Growth of Branched Single Crystalline GaAs Whiskers on Si Nanowire Trunks

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Introduction

Any successful method to integrate GaAs on Si by means of combining silicon integrated circuits with the optical and electronic opportunities of GaAs offers a significant impact to semiconductor technology. However, there are fundamental difficulties involved with the growth of a polar compound semiconductor on a nonpolar substrate such as GaAs on Si, leading to the problem of inversion domains and boundaries at the interface. Moreover, the lattice mismatch and the difference in thermal expansion coefficients have to be dealt with to obtain high-quality hybrid substrates of crystalline GaAs on silicon substrates. Much work has been done on planar growth of III-V materials on Si using different approaches such as buffer layers, growth on patterned Si surfaces, and selected area growth from small openings. Most of the problems can be avoided by reducing the contact area of the compound semiconductor and silicon, by means of a NW approach. The crystal lattice of the compound semiconductor will be elastically deformed and the strain could be relieved near the NW interface.

In most cases, nanowire growth has been performed by using a catalyst in the so called vapor-liquid-solid (VLS) or vapor-solid-solid (VSS) growth mechanism. We developed an approach for producing III-V (GaAs) nanowhiskers, on group IV (Si) NWs. For the first time, we show that single crystalline hexagonal GaAs whiskers can be grown hetero-epitaxially on the side facets of single crystalline Si-NWs by combining VLS and MBE growth techniques [1]. This addresses both, the long-time challenge of integrating high performance III-V semiconductors with mainstream Si technology, and the possibility of 3-dimensional nanoelectronic and -photonic devices.

Experimental

Si-NWs were grown in a hot wall LPCVD reactor by the VLS growth mechanism, and a 2 nm thick Au film was used as a catalyst. Alternatively, Au colloids with a mean diameter of 80 nm were spin-coated on the cleaned and etched Si (111) substrates. The VLS growth was performed at 823 K with 2% SiH_4 in He as a precursor gas; typical growth times in this study were 30 min. For the subsequent growth of GaAs whiskers the Si-NW/Si(111) samples were indium bonded to a molybdenum carrier wafer for growth in a Mod-Gen II MBE. After loading into the vacuum chamber the samples were baked at 773 K heater temperature before transfer into the growth chamber. The samples were grown between 723 and 823 K by increasing the substrate temperature to the desired growth temperature in an As_4 flux. The GaAs branches were grown at a 2-D equivalent growth rate of 0.55 µm/h under an As_4 beam equivalent pressure of 1.5×10^{-5} mbar.

Results

Most of the Si-NWs synthesized via the VLS process on Si (111) substrates with a thin 2 nm Au layer as the catalyst grew upwards from the substrate in the <111> directions. To achieve individual and freestanding Si-NW trunks we used 80 nm gold colloids as catalyst for the VLS growth. The typical dimensions are about 104 nm for the diameter and 3 µm for the length, which appear to be uniform. The HRTEM and diffraction image in Fig. 1 (a) confirms the [111] growth direction and the single crystalline nature of the Si-NWs. Such images clearly show the Si (111) atomic planes (separation 3.14 Å) perpendicular to the NW axis. The Si-NWs are usually free of dislocations and stacking faults and are covered by a very thin amorphous oxide layer.

The SEM image in Fig. 1 (b) shows the typical morphology of the samples after the MBE growth with branched GaAs whiskers grown on Si-NW/Si(111) samples. The GaAs branches, which were grown perpendicular to the Si (111) NW trunks, appear as perfectly aligned star-like structures with a 6-fold symmetry. The low magnification image shows the large quantity of these star-like structures, which densely cover a 1x1 cm2 sample. Fig.1 (c) shows the top view SEM image of individual freestanding Si-NWs with hetero-epitaxially grown GaAs whiskers, which in the tilted view of Fig. 1 (d) appear as vertically oriented nanotrees with Si-NW trunks and GaAs branches.

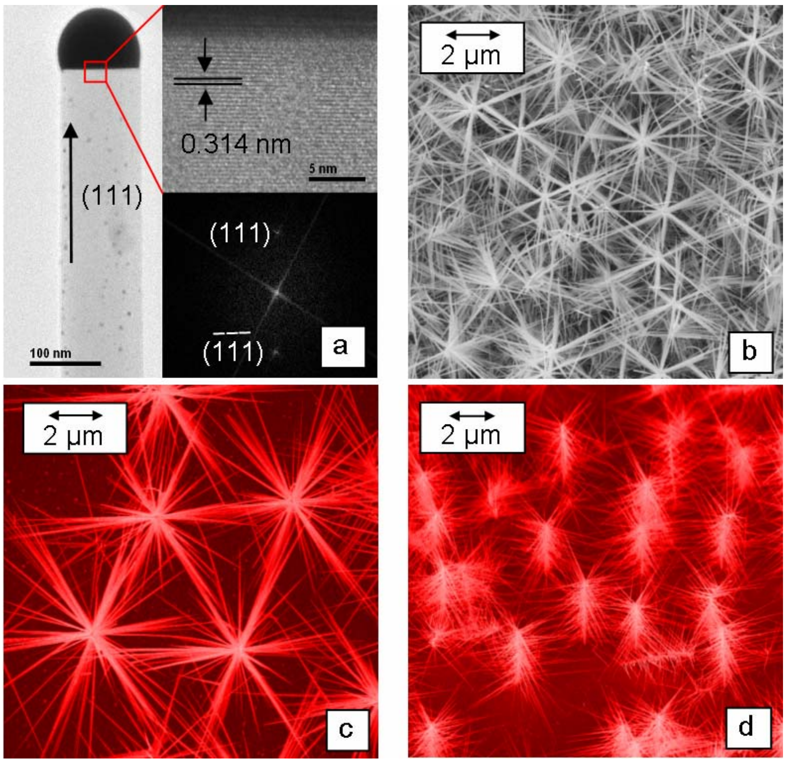

Fig. 1: (a) TEM, HRTEM and diffraction image of a single crystalline Si-NW with the catalytic Au nanoparticle on top. (b) low magnification SEM image showing the large quantity of GaAs nanowhiskers on Si-NWs; MBE growth of the GaAs nanowhiskers was performed at 723 K. (c) Top view SEM image of individual structures comprising GaAs branches with a 6-fold symmetry on the freestanding Si-NW trunks and (d) a 45° tilt SEM micrograph of GaAs branches growing perpendicular to the core Si (111)-NW trunks.

The Fig. 2 (a) shows the top view SEM image of a single nanotree with a 6-fold symmetry. TEM tilting experiments showed that the cross-section of each core Si-NW is a trigonal hexagon with three long and three short edges. The GaAs nanowhiskers grow on each of the six facets perpendicular to the core Si-NWs like branches on the trunks of a tree. The cross-sectional HRTEM image of such a nanotree in Fig. 2(b) shows these hexagon of the Si-NW trunk with alternating wider and narrower vertical {112} facets normal to the [111] growth direction. The angle between each of these adjacent facets of the Si-NWs is 120°, leading to the 6-fold symmetry as observed. In a few cases the GaAs whiskers grew preferably on the long facets of the hexagon leading to nanotrees with a quasi 3-fold symmetry (see Fig. 2 (c)), suggesting a minimal facet size for whisker nucleation. For long MBE growth times the core Si NW is covered by an unintentionally deposited GaAs layer (see Fig 2(b)) and some of the GaAs whisker growing later on this core-shell structure lose the crystallographic relation to the core Si-NWs.

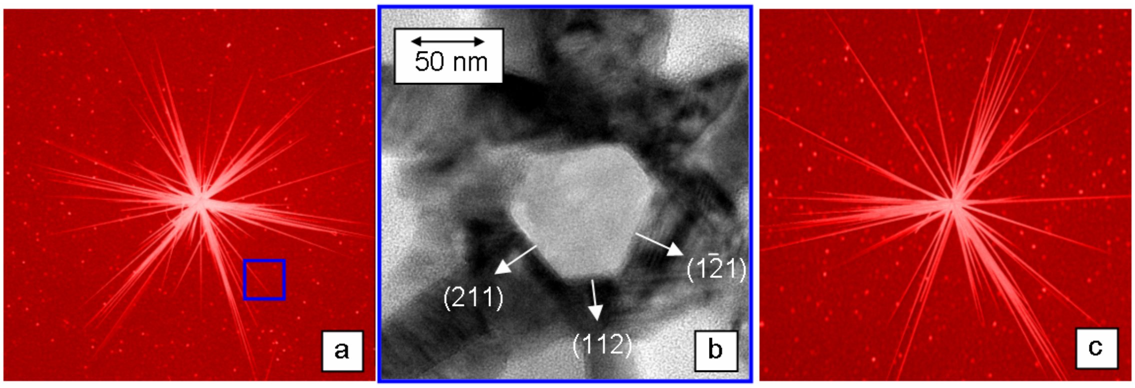

Fig. 2: (a) Top view SEM image of GaAs branches grown perpendicular to the free-standing Si-NW showing a 6-fold symmetry. (b) TEM image showing the cross-sectional hexagon of the Si-NW trunk with alternating wider and narrower vertical {112} facets normal to the [111] growth direction. (c) GaAs nanowhisker which grew preferably on the long facets of the Si-NW trunk with a quasi 3-fold symmetry.

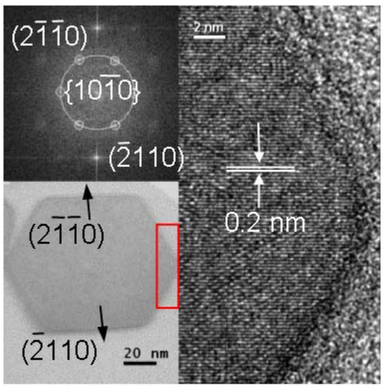

Fig. 3: Cross-sectional TEM and HRTEM image of a GaAs whisker and the respective diffraction pattern recorded along the nanowhisker axis.

The morphology and highly crystalline nature of the GaAs nanowhiskers were characterized by HRTEM and selected area electron diffraction (SAED) measurements (Fig. 3(a)). The TEM image shows the hexagonal cross-section of the GaAs whisker. The upper right SAED pattern was recorded along the nanowhisker axis and has been indexed as the diffraction along the [0001] zone axis of crystalline GaAs and the facets as {2110} atomic planes. The HRTEM micrograph shows a closer view on the area marked with the red rectangle in the TEM image with the (2110) atomic planes of the GaAs whiskers separated by 0.2 nm, consistent with the tabulated value (0.199 nm).

Figure 4 shows the SEM image of GaAs whiskers on Si-NWs for a short MBE growth time. Nucleation starts at the upper part of the Si-NW trunks and the first nanowhiskers grew perfectly aligned according to the Si-NW {112} facets. For longer growth times there is no significant length difference observed for the GaAs whisker as a function of their position along the Si-NW trunks, but GaAs whiskers deviating from the 6-fold symmetry appear to be shorter (see Fig. 2 (a)). These GaAs whiskers nucleate later on pre-existing GaAs precipitations formed on the Si-NW trunks ((see Fig. 2 (b)) and thereby lose the crystallographic relation to the core Si-NWs. The thickness of the unintentionally deposited polycrystalline GaAs shell on the Si-NW trunk increases with MBE growth time according to a 2-D equivalent GaAs growth rate of 0.55 µm/h.

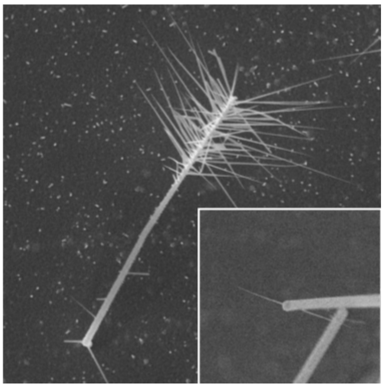

Fig. 4: GaAs nanowhiskers grown on a freestanding Si-NW trunk; MBE growth at 723 K for 524 s. The inset shows two GaAs whisker grown on opposite facets of a Si-NW; MBE growth at 723 K for 262 s.

Conclusion

Taken together, we showed hetero-epitaxial growth of branched III-V nanowhiskers on epitaxially grown group IV NWs realized by combined VLS and MBE methods. We demonstrate this for GaAs whiskers, a direct bandgap semiconductor with high electron mobility predestined for nanophotonics, grown on Si (111) nanowires. The hetero-epitaxial growth and the good crystallinity of the Si-NWs and GaAs whiskers were confirmed by HRTEM.

This work is far from complete in the aspects of synthesizing and the growth mechanism; however, we feel that the nanostructures reported here will have a high potential to be combined with Si for high-speed electronic and photonic applications.

References

[1] Alois Lugstein, Aaron Maxwell Andrews, Mathias Steinmair, Youn-Joo Hyun, Emmerich Bertagnolli, Matthias Weil, Peter Pongratz, Matthias Schramböck, Tomas Roch, Gottfried Strasser, "Growth of branched single crystalline GaAs whiskers on Si nanowire trunks", Nanotechnology 18, 355306 (2007).