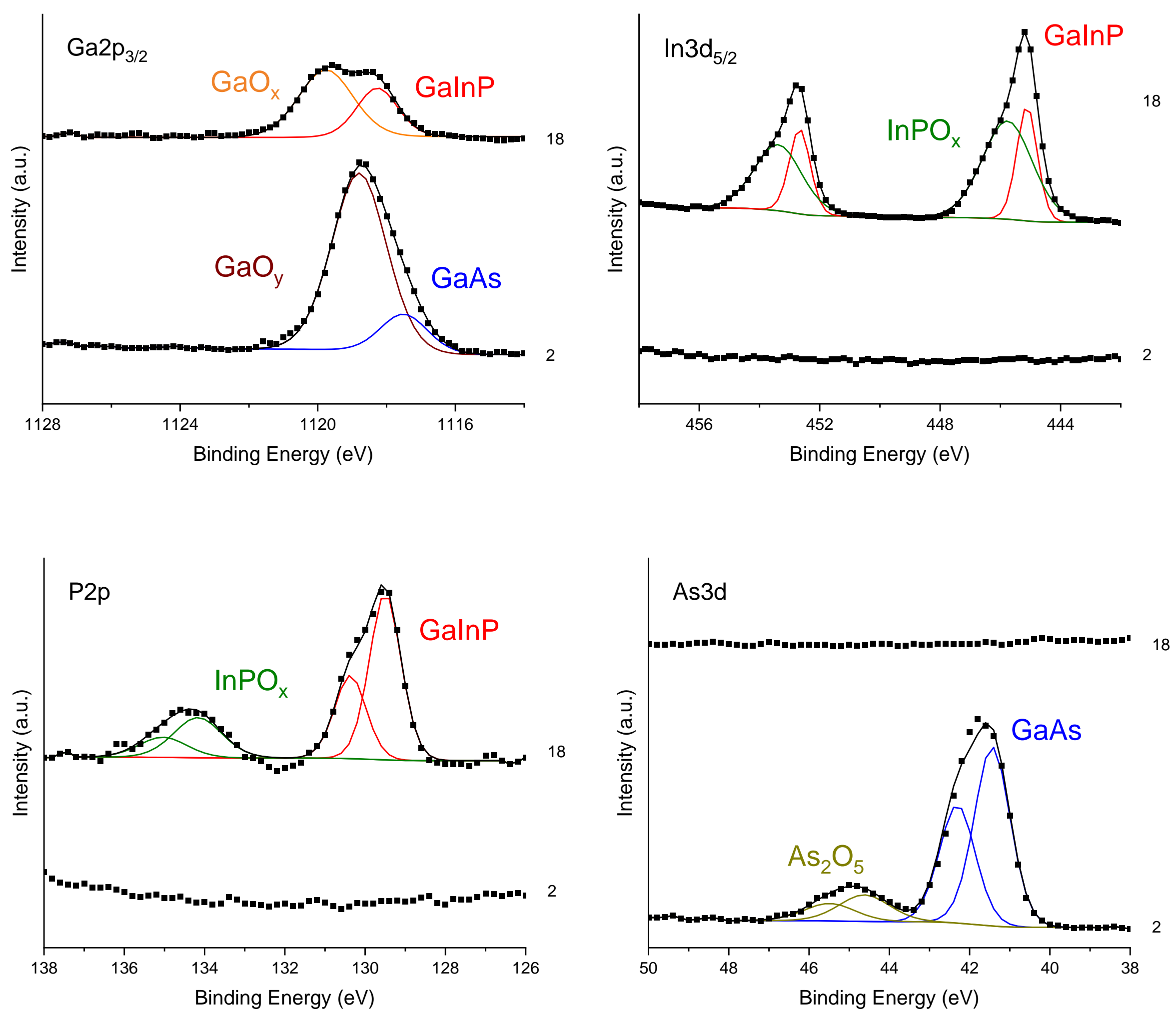

Figure S10: $Ga2p_{3/2}$, $In3d_{5/2}$, P2p, and As3d core level spectra recorded at positions 2 and 18 to show the fitting methodology and the peak identification that has been used for the BE shifts depicted in **Figures S11 and S12**.

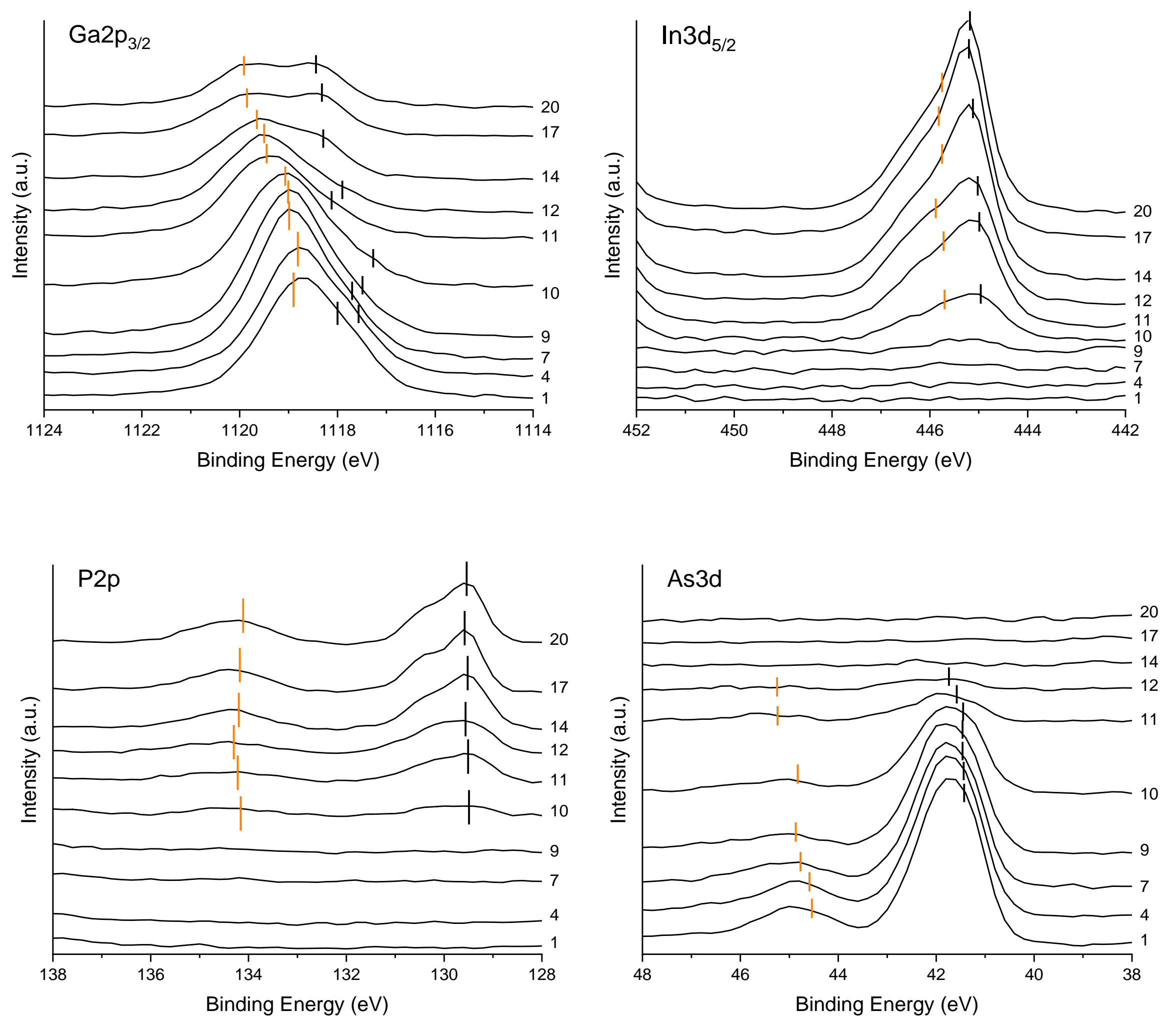

Figure S11: $Ga2p_{3/2}$, $In3d_{5/2}$, P2p, and As3d core level spectra were recorded at different positions with a step width of 75 µm. The spectra were then fitted to distinguish the GaInP and the GaAs states from the more oxidized ones. The vertical lines correspond to the peak maxima of the GaInP and GaAs states. They were then used to plot **Figure S12**.

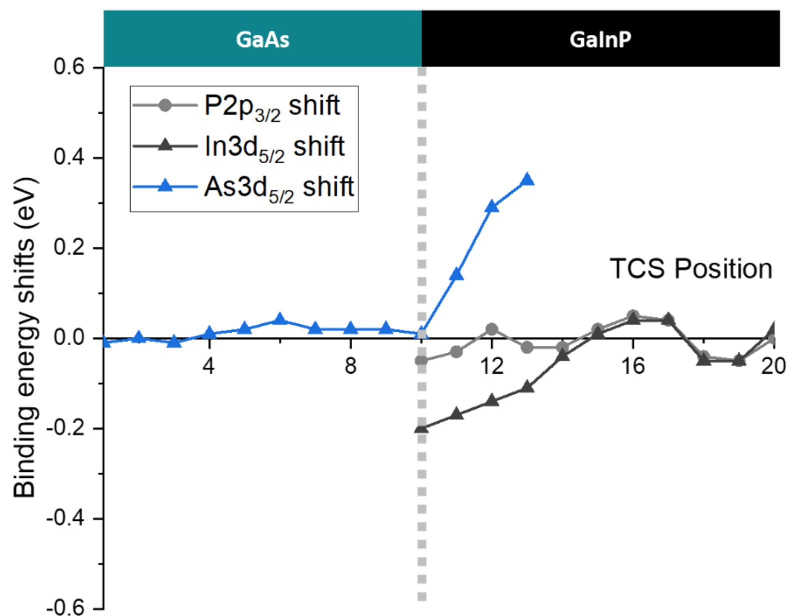

Figure S12: Binding energy shifts of As3d$_{5/2}$, In3d$_{5/2}$, and P2p$_{3/2}$ core levels. The peak maxima of the GaAs and GaInP states are depicted in **Figure S11**.

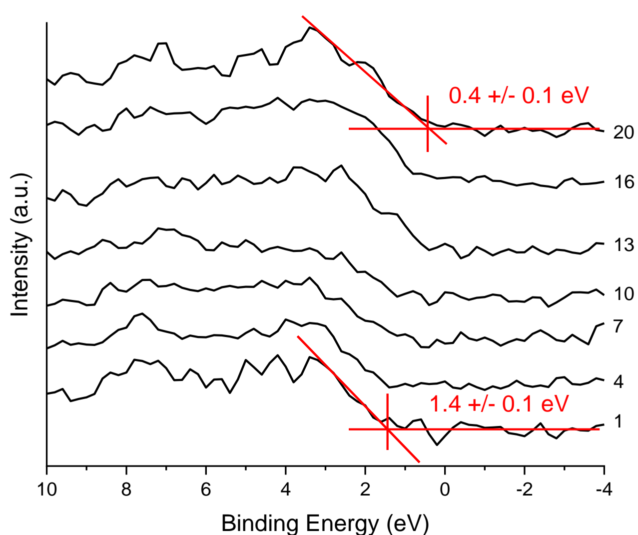

Figure S13: VB edge measured by XPS along the TCS of GaAs|GaInP. With, as a reminder, position 1 corresponds to pure GaAs, position 20 corresponds to pure GaInP, and the interface is evidenced at position 10. It is unclear if due to the high kinetic energy the oxidation states at the surface of the TCS alter the VB edge.

3. Experimental section

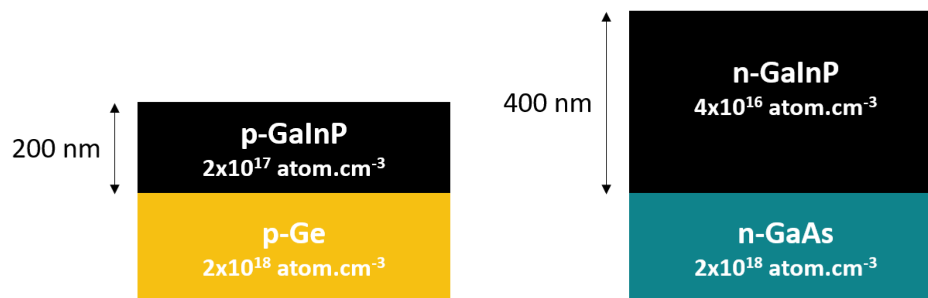

Figure S14: Schemes and details of the two III-V semiconductor devices.

Table S1: Details on the parameters used for the polishing procedure with the MULTIPREP™ polishing system: *t* is the duration of the polishing step; *d* is the grain size of the diamond lapping film; *r* is the rotation speed of the polishing plate; *S* is the sample load (the higher it is the more force is applied to the samples); *α* and *ρ* are the axial and radial angles applied between the sample and the polishing plate.

	Number of polishing steps	*t* (min)	*d* (µm)	*r* (rpm)	*S* (g)	α (mm)	ρ (mm)
Ge|GaInP n°1[a]	1	3	0,25	100	300	0°	0°
Ge|GaInP n°2[b]	1	2.7	0,25	100	400	0°	0°
GaAs|GaInP	3	1	0,25	100	100	0°	+0.1°
		3			100		+0.06°
		1			200		+0.06°

[a]Sample discussed in the main manuscript
[b]Sample discussed in **Figure S6** and briefly in the main manuscript as a duplicate.