Supporting Information

MBE growth of GaAs/InAs core-shell nanowires and fabrication of InAs nanotubes

Torsten Rieger[1,2], Martina Luysberg[3], Thomas Schäpers[1,2,4], Detlev Grützmacher[1,2] and Mihail Ion. Lepsa[1,2]*

[1] Peter Grünberg Institute 9, Forschungszentrum Jülich, 52425 Jülich, Germany

[2] Jülich Aachen Research Alliance for Fundamentals of Future Information Technology (JARA-FIT), Germany

[3] Peter Grünberg Institute 5 and Ernst Ruska-Centre (ER-C) for Microscopy and Spectroscopy with Electrons, Forschungszentrum Jülich, 52425 Jülich, Germany

[4] II. Institute of Physics, RWTH Aachen University, 52074 Aachen, Germany

E-Mail: t.rieger@fz-juelich.de

1. Substrate preparation and optimized growth parameters of GaAs/InAs core-shell NWs

Substrate preparation:

- GaAs (111)B wafers were covered with a spin-coated solution of Hydrogen Silsesquioxane (HSQ) and Methylisobutylketone (MIBK) 1:20 in volume.
- Baked at 300°C for 10 minutes.
- Loaded into the MBE system.

Growth details (for the best shell morphology):

- Wafers were heated to 590°C for 30 minutes under constant As flux (10^{-6} torr).
- GaAs growth was initiated by opening the Ga shutter (Ga rate 0.075 µm/h).

- 45 minutes growth of GaAs.
- Consumption of the Ga droplet by keeping the same substrate temperature (590°C) and As flux (10-6 torr) for 15 minutes.
- Ramping down the substrate temperature to 490°C.
- Opening the In shutter in order to start the shell growth.

Growth Parameters:

	GaAs core	**InAs shell**
Temperature	590°C	490°C
Ga rate	0.075 µm/h	---
In rate	---	0.1 µm/h
As flux	10^{-6} torr	10^{-6} torr

2. EDX spectra of the upper part of the GaAs/InAs core-shell NW

Three EDX spectra were acquired on a single NW at different positions marked in Fig. S1a. Point A corresponds to the top end of the NW, Point B is the part not showing any Moiré fringes and Point C is the main part of the NW. The EDX spectra are shown in Fig. S1b. As seen, the Indium is detected in the main part and on the top end while there is no Indium grown at Point B.

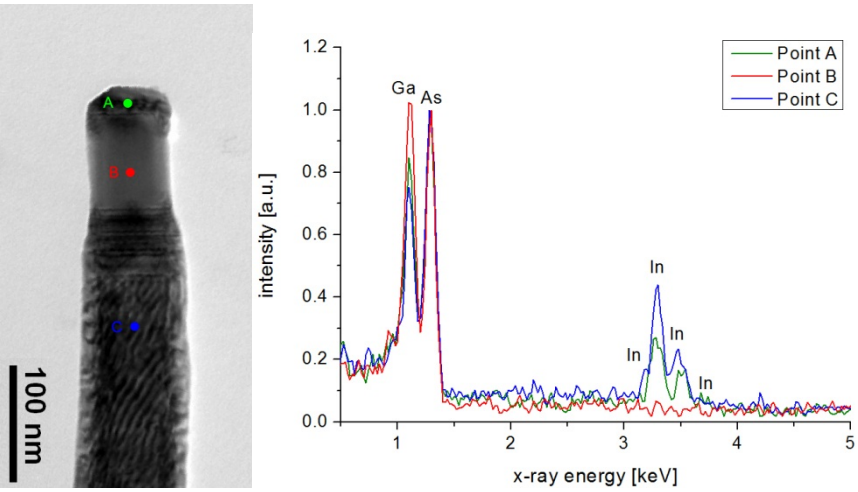

Figure S1. (a) TEM micrograph of the upper part of a GaAs/InAs core-shell NW showing three positions where EDX spectra were acquired. (b) EDX spectra taken from the positions marked in (a).

3. TEM of InAs islands

We have examined by TEM the core-shell NWs after one minute of shell growth time in order to further point out the islands-based growth mechanism as well as to investigate the nucleation of the islands. In Fig. S2, a TEM micrograph is shown which does not show Moiré fringes around the entire NW. The Moiré fringes appear only on the positions where the two crystal lattices overlap. Thus, when the fringes are not observed, there is only GaAs

demonstrating that the growth of the shell starts from islands nucleated on the different side facets of the NWs.

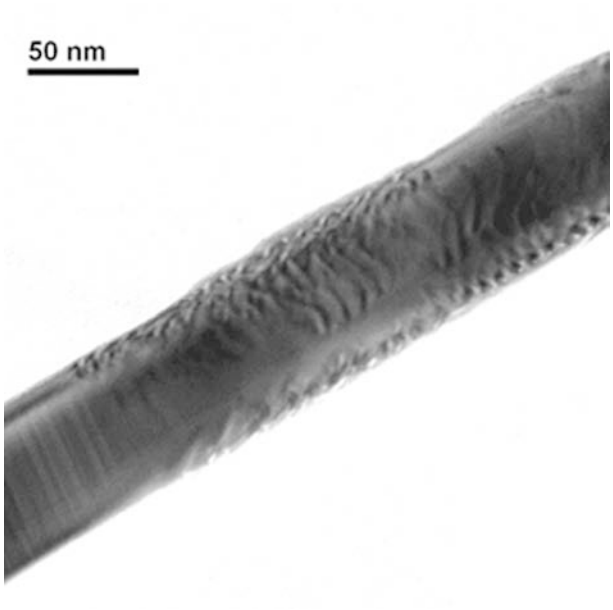

Figure S2. TEM micrograph after 1 min shell growth time

Fig. S3 shows a HRTEM micrograph of a single InAs island. No stacking fault or rotational twin are found which demonstrates that the island nucleation does not necessarily starts at a twin or stacking fault.

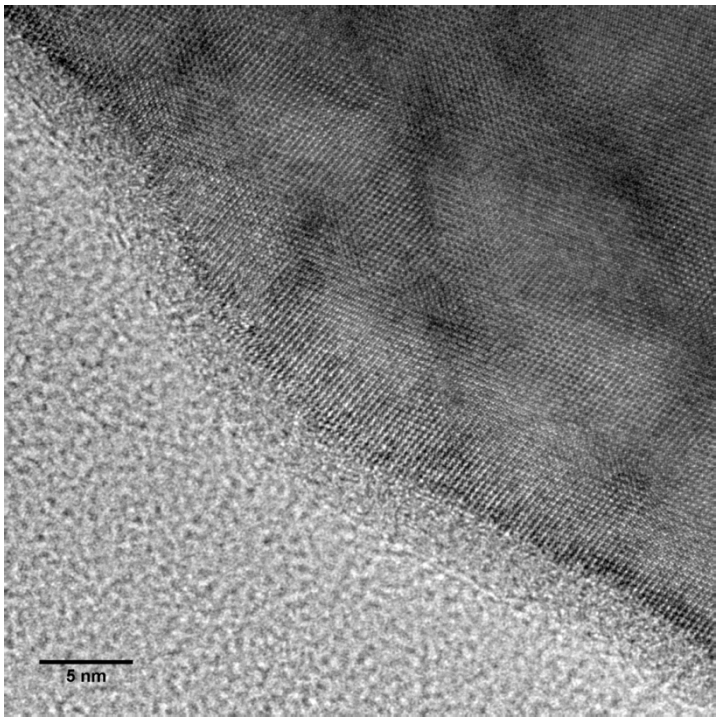

Figure S3. Single InAs island without stacking fault.

In Fig. S4, a HRTEM micrograph is presented, showing three misfit dislocations in an InAs island. One of them is located at a rotational twin in the core, while the other two are very close together. The latter two might have the same origin and are just due to a cross slip of the dislocation. This cross slip can help relieving the tangential strain.

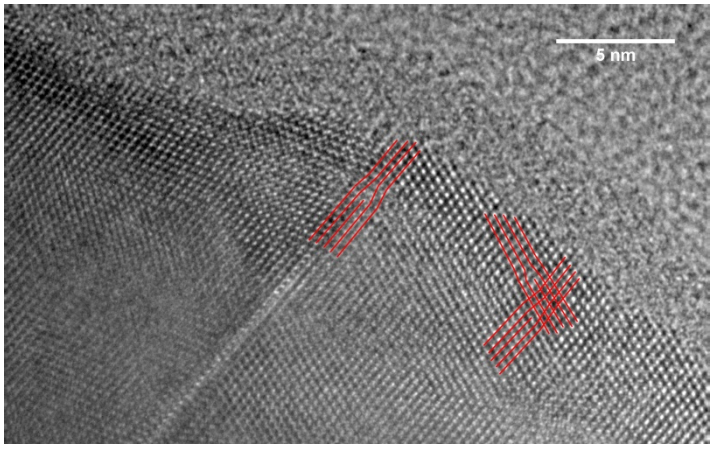

Figure S4. HRTEM of different misfit dislocations in an InAs island.

4. Additional information about defects in the InAs shell observed by TEM

Other defects in the crystal structure of the InAs shell are illustrated in Fig. S5 by TEM micrographs. The red guiding lines in Fig. S5a draw the attention on stacking faults which are perpendicular on other <111> directions. Also, stacking faults not being perpendicular to any <111> direction (islands boundaries) and threading dislocations are revealed in Fig. S5b by careful examination of the Moiré fringes observed in the TEM micrograph of NWs with a 3 min shell growth.

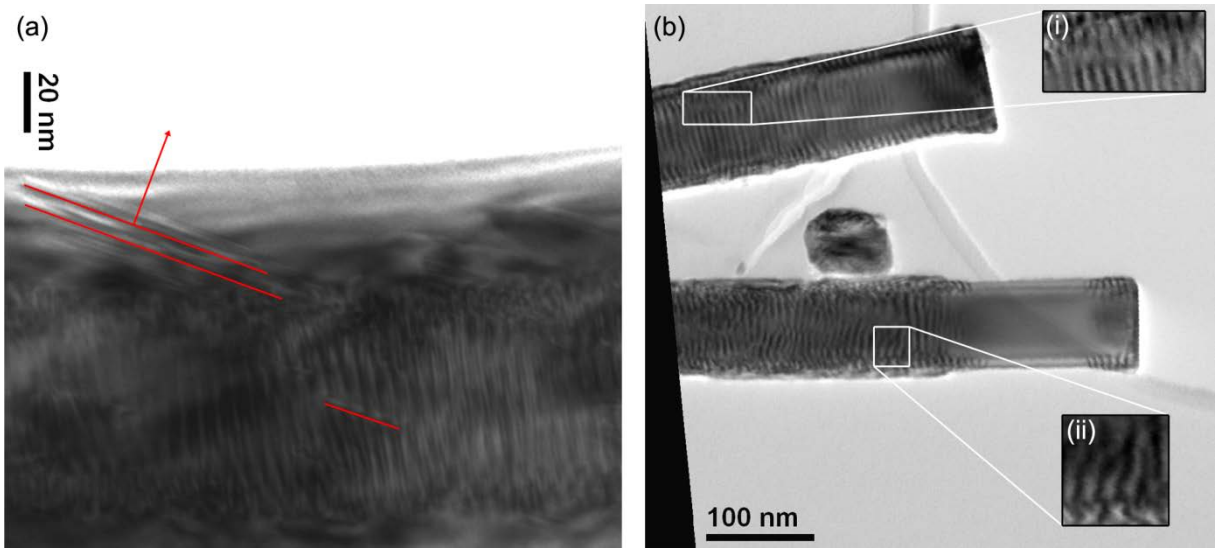

Figure S5. (a) TEM micrograph of stacking faults in other <111> directions. (b) TEM micrograph of GaAs/InAs core-shell NWs after 3 min of shell growth. (Inset (i)) Moiré fringes showing stacking faults in the shell, which are not adopted from the core and (Inset (ii)) Moiré fringes showing a threading dislocation.

Both defects, islands boundaries and threading dislocations, are supposed to appear due to the coalescences of the islands as schematically depicted in Fig. S6: in the beginning, the islands nucleate on arbitrary positions on the NW side facets. Already in this stage, the islands contain misfit dislocations (Supporting Information, section 2). As the islands do not influence each other, the dislocations inside the islands are on the energetically favorable

positions. When the islands coalesce, at the boundaries between different islands the local atomic arrangement does not match exactly. As the result, stacking faults as well as threading dislocations are formed.

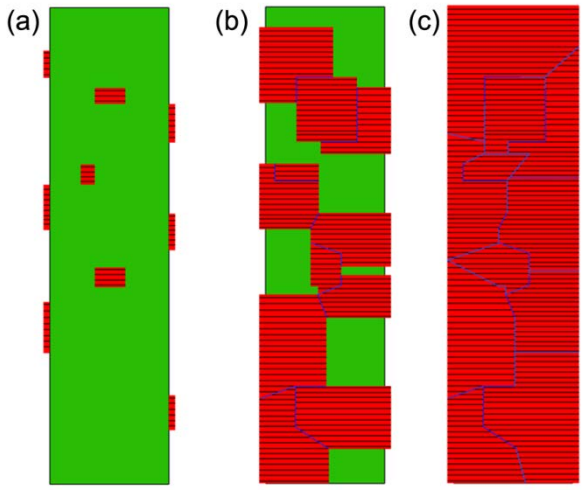

Figure S6. Growth mechanism of the InAs shell: (a) Islands formation with dislocations (b) and (c) Island coalescence forming boundaries containing islands boundaries and threading dislocations.

5. HAADF micrograph and EDX profile of a GaAs/InAs core-shell NW

In Fig. S7, an HAADF micrograph and the corresponding EDX profile of a core-shell NW are presented. The results confirm the core-shell geometry and can be compared with the EDX profile model estimations from the next section.

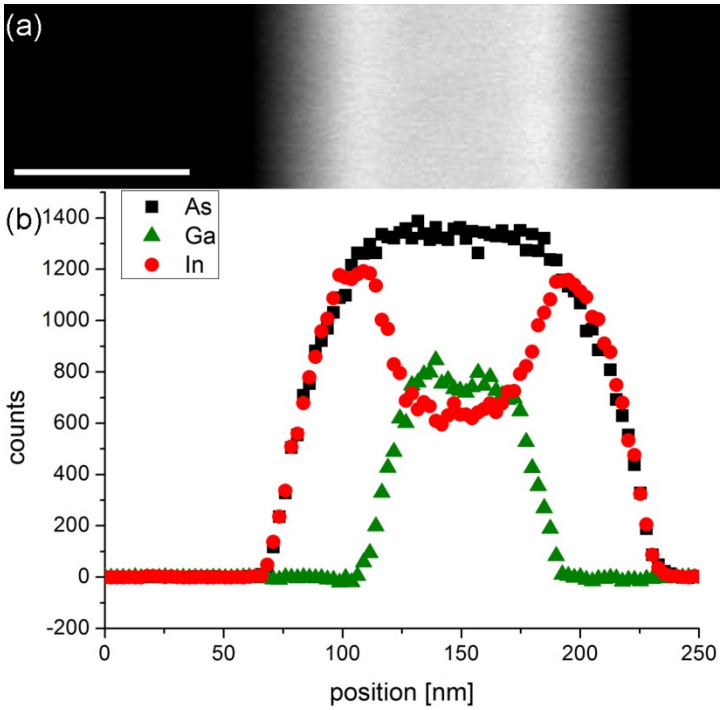

Figure S7. HAADF micrograph of a GaAs/InAs core-shell NW (a) and the corresponding EDX profile (b). The scale bar in (a) is 100 nm.

6. Model of the EDX profile of core-shell nanowires and nanotubes

The model assumes that the X-Ray intensity is proportional to the number of atoms, thus also the (local) thickness of the core $t_c(x)$ and shell $t_s(x)$ along the diameter. With a hexagonal morphology of the core and shell, d_c and d_{cs} being the diameter of the core and the core-shell nanowire, respectively, as well as setting $x = 0$ to the center of the wire, one gets

$$t_c(x) \propto \begin{cases} 0 & \text{for} \quad |x| > \frac{d_c}{2} \\ d_c \cdot \frac{\sqrt{3}}{4} & \text{for} \quad |x| \leq \frac{d_c}{4} \\ d_c \cdot \frac{\sqrt{3}}{4} - \left(x - \frac{d_c}{4}\right)\tan(60°) & \text{for} \quad \frac{d_c}{2} \geq |x| > \frac{d_c}{4} \end{cases}$$

and

$$t_s(x) \propto \begin{cases} d_{cs} \cdot \frac{\sqrt{3}}{4} - t_c(x) & \text{for } |x| \leq \frac{d_s}{4} \\ \left[d_{cs} \cdot \frac{\sqrt{3}}{4} - \left(x - \frac{d_{cs}}{4}\right) \cdot \tan(60°)\right] - t_c(x) & \text{for } |x| > \frac{d_s}{4} \end{cases}$$

Thus, in a GaAs/InAs core-shell nanowire, Ga signal will follow $t_c(x)$ while the In signal follows $t_s(x)$. The As signal will follow the shape of the nanowire:

$$t_{As}(x) \propto \begin{cases} d_{cs} \cdot \frac{\sqrt{3}}{4} & \text{for } |x| \leq \frac{d_s}{4} \\ d_{cs} \cdot \frac{\sqrt{3}}{4} - \left(x - \frac{d_{cs}}{4}\right) \cdot \tan(60°) & \text{for } |x| > \frac{d_s}{4} \end{cases}$$

Fig. S8 shows the expected signals for different core-shell geometries (a, b), nanotubes (c), nanotubes with a thin alloyed interface (d) as well as core-shell nanowires with a thin alloyed interface (e). Red lines correspond to the In signal, green ones to the Ga signal. In the schematically shown geometries, InAs is shown in red, GaAs in green and alloyed regions in blue. The model results can be compared with the experimental data presented in Figs. S7 and 5e for the core-shell NWs and InAs nanotubes, respectively.

As seen, a small alloying at the interface will let the EDX signals in the in core-shell NWs almost unaffected, whereas in the nanotube, the alloying is easily seen by the presence of the Ga signal as well as an small increase in the Ga signal near the inner edges of the tubes. This small increase can even be observed in the EDX profile shown in Fig. 5e.

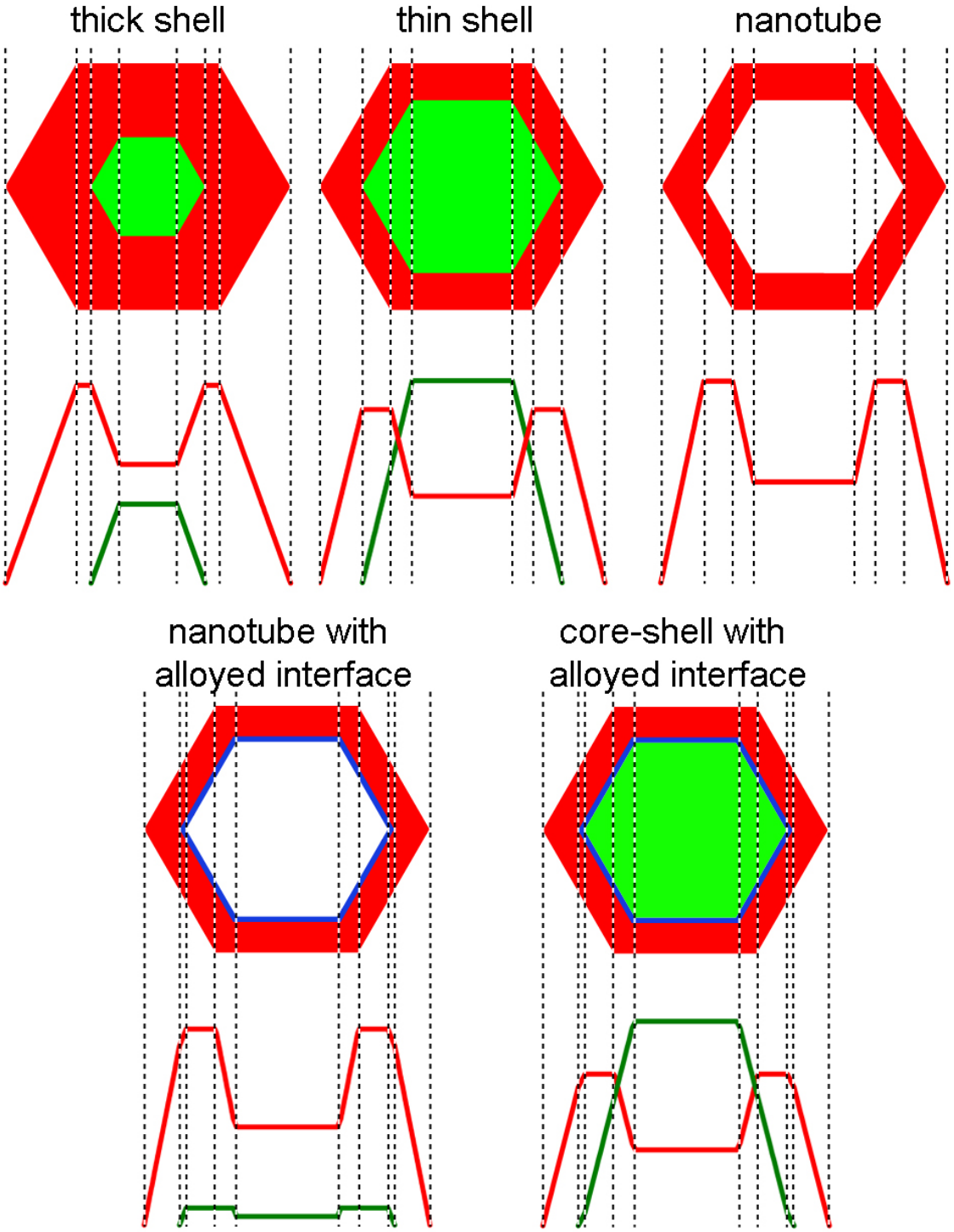

Figure S8. Geometry and expected EDX profiles for core-shell and nanotube structures with and without alloyed interfaces.