Characterization of core-shell GaAs/AlGaAs nanowire heterostructures using advanced electron microscopy

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Abstract. To explore the unique properties of the nanoscale, advanced fabrication and characterization techniques are required. Specifically, analyses in two orthogonal directions, plan-view and cross-section, were used to prove the core-shell morphology of GaAs/AlGaAs nanowires and determine their cross-section to be hexagonal. High-resolution transmission electron microscopy and high angle annular dark field scanning transmission electron microscopy confirmed the core-shell interface to be defect-free, coherent, and sharp (<1nm). Energy dispersive X-ray spectroscopy determined the shell composition to be Al₀.₉Ga₀.₁As uniformly along the length of the nanowire. These results demonstrate the power of electron microscopy to aid the development of semiconductor nanotechnology.

1. Background and motivation

Semiconductor nanowires have emerged as a promising new platform for nanoscale electronics, optoelectronics, and sensors. Core-shell nanowire heterostructures possess three distinct advantages over thin film systems: defect-free integration of highly misfit materials [1], the potential to study one dimensional physics [2] and the potential to create integrated nanowire devices [3].

The GaAs/AlGaAs system is of significant interest for high mobility electronics because it possesses intrinsically high electron mobility that can be further enhanced in two dimensional GaAs/AlGaAs heterostructures. GaAs/AlGaAs core-shell nanowire heterostructures would represent a unique model-system to achieve one dimensional carrier confinement. Initial studies [4,5] have demonstrated that core-shell nanowire fabrication may be realized in GaAs/AlGaAs, and in-depth structural analysis of GaAs/AlGaAs [6] core-shell nanowires is needed to develop nanowire devices.

In this work, the controlled growth of core-shell GaAs/AlGaAs nanowires is demonstrated. Chemical analyses by energy dispersive X-ray spectroscopy (EDX) confirm the core-shell morphology with a shell composed of Al₀.₉Ga₀.₁As. Structural analysis by transmission electron microscopy (TEM) and lattice-resolved high angle annular dark field (HAADF) scanning transmission electron microscopy (STEM) confirms the epitaxial shell deposition and the entire nanowire heterostructure to be defect-free and single crystalline.
2. Experimental
Core-shell GaAs/AlGaAs nanowires were fabricated by the vapour-liquid-solid mechanism [7] in a metal-organic chemical vapour deposition reactor as described previously [6]. Plan-view samples were prepared by ultrasonically removing nanowires in ethanol suspension and then depositing onto lacy formvar/carbon TEM grids. Cross-sectional samples were prepared as shown in Figure 1a, a plastic coverslip was rubbed on the surface of the as-grown substrates to collect the nanowires. The plastic coverslip was then embedded in epoxy and sliced approximately 75nm thick by microtomy (Leica and RMX using a Diatome Histo diamond blade).

The morphologies of as-grown nanowires were studied using a JEOL 6320FV scanning electron microscope (SEM). The structure and chemical composition of individual nanowires were characterized using a JEOL 2010F TEM, a VG HB603 STEM equipped with EDX, as well as a JEOL 2200FS aberration-corrected STEM. During high angular annular dark field (HAADF) STEM imaging of cross-sectional samples without carbon coating, the Z-contrast was clear but strong sample charging effects were observed, as shown in the fast Fourier transforms (FFT) shown in Figure 1b. After the samples were carbon coated, the sample charging was minimized, but this had considerable effects on the contrast at the core-shell interface. Therefore cross-sectional sample were imaged without carbon coating, coated with carbon, and then imaged again to determine both crystal structure and compositional profile.

![Figure 1](image-url)

**Figure 1.** a) A schematic of the TEM sample preparation procedures. b) HAADF STEM images with corresponding FFT for cross-sectional samples with and without carbon coating.

3. EDX chemical analysis
The compositions of the nanowires were determined by EDX analysis in both plan-view and cross-sectional view. EDX profiles were then fitted to a numerical model with Gaussian beam broadening:

$$
\Phi_Z(x) = \frac{1}{\sigma\sqrt{2\pi}} \int_{-\infty}^{\infty} h_Z(x') e^{-\frac{(x-x')^2}{2\sigma^2}} dx'
$$

(1)

Here $h(x')$ is the thickness profile integrated over the sample height of element Z and $\sigma$ is the Gaussian beam broadening width. The results from both plan-view and cross-sectional analysis were consistent with a GaAs core and an Al$_{x}$Ga$_{1-x}$As shell with $x = 0.89 \pm 0.03$. For spectra acquired on the VG HB603 $\sigma = 0.5$ nm. For results taken on the JEOL 2010F $\sigma = 3-4$ nm.
**Figure 2.** a) EDX linescan data from a core-shell nanowire collected in plan-view and fitted to the model shown in equation 1. Inset is BF-STEM image of the nanowire. b) EDX linescan data from a core-shell nanowire collected in cross-section and fitted to the model shown in equation 1. Inset is DF-STEM image of the nanowire c) HAADF STEM image and corresponding EDX chemical maps.

4. Structural analysis

The structures of the nanowires were analyzed using SEM, TEM, and lattice resolved HAADF STEM. SEM inspection, as shown in Figure 3a, reveals that the nanowires grow vertical to the substrate. Bright-field (BF) TEM analysis, as shown in Figure 3b, reveals that the nanowires are zinc-blende single crystals growing along the <111> direction. The nanowires were imaged at multiple tilt angles and no dislocations or twin planes were observed.

For more detailed information about the core-shell interface, lattice-resolved HAADF STEM images, as shown in Figure 3c with <111> zone axis, of the GaAs/AlGaAs interface were taken. From these images it can be seen that the core-shell interface is epitaxial and coherent. Comparison of the HAADF-STEM with the corresponding BF-STEM, as shown in Figure 3d, demonstrates that the contrast in the HAADF image is caused by compositional variation. FFT of the lattice-resolved images determines the facets to be {110}.

**Figure 3.** a) SEM image of as-grown core-shell nanowires. b) BF-TEM image of core-shell nanowire. Inset is a corresponding SAD pattern. c) Lattice-resolved HAADF STEM image of the core-shell interface in cross-sectional view. Inset is FFT. d) The BF-STEM image corresponding to c).

5. Anisotropic shell deposition

In addition to the commonly observed regular hexagonal cross-section of GaAs/AlGaAs nanowire heterostructures [5], anisotropic shell deposition has also been observed. Plan-view STEM, as shown in Figure 4a, shows a core-shell morphology and appears similar to core-shell nanowires with regular...
hexagonal cross-section (Fig 3b). However, the anisotropic nature of the shell is clearly revealed in cross-sectional HAADF STEM, as shown in Figure 4b. This anisotropy is related to crystallographic facets of the nanowire core. If all facets of the nanowire core belong to one family of planes (\{110\} or \{112\}), then shell deposition is uniform. However, if the facets of the nanowire core belong to multiple families of planes, the shell deposition will be anisotropic. From FFTs of lattice-resolved HAADF STEM images, the shell thickness is seen to be greater on \{110\} facets than on \{112\} facets.

Lattice-resolved HAADF STEM images, as shown in Figure 4c, also reveal the shell to be amorphous. This is further demonstrated by a STEM shadow image, as shown in Figure 4d, in which the crystalline shell shows clear Kikuchi lines whereas the shell does not. The amorphous nature of the shell is unclear, but is likely due to oxidation of the Al-rich AlGaAs shells. Further investigation is underway to investigate growth on different nanowire facets.

Figure 4. a) DF-STEM image of anisotropic shell nanowire. b) HAADF STEM image of anisotropic core-shell nanowire in cross-sectional view. c) Lattice-resolved HAADF STEM image of core-shell interface. d) “Shadow” image of anisotropic shell nanowire in cross-sectional view.

6. Summary
In summary core-shell GaAs/AlGaAs nanowire heterostructures have been characterized using multiple electron microscopy techniques. HAADF STEM imaging gives information about both the structure and composition profile at the GaAs/AlGaAs interface. EDX analyses confirm the core-shell morphology and consistently determine the shell to be Al_{x}Ga_{1-x}As with \(x = 0.89 \pm 0.03\). BF-TEM and lattice resolved HAADF STEM structural analysis demonstrates the core-shell interface to be epitaxial, coherent, and sharp. As evidenced by anisotropic shell nanowires, both cross-sectional and plan-view analysis are necessary to determine the morphology of core-shell nanowires.

Acknowledgements
The authors thank EA Fitzgerald for access to MOCVD facilities, the MIT Center for Materials Science and Engineering, an NSF-funded MRSEC, for use of electron microscopy facilities, and ST Boles for MOCVD assistance and helpful discussions. SG acknowledges 3M, the Interconnect Focus Center, MIT startup funds, and the NSF CAREER Award (DMR-074555) for financial support. A portion of this research was conducted at the SHaRE User Facility, which is sponsored by the Division of Scientific User Facilities, Office of Basic Energy Sciences, U.S. Department of Energy.

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