Methods and Supplementary Information:

Nanowire Synthesis

GaP and InP nanowires were synthesized using a pulsed laser vaporization apparatus (Fig. 1B). A target of well-mixed GaP (or InP) and Au powders ($(GaP)_{0.95}Au_{0.05}$ or $(InP)_{0.95}Au_{0.05}$) was pressed under uniaxial pressure in a Carver laboratory press at ~4,000 psi. To enhance mechanical stability the targets were then compacted further using an isostatic press (Autoclave Engineers) for 3 minutes at a pressure of 30,000 psi. Two quartz tubes were centered in a tube furnace as shown schematically in Fig. 1B. The carrier gas was first passed through the space between the outer and inner tube to pre-heat before entering the reaction zone. The target was placed in the inner tube (inner diameter 1 inch) just outside the furnace to avoid thermal evaporation of the target materials. A pulsed Nd: YAG laser (Telescopic Series SL803, Spectron Laser Systems) with a pulse width of 15 nanoseconds and repetition rate of 10Hz was used to vaporize the targets. Both the fundamental (1064nm at 850mJ/pulse, delayed by ~40 nanoseconds) and first harmonic (532nm at 450mJ/pulse) lines were present. The plume generated by the laser was carried down the central quartz tube by a flow of 100 sccm Ar at a pressure of 200 Torr. Temperature profiles such as shown in Fig. 1c for GaP were measured using a type K thermocouple. Similar profiles (i.e., flat midzone profile and gradient midzone profile) were used for InP. For GaP studies, the temperature at the center of the furnace was ~ 900 °C and for InP it was ~ 820 °C. A midzone temperature gradient was established by offsetting the left zone by –100 °C and the right zone +100 °C, while keeping the center zone 900 °C for GaP and 820 °C for InP. These offsets generated midzone gradients dT/dz ~ 3.4 °C /cm.

The as-grown nanowires were collected from the inner tube wall at point B (Fig. 1) ~ 5-10 cm away from the end of the tube furnace. Above Fig. 1C, we indicate the approximate drift time vs position for the nanoparticle/nanowire to drift in the carrier gas from the target (carrier gas velocity ~ 20 cm/min).

Transmission Electron Microscopy

To prepare grids for TEM, the GaP and InP nanowires were first dispersed in isopropanol by ultrasonication to obtain a metastable suspension. Ultrasonication must be carried out at low power to prevent breaking the nanowires. The time used to obtain a uniform suspension was only ~10 seconds. Then a drop of the nanowire suspension was placed onto a copper/lacy carbon TEM grid (Electron Microscopy Sciences, Inc) and the solvent was allowed to vaporize before introducing the grid into the microscope. TEM images were collected using a JOEL 2010F transmission electron microscope operated at 200 kV. Fourier transform and filtering were done using Gatan Digital Micrograph (Gatan, Inc.) described as follows. A region-of-interest was chosen from HRTEM images and then a Fourier transform was carried out to see the electron diffraction pattern. The spots diffracted from specific planes were selected and masked, and then an inverse Fourier transform was carried out. After filtering, certain lattice fringes were enhanced such that structural detail can be seen. The distance between adjacent twinning boundaries was also measured by Gatan Digital Micrograph.