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PIER PhD Project 11

Structural analysis of single droplet-etched epitaxial nanostructures – a combined surface X-ray diffraction and real space imaging approach

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Semiconductor quantum dots (QD) are highly promising building-blocks for advanced applications like sources of single indistinguishable photons for quantum cryptography or qubits for quantum computing. We focus in this project on novel droplet-etching based epitaxial QDs fabricated in a self-assembled fashion by molecular beam epitaxy (MBE). Local droplet etching (LDE) represents a novel approach for in-situ top-down nanostructuring without the need of lithographic steps and, thus, fundamentally extends the MBE fabrication of advanced semiconductor nanostructures. Figures 1a-d show examples of low-density nanoholes after Al droplet etching in AlGaAs.

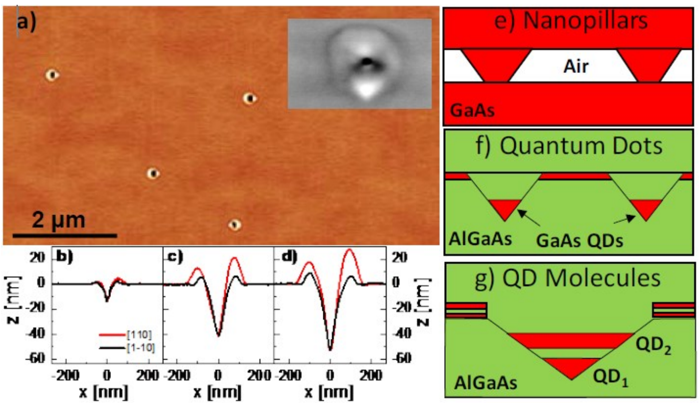

Fig.1 a) Top-view AFM image of low density droplet etched nanoholes in AlGaAs. The inset shows a high resolution SEM image of a selected nanohole. b)-d) AFM linescans of nanoholes with size tuned by process parameters. e) Schematic of GaAs nanopillars in air-gap heterostructures. f) Schematic of GaAs QDs in nanoholes. g) Schematic of a vertically stacked quantum dot molecule fabricated self-aligned in a nanohole.

With this proposal we address the need for one-to-one structure and property characterization of single droplet etched nanoholes at the atomic scale. High resolution X-ray measurements on these nanoholes will be performed in order to clarify the mechanisms of their formation. We furthermore aim to use the nanoholes as templates to fabricate, e.g., GaAs nanopillars (Fig. 1e) or QDs (Fig. 1f,g), to gain insight into the relation between the QD electro-optical properties and their morphology and material composition.

The project is promoted by two research groups at University of Hamburg and DESY. Sample fabrication with MBE, characterization with atomic force microscopy (AFM), and optical experiments using single-dot photoluminescence (PL) will be performed in the research group of W. Hansen at the Institut für Nanostruktur- und Festkörperphysik (INF). Single nanoholes are selected, marked for re-localization and are excavated or area-selectively thinned using focused-ion-beam (FIB) and structurally and chemically characterized by high resolution X-ray diffraction and SEM-based scanning transmission electron microscopy (STEM) in the research group of A. Stierle at the DESY NanoLab.

For local droplet etching, Al, Ga, or In droplets can be used during MBE for drilling nanoholes into GaAs, AlAs, or AlGaAs surfaces [1]. Variations of the process conditions provide a control on the nanohole structural properties over a wide range. E.g., the hole density is varied from $4x10^6$ cm^2 up to $2x10^9$ cm^2 and the depth from 1 nm up to 125 nm (Figs. 1b-d). However, the dynamics of the droplet etching process as, e.g., the influence of the process parameters like the Arsenic background pressure is so far largely unclear. We aim to gain insight from determining the structure and material composition of the rings around the hole opening, which is assumed to originate partly from inside the nanoholes and re-deposited during droplet etching. AFM and SEM imaging show that the top surface inside a nanohole is faceted, permitting to analyze their atomic structure using surface X-ray diffraction and measuring characteristic facet signals appearing as streaks around the substrate Bragg peaks. Identification of these streaks should give first insights into the order and atomic composition of the uppermost surface layer. Ensembles of unfilled nanoholes will thereby act as a reference system for the high resolution XRD investigations and will be compared with AFM and SEM characterization. We will furthermore follow the time-dependence of the etching process by monitoring the etching and faceting of nanoholes by in-situ surface X-ray diffraction using a portable vacuum chamber.

To localize single, pre-selected nanoholes for surface X-ray diffraction in a nanofocused X-ray beam we will use a “Advanced Nanoobject Transfer and Positioning” protocol to be developed at DESY NanoLab within the EU Horizon 2020-funded program “Nanoscience Foundries and Fine Analysis (NFFA)”. This µm a) Air AlGaAs GaAs QDs e) Nanopillars g) QD Molecules QD2 AlGaAs QD1 GaAs f) Quantum Dots protocol comprises i) the SEM analysis of the nanoholes at DESY NanoLab, ii) the pre-selection and marking of the nanoholes using electron-beam assisted deposition of metal-organic precursors as marker material, iii) defining a coordinate system with marker and nanohole positions, iv) transfer to a dedicated nanofocused Xray beamline at a third-generation synchrotron source like PETRA III or ESRF, and v) re-localization of the nanoholes utilizing the markers and the coordinate system based on their X-ray fluorescence signal.

In order to functionalize a template of droplet etched nanoholes, they are epitaxially filled with materials different from the substrate. A first example are ultra-short GaAs nanopillars (Fig. 1e) separating two epitaxial layers in a fully crystalline so-called air gap heterostructure (AGH). Nanopillars in AGHs act as ballistic quantum-point contacts for phonons and such structures are highly interesting for thermoelectric applications due to their low thermal conductivity [2]. The structural properties of such nanopillars are not accessible with AFM und will be investigated with XRD. Interesting points are, e.g, the size, shape and composition of the pillars as well as a possible bending and the strain-status of the suspended top-layer.

Partial filling of nanoholes in AlGaAs with GaAs yields strain-free GaAs quantum dots (QDs, Fig. 1f). The droplet-etched GaAs QDs demonstrate a tunable optical emission wavelength from 700 to 800 nm [3], narrow exciton lines with widths down to 25 µeV, and low exciton fine-structure splittings down to 4 µeV. Autocorrelation experiments demonstrate clear single-photon emission. In addition, vertically coupled quantum dot molecules (QDM, Fig. 1g) are fabricated by filling of nanoholes with two quantum dots separated by a thin tunnel barrier. In optical experiments, the QDMs show clear signatures of moleculeresonant states. Within this proposal we aim to investigate these QD-based structures with single-dot photoluminescence (PL). However, so far the interpretation of the optical experiments on the QD-based structures lacks the detailed knowledge of the structure. A more dedicated structural analysis is important to interpret single QD optical properties like linewidth and lifetime. For QDMs, the tunneling of charge carriers through the barrier separating both QDs crucially depends on the local barrier thickness and composition. The methods developed in the project for addressing single nanoholes will be applied also for measurements of single droplet-etched GaAs QDs. The QDs are embedded in AlGaAs and buried 10 nm – 100 nm below the surface. Since the position of the buried dots is visible by small hillocks on the surface, we can prepare markers also to address single buried QDs or QDMs. We will prepare cross-section samples by the FIB technique for structural analysis with high resolution scanning electron microscopy using the STEM detector at DESY NanoLab that provides excellent contrast and high spatial resolution. Additional measurements using atom probe tomography (APT) are planned in collaboration with Paul Koenraad in Eindhoven, Netherlands. Here, the definition of markers is essential for the selection of single nanoobjects. APT provides in real space with subten nanometer resolution insight into the chemical composition of single QDs and QDMs.

Working plan for the PhD student:

The PhD student will be directly supervised by Dr. Thomas Keller in the Stierle research group and Dr. Christian Heyn in the Hansen Group. The PhD student will create surface nanoholes and quantum dots by MBE growth and hole filling. In a next step, single nanoholes are pre-selected, marked using Pt-based markers and prepared for the structural and chemical characterization using high-resolution XRD at nanofocused X-ray beamlines. These demanding XRD measurements are feasible at third generation synchrotron sources. XRD measurements of single QDs after selective-area FIB thinning will be compared with STEM and APT investigations. Finally, the insight into the nanostructure morphology will be used to understand the optical properties of the QDs determined in the Hansen group and fed back to optimize the process of nanohole creation and to design advanced nanohole / QD systems.

References

[1] Ch. Heyn, Th. Bartsch, S. Sanguinetti, D. E. Jesson, and W. Hansen, Nanoscale Res. Lett., 10, (2015) 67.
[2] Th. Bartsch, M. Schmidt, Ch. Heyn, and W. Hansen, Phys. Rev. Lett. 108, (2012) 075901.
[3] Ch. Heyn, A. Stemmann, T. Köppen, Ch. Strelow, T. Kipp, S. Mendach, and W. Hansen, Appl. Phys. Lett. 94, (2009) 183113.
[4] B. Arndt, H. Noei, T. F. Keller, P. Muller, V. Vonk, A. Nenning A. K. Opitz, J. Fleig, U. Rutt, A. Stierle, Thin Solid Films 603, (2016) 56.

Specific requirements/experiences of PhD candidates:

Excellent knowledge of fundamentals in solid-state physics, experience in at least one of the following fields: semiconductor physics, semiconductor epitaxy, experimental x-ray methods