Supporting Information for "Electron Mobilities Approaching Bulk Limits in "Surface-Free" GaAs Nanowires"

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Experimental details

Nanowire growth

Prior to growth, GaAs(111)B substrates were functionalized with poly-L-lysine. Colloidal Au nanoparticles of 50 nm diameter were then applied to the substrates. Nanowires were grown at a

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pressure of 100 mbar and a total gas flow rate of 15 slm. Trimethylgallium (TMGa), trimethylaluminium (TMAl), and arsine (AsH_3) were used as the Ga, Al, and As sources, respectively. GaAs nanowire cores were grown via a two-temperature procedure, comprising an initial nucleation step at 450 °C for 1 minute followed by 45 minutes growth at 375 °C.[1] AlGaAs shell growth and GaAs cap growth were performed at 750 °C. For AlGaAs shell growth, the group III vapor phase fraction, TMAl/(TMAl + TMGa), was $\chi_{v,Al} = 0.5$, yielding a shell composition of $Al_{0.4}Ga_{0.6}As$.[2]

Electron microscopy

Field emission scanning electron microscopy (FESEM) was carried out using a Hitachi S4300 FESEM at an accelerating voltage of 3 kV.

Cross-sectional TEM samples were prepared by embedding the nanowires in resin and microtoming to obtain thin slices of approximately 50 nm in thickness. TEM was performed using a Phillips CM300 TEM operated at 300 kV. Figure S.1 illustrates cross-sectional TEM images of core–shell–cap nanowires grown with different AlGaAs shell thicknesses.

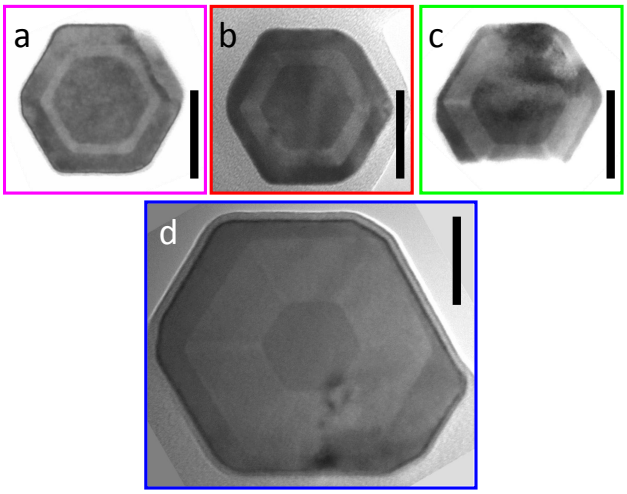

Figure S.1: Bright field TEM images of cross-sections of GaAs/AlGaAs/GaAs core–shell–cap nanowires. The AlGaAs shell thicknesses are (a) 5 nm (b) 10 nm, (c) 16 nm and (d) 34 nm. Scale bars are 50 nm.

Photoluminescence

Figure S.2 shows a typical photoluminescence spectrum, with an emission peak at 1.43 eV (865 nm), characteristic of band-edge emission from GaAs.

For time-correlated single photon counting measurements, the sample was excited at 770 nm with the output from a mode-locked Ti:Sapphire laser oscillator generating 100 fs pulses at an 82 MHz repetition rate.

For photoluminescence up-conversion measurements, the sample was excited at 750 nm with the output from a mode-locked Ti:Sapphire laser oscillator generating 100 fs pulses at an 82 MHz repetition rate. The PL was gated optically in a β-barium borate crystal using a separate part of the laser output that was subjected to an adjustable time delay relative to the excitation pulse. Time-resolved PL was measured at 865 nm with a liquid-nitrogen cooled CCD detector. The spectral resolution of the PL up-conversion system at the detection wavelength was 32 meV with a temporal resolution of 250 fs.

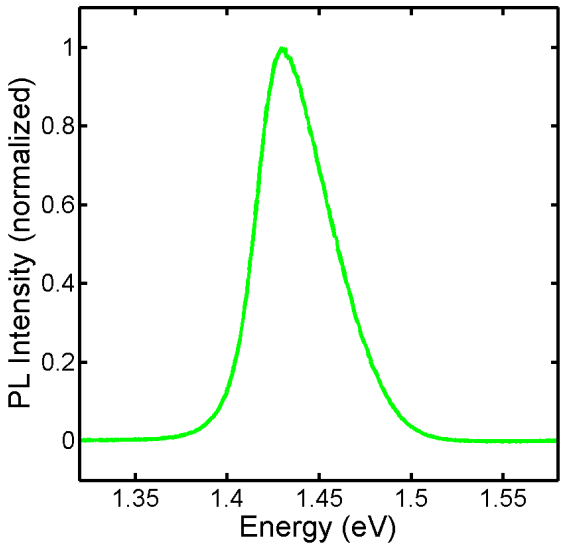

Figure S.2: Photoluminescence spectrum of GaAs/AlGaAs core–shell–cap nanowires with 16 nm thick AlGaAs shells. The photoexcitation pump fluence was 3 μJ/cm^2.

Effect of GaAs cap on mobility

As reported in the main manuscript, both the AlGaAs shell and the GaAs cap play a role in separating charge carriers in the GaAs core from scattering sites on the nanowire surface. Figure S.3 compares the electron mobilities measured in core–shell and core–shell–cap nanowires. The core–shell–cap nanowires exhibit higher electron mobilities due to the addition of the GaAs cap.

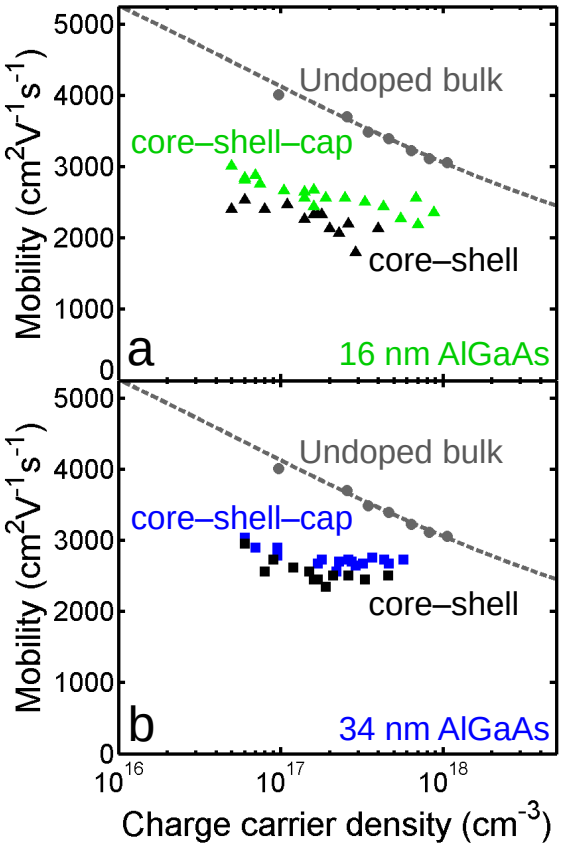

Figure S.3: Carrier mobilities versus photoexcited carrier density measured for core–shell (black symbols) and core–shell–cap (colored symbols) nanowires with (a) 16 nm thick AlGaAs shells and (b) 34 nm thick AlGaAs shells. The carrier mobilities were extracted from photoconductivity spectra obtained at various fluences and times after photoexcitation. Electron mobilities, determined by Sharma *et al.*,[3] for photoexcited undoped bulk GaAs are also shown (gray circles). The dashed line is an empirical fit to the bulk data using the Caughey–Thomas relation.[3,4]

Effect of annealing

To examine the effects of annealing, bare GaAs nanowires were grown as described in the main manuscript, then subjected to an annealing step at 750 °C for 12 minutes. Thus, these bare nanowires were exposed to the same temperature conditions as the GaAs/AlGaAs/GaAs core–shell–cap nanowires with 16 nm shells, including temperature ramp and precursor flow stabilization steps. Figure S.4 shows SEM images of unannealed and annealed GaAs nanowires, together with photoconductivity data. Annealing does not appear to change the morphology of the nanowires. The photoconductivity lifetimes, photoconductivity spectra and carrier mobilities of the annealed GaAs nanowires were indistinguishable from those of the unannealed GaAs nanowires. This suggests that annealing does not significantly affect either the carrier mobility or the carrier lifetime.

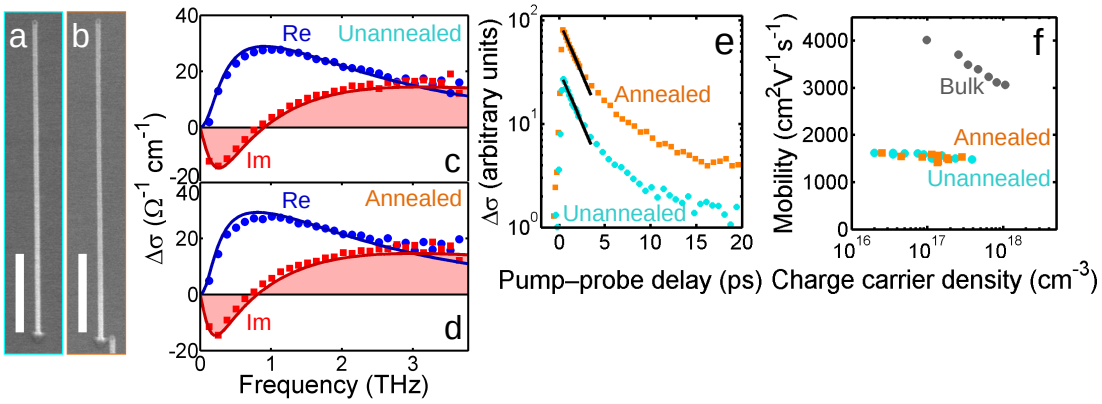

Figure S.4: Comparison of unannealed and annealed GaAs nanowires. SEM images of (a) unannealed and (b) annealed GaAs nanowires. Photoconductivity spectra obtained at 4 ps after photoexcitation with a fluence of 10 μJ/cm^2 for (c) unannealed and (d) annealed nanowires. (e) Photoconductivity ($\Delta\sigma$) decays measured for annealed (orange squares) and unannealed (light blue circles) bare GaAs nanowires. Decays were measured under a photoexcitation fluence of 10 μJ/cm^2. Straight lines are monoexponential fits to the decays within the first 3 ps, with time constants of 2.2 ps. Decays are scaled for clarity. (f) Carrier mobilities versus photoexcited carrier density measured for annealed (orange squares) and unannealed (light blue circles) bare GaAs nanowires. The carrier mobilities were extracted from photoconductivity spectra obtained at various fluences and times after photoexcitation. Electron mobilities, determined by Sharma *et al.*,[3] for photoexcited undoped bulk GaAs are also shown (gray circles).

References

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