Computational design of core-shell nanowire crystal-phase quantum rings for the observation of Aharonov-Bohm oscillations

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***Abstract*—We present a systematic design process of quantum rings formed by combining radial compositional and axial crystal-phase heterostructures in GaAs/AlAs core-shell nanowires. We focus on the modelling and design aspect and finally demonstrate that core-shell GaAs/AlAs nanowires containing atomically flat polytype segments can be systematically tailored for studies of the Aharonov-Bohm effect of neutral and charged excitons.**

I. INTRODUCTION

Semiconductor nanowires exhibit unique properties which potentially enable us to design nanostructures that cannot be obtained in planar epitaxial growth. In particular, the free side facets facilitate elastic relaxation such that compositional heterostructures along radial and axial directions can be produced with much larger lattice mismatch than in planar systems. Moreover, nanowires produced from III-V materials exhibit polytypism, i.e. the coexistence of different crystal phases of the same chemical composition within the same nanowire. As distinct crystal phases from the same material have different electronic properties, they can be employed to design crystal-phase heterostructures with atomically flat interfaces and only minor lattice mismatch, that are free of alloy fluctuations [1]. Semiconductor nanowires therefore provide the opportunity to combine compositional and crystal-phase geometries to design a new class of quantum-physical nanostructures. In the following, we introduce a combination of a radial compositional and an axial crystal-phase heterostructure to obtain a new design of high-quality quantum rings with experimentally accessible specifications for studies of the optical Aharonov-Bohm effect. This fundamental quantum mechanical phenomenon results from the phase shift acquired by an electronic excitation in a ring threaded by a magnetic field, and is in essence a property of charged particles. However, it has also been observed for neutral excitons in quantum rings if electrons and holes are bound to rings of different radii where it manifests itself in

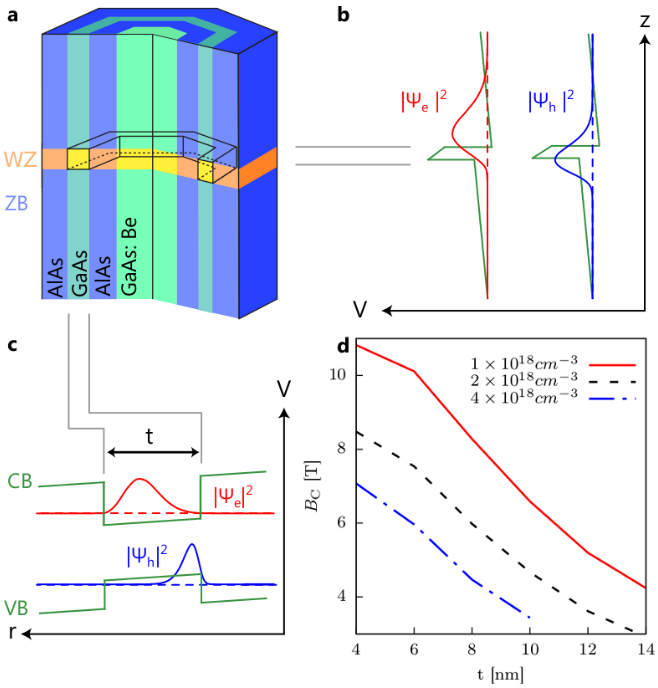

Fig. 1. a: Schematic view of a GaAs/AlAs crystal-phase quantum ring. The zincblende GaAs core (green) of diameter of 30 nm is doped with Be. The GaAs shell quantum well (turquoise) of thickness t is clad by AlAs shells with thicknesses of 15 nm. A wurtzite (WZ) segment propagates through the entire diameter of the nanowire (orange and yellow) b, c: Schematic representation of the conduction and valence band edges in axial (b) and radial (c) direction with the respective electron (red) and hole (blue) ground state charge densities. d: Critical magnetic field to achieve a 2π phase shift as a function of GaAs shell thickness for different acceptor densities.

oscillations of emission wavelength and intensity [2].

II. THEORY-GUIDED DESIGN

The excitonic Aharonov-Bohm effect has been studied using droplet-epitaxially grown volcano-shaped quantum rings in the past. However, these systems suffer from imperfections that induce decoherency and the characteristic design parameters of such quantum rings are hard to control. A good experimental setup for the observation of the excitonic Aharonov-Bohm effect has two fundamental – contradicting – requirements. On the one hand, the critical magnetic field B_c required to obtain a 2π phase shift of the exciton is given as

$$B_c = \Phi_0 / \left(\pi |\langle r_\mathrm{e}^2\rangle - \langle r_\mathrm{h}^2\rangle|\right) \tag{1}$$

with r_e^2 and r_h being the mean squared electron and hole ground state radii. It is obvious that experimentally accessible critical magnetic fields require a sufficient spatial electron-hole separation. On the other hand, a large separation between electron and hole will impede excitonic recombination. A design process of suitable structures thus requires an electron-hole separation large enough to warrant reasonable magnetic fields while still allowing the radiative recombination of the indirect exciton.

Our suggested structure is a core-shell GaAs/AlAs nanowire, dominated by the zincblende phase, incorporating a small number of thin wurtzite segments, e.g. twin boundaries (cf. Fig. 1 a). The active layer is a GaAs shell of thickness t. The wurtzite segments ensure axial confinement of electrons and holes (Fig. 1 b). A Be-doped GaAs core induces a radial potential and thus a radial electron-hole separation due to surface Fermi level pinning (Fig. 1 c). The spatial separation between electron and hole can now be controlled by two parameters, namely the GaAs shell thickness, t, and the core acceptor density $\mathrm{N}_{\mathrm{A}^-}$.The design process to identify promising structures for systematic studies of the optical Aharonov-Bohm effect was based on numerical simulations considering realistic structures that can be obtained by molecular beam epitaxy. The material system GaAs/AlAs was selected for the binary compositional heterostructure due to its small lattice mismatch and effective electron masses small enough to maintain excitonic recombination despite a spatial separation between electron and hole. p-type doping of the core can be realized in GaAs using Be and the core diameter can be reduced to below 30 nm by partial thermal decomposition [3,4]. The thickness of the AlAs barriers was chosen such that it prevents tunneling from the active shell into the core and finally oxidation of AlAs was prevented by an outer GaAs shell. Promising candidates for the observation of the optical Aharonov-Bohm effect were identified by systematic variation of the GaAs shell thickness and the core doping density. The employed formalisms for this purpose were an eight-band $\mathbf{k}\cdot\mathbf{p}$ model in combination with linear elasticity theory. Both formalisms were implemented within a plane-wave framework [5,6]. With core doping densities of 2 to 4 $\times 10^{18}$ cm^{-3} and shell thicknesses between 5 and 10 nm, critical magnetic fields below 8 T can be achieved (cf. Fig. 1 d).

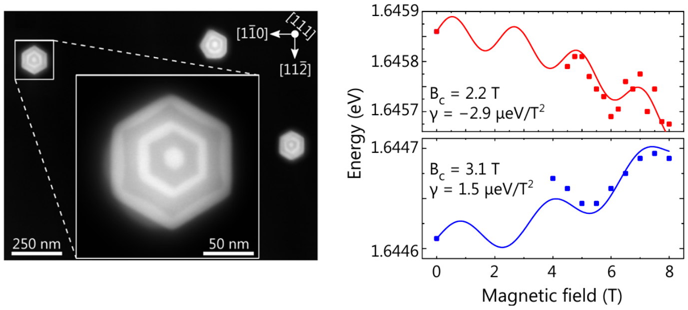

Fig. 2. Left: plan-view transmission electron micrograph of the GaAs/AlAs core-multishell nanowires. Right: Dependence of the average energy of the Zeeman split states on magnetic field for two different rings.

III. EXPERIMENTAL VERIFICATION

Guided by these results and considerations, GaAs/AlAs core-multishell nanowires were fabricated by Ga-assisted vapor-liquid solid growth in molecular beam epitaxy. Fig. 2 (left) shows a plan-view transmission electron micrograph confirming core and shell dimensions close to the designated values. Raman spectroscopy indicates a core Be concentration of 3 to 6 $\times 10^{18}$ cm^{-3}.

Photoluminescence spectra exhibit a broad band originating from recombination in the GaAs shell, superimposed by two narrow lines that we attribute to emission from twin boundaries in the GaAs shell, i.e. quantum rings. The respective transitions split with increasing magnetic field as a result of Zeeman and diamagnetic effects. However, the evolution of the transition energies with increasing magnetic field is not parabolic but exhibits an oscillatory behavior where the split states oscillate in phase. This oscillatory magnetic-field dependence is shown in Fig. 2 (left) for two different quantum rings. We attribute these oscillations to the optical Aharonov-Bohm effect and extracted critical magnetic fields of 2.2 and 3.1 T for the two nanowires under consideration.

In summary, we have introduced core-shell crystal-phase quantum rings for systematic studies of the optical Aharonov-Bohm effect which allow for a high degree of control of structural and physical parameters.

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