Supporting Information for

“Lattice-matched InGaAs-InAlAs core-shell nanowires with improved luminescence and photoresponse properties”

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Additional experimental details

Morphologies of InGaAs-InAlAs core-shell nanowires

Figure S1 displays scanning electron microscopy (SEM) images of all InGaAs-(core-only) and InGaAs-InAlAs core-shell NW samples as grown with different Ga/Al-contents [x(Ga)/y(Al)] on Si (111). All NW arrays exhibit a high growth yield of >90% and clearly
show the increased diameter upon passivation by the radial InAlAs shell. Except for the NW array with $x(Ga) \gamma(Al) = 0.35$, all samples show similar NW lengths and diameters, as also summarized in Table S1.

**Figure S1.** SEM images of all InGaAs NWs (top row) and InGaAs–InAlAs core-shell NWs (bottom row) as grown with different $x(Ga) \gamma(Al)$ of (a,b) = 0.2, (c,d) = 0.35, (e,f) = 0.47, and (g,h) 0.63. The scales bar is equivalent for all images. The values given for composition are actual values determined by HRXRD.

**Table S1.** Summarized dimensions (length, diameter) of the InGaAs NWs (core-only) and InGaAs-InAlAs core-shell NWs for the various different $x(Ga) \gamma(Al)$. Also, for the core-shell NW structures the shell thickness and the length of a top segment that extends in the axial growth direction beyond the InGaAs NW core are given as extracted from the SEM data. Mean values and standard deviation are derived from approximately 20 NWs measured for each sample.

| $x(Ga) \gamma(Al)$ | NW structure     | length (nm) | diameter (nm) | shell thickness (nm) | top segment (nm) |
|-------------------|------------------|-------------|---------------|----------------------|-----------------|
| 0.2               | InGaAs core      | 787 ± 42    | 135 ± 5       | -                    | -               |
|                   | InGaAs-InAlAs    | 788 ± 43    | 148 ± 4       | 6.2 ± 0.4            | 0.6 ± 0.1       |
| 0.35              | InGaAs core      | 411 ± 39    | 149 ± 8       | -                    | -               |
|                   | InGaAs-InAlAs    | 664 ± 61    | 187 ± 7       | 19 ± 2               | 253 ± 7         |
| 0.47              | InGaAs core      | 638 ± 31    | 126 ± 6       | -                    | -               |
|                   | InGaAs-InAlAs    | 661 ± 45    | 138 ± 6       | 6.2 ± 0.6            | 23 ± 3          |
| 0.63              | InGaAs core      | 658 ± 37    | 128 ± 8       | -                    | -               |
|                   | InGaAs-InAlAs    | 688 ± 38    | 141 ± 8       | 6.6 ± 0.6            | 30 ± 3          |
Finite element modelling (FEM) Simulations of XRD patterns

In order to analyze potential contributions from the InAlAs shell to the overall very small strain components, we correlated the Bragg peak positions of the XRD patterns with simulations. This allows us to estimate the approximate composition of the InAlAs shell. In particular, we exemplify this procedure for the NW sample with highest $x(Ga)/y(Al) \approx 0.64$. For the lattice parameters of the core-only NWs, we evaluate the strain with respect to pure WZ material from the Bragg peak positions, assuming a biaxial strain state due to the mutual strain of ZB and WZ segments. For the core-shell NWs, the diffraction peaks are dominated by the signal from the core, as the shell thickness is very small. Hence, we see the influence of the shell only via shifts of the core Bragg peak. To quantify the shell composition, we performed finite element modelling (FEM) of the NWs using the geometrical parameters obtained from SEM and TEM. The chemical composition of the core was fixed to that of the respective core-only NWs, and the shell composition was varied between $0.4 < y(Al) < 0.8$. From the resulting strain distribution, we calculated the x-ray diffraction patterns and determined the Bragg peak shifts as a function of the shell composition, as shown in Figure S2. We find that all samples exhibit only very small peak shifts, and the maximum deviation of shell composition $y(Al)$ from the core composition $x(Ga)$ is 7%. This corresponds to in-plane strain values of the core below 0.1% and below 0.4% for the shells.
Figure S2. Simulated strain component for a fixed Ga-content of \( x(Ga) = 0.64 \) in the core and a variable shell composition: \( 0.4 < y(Al) < 0.8 \). The measured value is shown by the red datapoint, and from the confidence interval the maximum deviation is \( \sim 7\% \).

Photoresponse and EQE measurements

We measured the spectral photo-response and external quantum efficiency (EQE) of the surface passivated InGaAs-InAlAs core-shell NW device. This was realized by dispersing the light emitted from a tungsten-halogen lamp via a monochromator (Spex 340E, 1200 1/mm) onto the sample. The resulting wavelength-dependent photocurrent was detected by a pyroelectric detector and lock-in technique without bias. Calibration was further performed against a standardized Si solar cell, allowing to derive absolute values for EQE. As displayed in Figure S3 a photo-response is observed to illumination in the range of \( \sim 700 – 1100 \) nm, with a maximum EQE of \( \sim 15\% \) at \( \sim 900-1000 \) nm which is typical for such NW/Si PV cell [1,2]. Approaching the Si absorption edge (\( \lambda > 1100 \) nm) the photoresponse is drastically decreased and EQE vanishes. This suggests a negligible contribution of the NWs to the photocurrent while most carriers are generated in the Si substrate. In addition, the photoresponse towards the ultraviolet region is also strongly reduced.
Figure S3. External quantum efficiency (EQE) for the surface passivated InGaAs NW array on Si as measured without bias via lock-in technique.

This is consistent with recent calculations of the illumination-wavelength dependent absorption cross-section in InAs-NW/Si photodiodes, i.e., high-energy photons have only short penetration depths and do not sufficiently reach the p-n heterojunction at the substrate interface [2]. Further improvements in EQE will need to consider specific strategies, including optimization of the interwire distance, size and Ga-content of the NW arrays. All these will impact the absorption cross-section and additional spectral matching with the solar spectrum. Ultimately, decoupling the effective absorption and minority carrier collection will require implementation of the surface passivated InGaAs-InAlAs NWs into radially arranged p-n heterojunctions.

[1] W. Wei, X. Y. Bao, C. Soci, Y. Ding, Z. L. Wang, and D. Wang, Nano Lett. 9, 2926 (2009).

[2] A. D. Mallorqui, E. Alarcon-Llado, E. Russo-Averchi, G. Tütüncüoglu, F. Matteini, D. Rüffer, and A. Fontcuberta i Morral, J. Phys. D 47, 394017 (2014).