Diffraction at GaAs/Fe_3Si core/shell nanowires: the formation of nanofacets

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Nanowires (NWs) combining a semiconductor and a ferromagnet in a core/shell geometry allow for a magnetization along the wire and thus perpendicular to the substrate surface.[1] Fe_3Si is lattice matched to GaAs. However, the difference of the surface energies of both materials leads to the strain-free Volmer-Weber island growth mode of Fe_3Si on GaAs.[2] Thus the initially isolated Fe_3Si islands can grow to their equilibrium shape before coalescence. The GaAs cores are facetted as hexagonal pillars. The Fe_3Si shells are not obliged to replicate the {110} oriented sidewalls of the cores, and indeed we find growth conditions, where they do not.

MBE grown GaAs/Fe_3Si core/shell NW structures on Si(111) were characterized by SEM, X-ray diffraction, and TEM. The X-ray experiments were performed in grazing incidence geometry (cf. sketch in Fig. 1) at the beamline BM 02 of the European Synchrotron Radiation Facility (ESRF) in Grenoble. The energy of the beam was 10 keV. The detector size is 560×120 pixels of size 130×130µm^2 each. In order to minimize the penetration depth into the substrate and the film between the NWs we took a very small angle of incidence of 0.04°. In this way the signal of the ensemble of NWs was always dominating the diffraction pattern. This was checked by observation of the vanishing signal of the single-crystal Si substrate, which would dominate the diffraction curve at larger angles of incidence. TEM images and selected area diffraction (SAD) patterns are acquired with a JEOL 3010 microscope operating at 300 kV. The cross-section TEM methods provide high lateral and depth resolutions on the nanometer scale, however they average over the projected thickness of the NW.

Figure 1 shows an SEM micrograph of the GaAs/Fe_3Si core/shell NWs. A relatively low area density of well oriented NWs $\rho_{NW} = 5\times10^8$ cm^{-2} is found as well as some hillocks. Regular step patterns are clearly visible on the sidewalls of the NWs. A poly-crystalline film covers the substrate surface between the NWs. The geometry of XRD in grazing incidence geometry is sketched on top of the micrograph. K_i, K_f, and K_d are the wave-vectors of the incident, reflected, and diffracted beams, respectively.

Figure 2 depicts the XRD curves of the (-220) reflection of GaAs/Fe_3Si core/shell NWs. The shells were grown at a temperature of 200°C. A radial ω–2Θ scan (thick red line) along the [-110] direction and an angular ω-scan perpendicular to the [-110] direction (thin black line), together with their full widths at half maximum (FWHM) are given. The FWHM of the diffraction peak amounts to 0.55° in the angular direction, whereas the FWHM in the radial direction 0.08° is lower by more nearly a factor of seven. The FWHM in angular direction here corresponds to the range of twist of the NWs, as in-plane reflections are used.[3] The range of tilt of GaAs NWs was measured earlier to be near 0.28° using symmetrical out-of-plane measurements. The radial scan shows thickness fringes (marked by arrows in Fig. 2) corresponding to a thickness of 14.3 nm equal to the Fe_3Si shell thickness.

Figure 3 demonstrates in-plane reciprocal space maps of the (-220)-reflection of GaAs NWs (left) and GaAs/Fe_3Si core/shell NWs (center and right). The growth temperatures of the Fe_3Si shells were 200°C (center) and 300°C (right). The crystallographic directions are sketched below. The facets of the pillar shaped cores

with hexagonal cross-section are clearly distinguished by streaks in the diffuse scattering pointing along the <110> directions. In the central map core streaks along the <110> directions are still visible although they are superimposed by additional streaks along the <1-21> directions originating from the tilted {1-11} facets of the Fe_3Si shells. In the map shown on the right side only streaks of the shell facets remain. Additional maxima above the main peak indicate interface reactions between Fe_3Si and GaAs occurring at T_S = 300°C.
Figure 4 demonstrates a multi-beam bright-field TEM micrograph and the corresponding SAD pattern illustrating the orientational relationship of a GaAs/Fe_3Si core/shell NW. We observe the coincidence of the core- and shell-orientations, i.e. here the Fe_3Si growth is predominantly epitaxial on the GaAs.[4] The straight line drawn on the SAD pattern near the (11-1) reflection is oriented perpendicular to the nanofacets of Fe_3Si visible in the corresponding micrograph. The separation of the diffraction spots along this line corresponds to the (111) net plane distance of Fe_3Si indicating that the nanofacets are mainly {111}-oriented. For the SAD the substrate was first oriented near the [011] zone axis. Then the NW had to be tilted a little further in order to keep the [011] zone axis orientation of the GaAs NW, because the axis of the NW was not exactly perpendicular to the Si surface. In the SAD pattern from a single core/shell NW the Fe_3Si maxima are stronger probably due to larger volume fraction of the shell. The fundamental reflections of the Fe_3Si are more intense than the super-lattice maxima.[5] Nevertheless we can distinguish higher order maxima evidencing, that the NW is properly oriented and that the crystallographic orientations of core and shell basically coincide. This underlines that a growth temperature of 200°C is well suited with respect to reach a highly perfect strain-free structure of the Fe_3Si shells. Density functional theory calculations reveal that the formation of {111} facets reduces the overall surface energy of the shells. Consequently a non negligible material transport occurs over distances small compared to the NW lengths. We thank the ESRF in Grenoble for providing beamtime during the experiment HC-1967. This work was supported in part by the Office of Naval Research through the Naval Research Laboratory's Basic Research Program. Some computations were performed at the DoD Major Shared Resource Center at AFRL.

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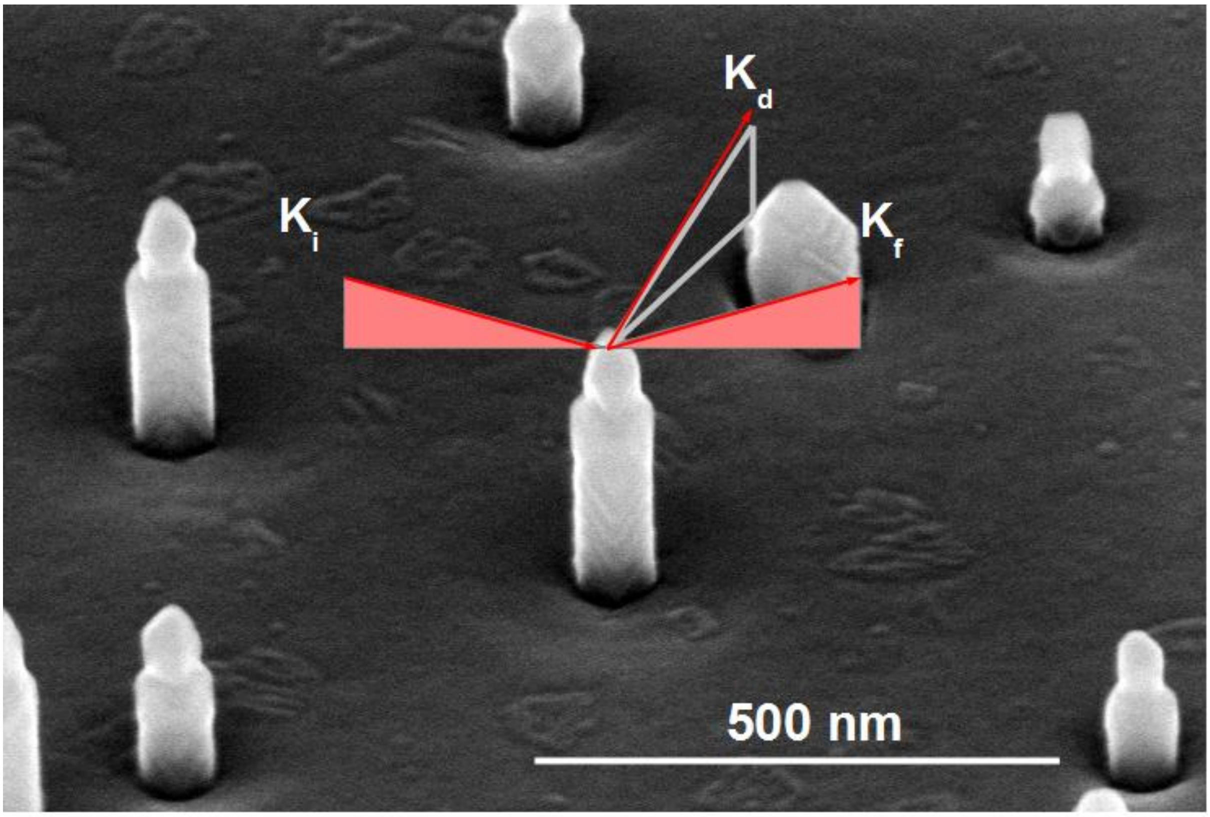

Figure 1: SEM image of GaAs/Fe_3Si core/shell NWs and islands grown by molecular beam epitaxy on a Si(111) substrate at a substrate temperature of T_S = 200°C. The geometry of XRD in grazing incidence geometry is sketched on top of the micrograph. Ki, Kf, and Kd are the wave-vectors of the incident, reflected, and diffracted beams, respectively.

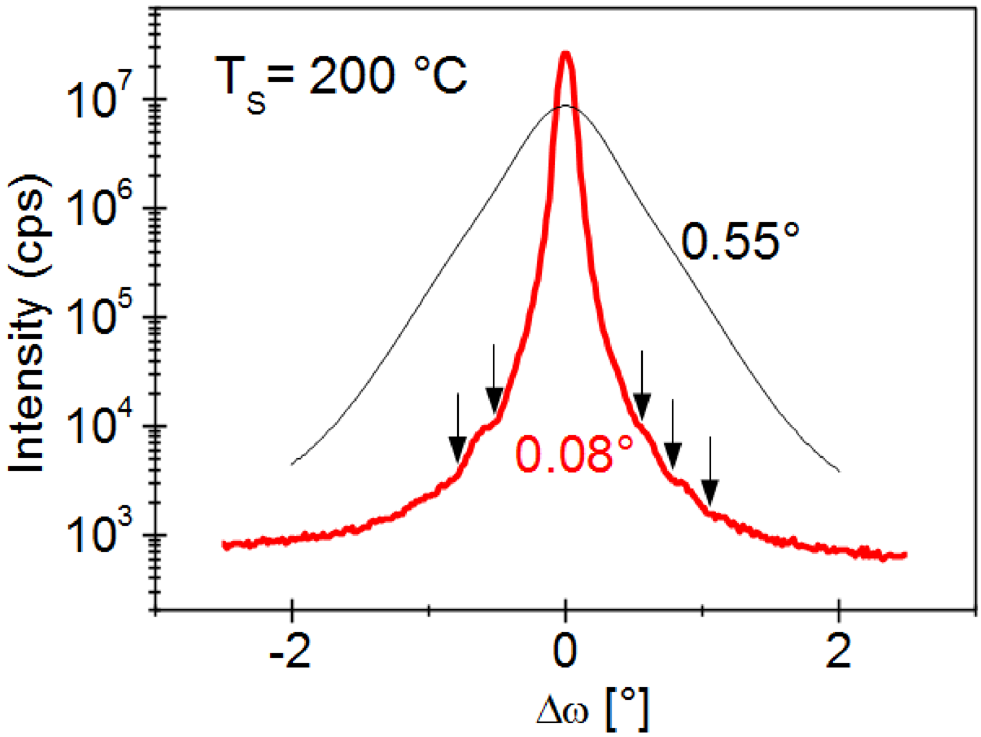

Figure 2: XRD curves of the (-220) reflection of GaAs/Fe_3Si core/shell NWs, T_S = 200°C. A radial ω2Θ-scan (thick line) along the [-110] direction and an angular ω-scan perpendicular to the [-110] direction (thin line), together with their full widths at half maximum (FWHM) are given. Thickness fringes are marked by arrows.

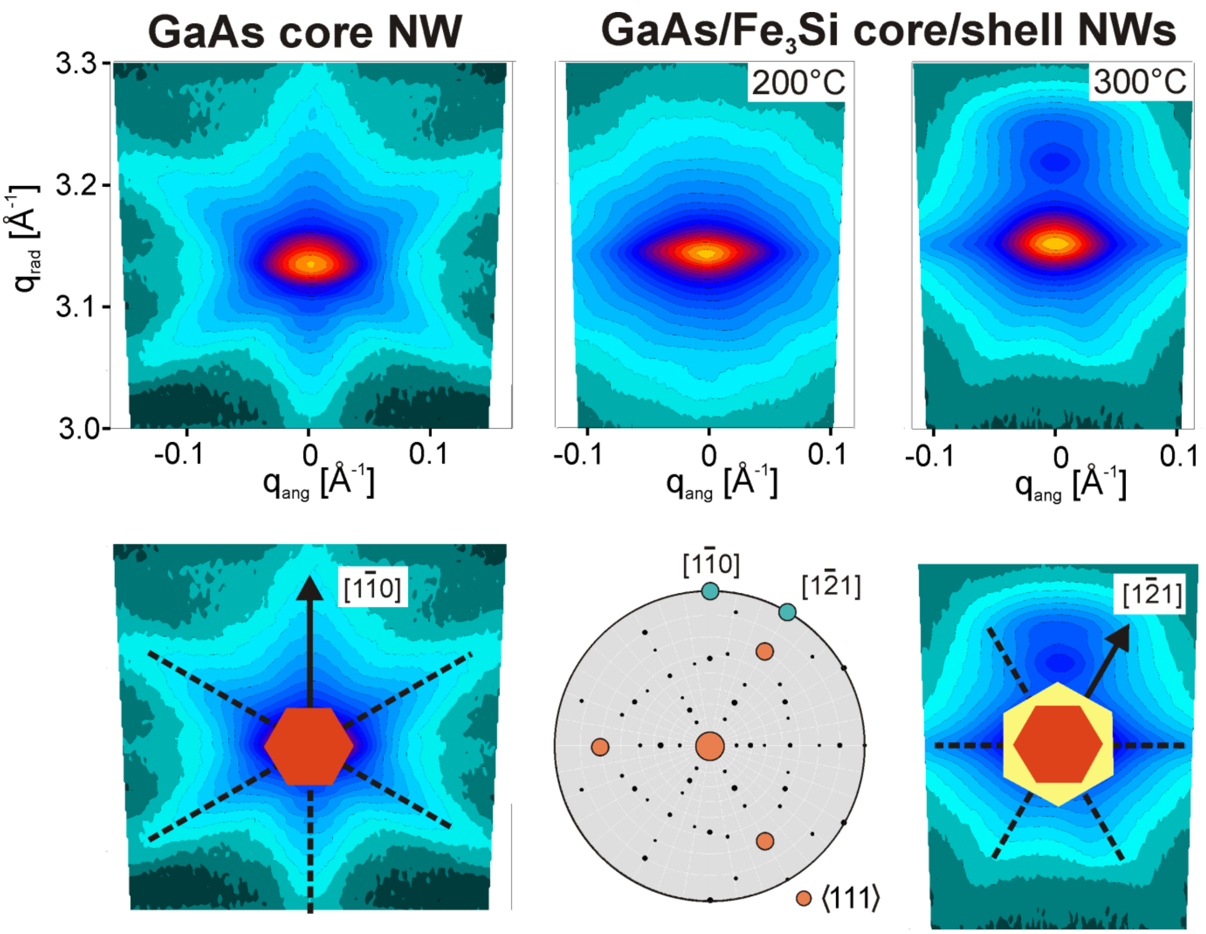

Figure 3: (-220) in-plane reciprocal space maps of [111]-oriented GaAs NWs (left) and GaAs/Fe_3Si core/shell NWs (center and right) grown by molecular beam epitaxy on a Si(111) substrate. The growth temperatures of the Fe_3Si shells are given above. The crystallographic directions together with the corresponding sidewalls are sketched below. In the symbolic stereogram (middle) the [1-10] and [1-21] directions are marked by blue circles and the different <111> directions by red circles. The radial direction of the scans is drawn vertically and the angular direction is directed horizontally in the figure.

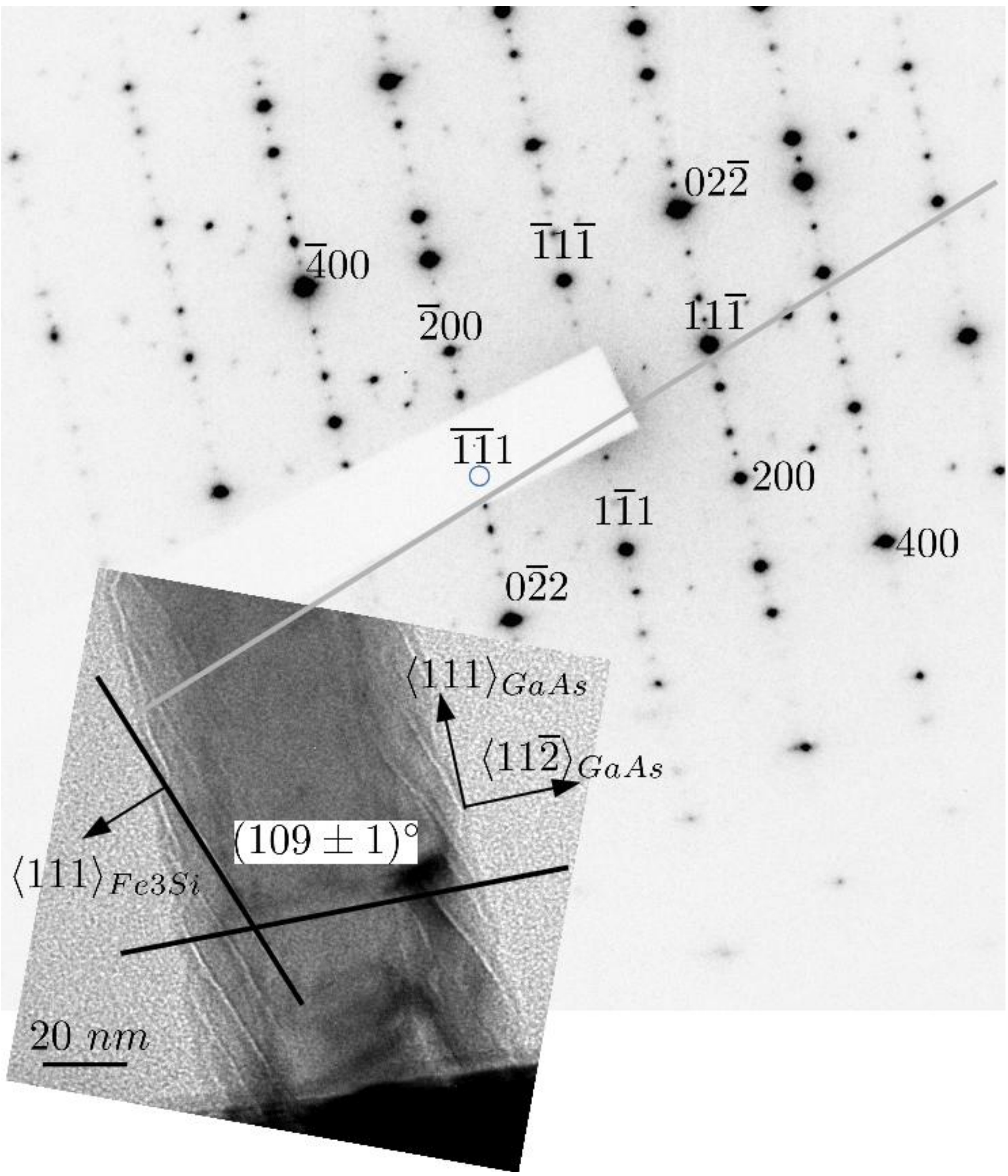

Figure 4: Multi-beam bright-field TEM micrograph and the corresponding SAD pattern illustrating the orientational relationship of a GaAs/Fe_3Si core/shell NW. The straight line near the (11-1) reflection is oriented perpendicular to the nanofacets of Fe_3Si visible in the corresponding micrograph.