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Fabrication and Optical Properties of Strain-free Self-assembled Mesoscopic GaAs Structures

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Abstract

We use a combined process of Ga-assisted deoxidation and local droplet etching to fabricate unstrained mesoscopic GaAs/AlGaAs structures exhibiting a high shape anisotropy with a length up to 1.2 μm and a width of 150 nm. We demonstrate good controllability over size and morphology of the mesoscopic structures by tuning the growth parameters. Our growth method yields structures, which are coupled to a surrounding quantum well and present unique optical emission features. Microscopic and optical analysis of single structures allows us to demonstrate that single structure emission originates from two different confinement regions, which are spectrally separated and show sharp excitonic lines. Photoluminescence is detected up to room temperature making the structures the ideal candidates for strain-free light emitting/detecting devices.

Keywords: Strain-free quantum dots, Local hole etching, Ga-assisted deoxidation, Molecular beam epitaxy

Background

Semiconductor quantum dots have been the subject to intensive research in the last three decades and one of the model systems for nanotechnology. One of the main mechanisms to fabricate these nanometer-sized structures is the Stranski–Krastanov growth mode of epitaxial lattice mismatch materials [1–3]. This method has the disadvantage that the obtained quantum dots are inherently strained, which modifies optical and electrical properties of the material [4]. In the last decade, several strategies have been developed to overcome this restriction and to obtain strain-free quantum dot structures [5–15]. One common strategy is the use of thickness fluctuations of an AlGaAs/GaAs/AlGaAs quantum well (QW) [13–15]. A second tactic is to pattern the substrates with holes and then grow a light-emitting structure of AlGaAs/GaAs/AlGaAs on top resulting in a quantum dot at the hole position in the initial template. Strategies for hole fabrication have included the in situ etching of holes [16, 17] and their overgrowth [7], Ga-assisted hole etching and filling [9, 11, 12, 18–23], or overgrowth of patterned substrates [8]. These strain-free structures constitute an own class of nano-sized optical emitters and have been used successfully in a variety of fascinating experiments and applied device structures such as studying of the exciton–exciton and exciton–phonon interaction in quantum dot ensembles [24], coherent optical controlled quantum states [25], single-photon sources [26–28], entangled photon sources [29], and tuning the optical emission by local straining [30, 31] or the optical control of the nuclear spin for potential quantum-computing applications [32–34].

Common current approaches share that the produced GaAs dots are rather shallow and it is hard to realize a good confinement. For dots defined by thickness fluctuations, this problem is intrinsic to the fabrication and the use of two-dimensional islands of the epitaxial growth [13]. When defining the hole template by etching, these holes tend to be shallow preventing the growth of thick lower AlGaAs barriers [7]. Alternatively, the bottom barrier is grown first and then structured by in situ etching, introducing a growth interruption exposing an AlGaAs surface [9, 12, 23]. Extended thick GaAs structures are commonly not found, even though they would offer an

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additional tuning parameter, broadening the optical and physical properties.

In this work, we fabricate a room temperature active elongated mesoscopic GaAs structure (MGS) with a high shape aspect ratio. The MGS have been made by molecular beam epitaxy combining Ga-assisted deoxidation [35, 36] with local hole etching [10, 18, 19, 22] in a machine equipped with a thermal As source. Our morphological study by atomic force microscopy (AFM) finds an average hole depth of 19 nm and ca. 200 nm hole diameter. The holes exhibit crystal facets as side walls making them hexagonally shaped. They are surrounded by GaAs mounds. By overgrowing these holes with different amounts of $Al_{0.33}Ga_{0.67}As$, the shape and size of the initial template can be tuned. We demonstrate that layers as thick as 20 nm can be grown without significantly decreasing the hole depth and sizes, reproducing the elongated mound surrounding the hole. We fill this hole-mound structure with different amounts of GaAs and then cap it with another upper 20-nm $Al_{0.33}Ga_{0.67}As$ barrier. Studies by transmission electron microscopy (TEM) of focused ion beam (FIB) prepared cross-sections together with AFM results allow us to develop a complete structural image of the MGS. Microphotoluminescence (μ-PL) investigations reveal room temperature (RT) optical emissions of the MGS. At low temperature, the PL spectrum is composed of closely spaced single excitonic lines in an energy range of 1.58 to 1.72 eV and the contribution of the 15-nm-deep hole-mound structures at ca. 1.55 eV. Temperature-dependent μ-PL investigation indicates an electronic coupling of these energetically closely spaced lines creating a unique mesoscopic system compromising a single dot formed by the deep hole coupled to thickness fluctuation dots formed by the elongated mound structure. Hence, the mesoscopic GaAs structures form a quantum dot system with high potential for application in optical devices.

Methods

All the samples were grown in a 2-inch custom-made solid-state molecular beam epitaxy machine (Dr. Eberl MBE-Komponenten GmbH) at LNNano (CNPEM, Brazil). Growth is monitored in situ by reflective high-energy electron diffraction (RHEED). The chamber uses a thermal As_4 cell opened and closed by a linear shutter that reduces the beam equivalent pressure (BEP) by ca. 100 times (1.2×10^{-5} to 2.7×10^{-7} mbar). This kind of cell needs, compared to valve cracker cells, some time to reach and stabilize the As_4 flux for a given temperature—for our machine ca. 15 min for typical temperature of 314 °C. During this stabilization time, the As flux continually increases non-linearly and finally saturates at its maximum value.

GaAs (001) wafer pieces with a previously grown GaAs buffer were In-glued on the 2″ Si wafer and introduced into the MBE and preheated at 300 °C. Hole templates were fabricated by a combination of Ga-assisted deoxidation and local Ga hole etching [10, 18, 22, 35, 36]. In the absence of an As counter pressure, the sample was exposed repeatedly 20 times to a Ga flux of one monolayer (ML) per minute for 30 s followed by a 30-s growth interruption at a substrate temperature of 450 °C. A streaky RHEED showed up after six to seven ML indicating a complete removal of the native oxide layer. During the Ga-assisted deoxidation process, we ramped the As cell to 290 °C corresponding to an increase of the low background pressure from 2×10^{-9} mbar to 1×10^{-8} mbar. After finishing the initial Ga deposition, the As cell temperature is raised to 314 °C and the gallium rate is increased to ~0.3 μm/h. The sample is annealed for 5 min during the time that the As cell needs to reach 314 °C with the shutter closed.

With the As cell shutter closed, we deposited Ga of an 5 nm equivalent GaAs thickness during 1 min and opened afterwards the As shutter. A further annealing of 15 min was carried out to stabilize the As counter pressure—the chamber background pressure rised from 2×10^{-8} mbar to 1.5×10^{-6} mbar (corresponding to a BEP of 2×10^{-5} mbar stabilizing a rate of ca. 0.4 ML/s for our machine). This process resulted in the formation of a hole surrounded by a long mound structure on an otherwise flat GaAs (001) surface.

On a typical hole-mound structure of the GaAs surface described above, we deposited $Al_{0.33}Ga_{0.67}As$ layers with different thickness to study the hole filling at the same substrate temperature (450 °C) used for the hole creation. In the second set of samples, we chose a particular template containing hole-mound structures with a 20-nm AlGaAs layer and deposited different GaAs layer thicknesses by pulsed epitaxy [12, 22] with a growth rate of 0.3 μm/h. The GaAs was deposited at 450 °C substrate temperature opening the shutter for 12 s (1 nm) and waiting for 36 s. The number of repetitions was tuned according to the nominal GaAs thickness for filling. Samples were annealed for 10 min ramping the substrate to 570 °C after GaAs deposition. In the third set ones, for photoluminescence studies, a top barrier of 20-nm $Al_{0.33}Ga_{0.67}As$ layer followed by a 5-nm GaAs cap were grown at 570 °C.

After every growth step, reference samples were taken out of the chamber and the surface studied by AFM. We used a NX10 instrument (Park System) in tapping mode with standard AFM cantilevers. Images were post-processed and statistical analysis carried out with the SPM software Gwyddion. For the holes, structures were measured using the line scan tool of the software, whereas the mesoscopic GaAs structures were analyzed using the grain finding function with the statistical analysis module.

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