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TEM-study of free-standing self-assembled InSb quantum dots grown on InAs surface

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1. Introduction

Semiconductor quantum dot (QD) based nanostructures have attracted a considerable attention in basic and applied physics due to their unique electronic properties, which enable a strong improvement in optoelectronic device performance. Quantum dots as zero-dimensional structure are promising for investigations due to their δ -function-like density of states, strong carrier localization, increased exciton binding energies, and enhanced oscillator strength. The three-dimensional (3D) confined structure induces quantum level spacing which is less sensitive to phonon decay and therefore may give rise to higher operating temperatures devices [1]. So far, the great part of studies have been devoted to the InAs QDs buried in GaAs or InP matrices that are capable of operating in the spectral range 1.3-1.55 µm important for the telecommunication applications [2,3]. It is evident that the photon energy of the active region of an optoelectronics device based on lowdimensional heterostructures such as quantum dots or quantum wells is limited by the energy gap of a matrix layer. To expand the spectral range covered by QD-based structures to the mid-infrared $(\lambda > 2 \mu m)$ region quantum dots obtained from a narrow-gap semiconductor such as InSb are required to produce in GaSb or InAs

ABSTRACT

We have investigated the morphology of free-standing self-assembled InSb quantum dots (QDs) grown by liquid-phase epitaxy (LPE) under conventional growth conditions on InAs(001) substrate. Two growth modes, Volmer–Weber for low-density ($5 \times 10^8 \text{ cm}^{-2}$) large QDs with 10–12 nm in a height and Stranski–Krastanow for high-density ($1 \times 10^{10} \text{ cm}^{-2}$) small QDs with a height of 3–4 nm, were identified in dependence on a critical growth temperature of InSb QDs formation. Characterization of the sample surface was performed using transmission electron microscopy (TEM) that allowed us to observe some features of the InSb QD shape which transforms from a full dome to truncated one with increasing QD lateral size from 15 to 40 nm, respectively. Using pseudo-moiré pattern appearing in plan-view diffraction TEM image the critical size of the InSb QD for plastic deformation was evaluated.

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matrices [4,5]. The InAs substrate allows the application of narrower compounds with the energy gap in the range of 0.35-0.65 eV. InSb/InAs QD heterostructures are most interesting as a potential platform for optoelectronic devices operating in the mid-infrared wavelength region $3-5 \,\mu$ m region where pronounced absorption lines of different gases are situated that enables detection of various chemical, explosive, and biological agents [6].

Various growth methods, including molecular beam epitaxy (MBE), liquid-phase epitaxy (LPE) or metalorganic vapor-phase epitaxy (MOVPE), were employed to obtain InSb QDs on InAs substrates. Arrays of InSb QDs with a height of 10 nm and diameter of 80 nm were formed with a surface density of $6 \times 10^9 \text{ cm}^{-2}$ using a special MBE technique combining atomic layer epitaxy (ALE) at T=390 °C or migration enhanced epitaxy (MEE) at T=410 °C, whereas for the conventional MBE method the density did not exceed 2×10^9 cm⁻² [7,8]. For modified approach of MOVPE, the droplet heteroepitaxy (DHE), under conditions of In droplets formation and TMSb supplying at 350 °C, the density of the InSb QDs was found to be 10^{11} cm⁻² and with almost one size distribution of about 20 nm [9]. In turn, the LPE method is an inexpensive and simple method which allows getting InSb/InAs QDs density starting with 10¹⁰ cm⁻² [10,11]. It was recently established that the substrate surface chemistry significantly effects on the dot size and shape and their density [12].

InSb QDs in the InAs matrix should possess unique electronic and optical properties due to type II energy band alignment. The

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type II band lineup heterojunction accommodates spatially indirect transitions in contrary to a straddling type I band lineup (InAs/GaAs, InAs/InP, etc.) with spatially direct transitions. Moreover, type II recombination transitions can reduce the effective gap of these transitions [13]. Formation of type II quantum dots results in an appearance of a luminescence line which persists up to high observation temperatures and excitation densities [14]. This luminescence originates from radiative recombination of 0D holes and spatially separated electrons attracted to them via Coulomb interaction. Most of the holes are captured and subsequently a dipole layer can be formed between the holes in the InSb dots and the electrons attracted from the surrounding InAs regions. This fieldinduced band-bending will confine the electron wave function closer to the InSb dots.

Despite the large lattice mismatch (7%), similar to the wellknown InAs/GaAs system [15], the formation and properties of InSb QDs on InAs matrix are studied to much lesser extent. In this work, we aim at obtaining initial data on the morphology of free-standing InSb QDs formed on InAs(001) surface by liquid phase epitaxy.

2. Experimental

The epitaxial growth was carried out in a horizontal LPE set-up equipped with a conventional graphite slider boat under purified H_2 flow on InAs(001) substrate in the wide temperature range T = 425-445 °C. The substrates were prepared for the deposition by the wet chemical polishing. The deposition of InSb was performed from In melt in which binary InSb compound had been fused as a source of antinomy. The formation of the InSb QDs was accomplished during a steady-speed sliding of the InAs substrate under the melt at the given temperature. Details of the QD growth were reported elsewhere [11,16].

The surface relief of the samples under study was examined at room temperature conditions by atomic-force microscopy (AFM) with Solver P47H microscope and Integra-Aura scanning probe complex manufactured by NTMDT (Russia). Measurements were performed in a tapping mode using NSG-01 silicon cantilevers with a resonance frequency of f = 150 kHz. Average impact force did not exceed F = 0.5 nN to avoid destruction of the sample surface [17]. Transmission electron microscopy (TEM) study was performed using a JEOL JEM2100FEG microscope operating at an accelerating voltage of 200 kV. Both diffraction contrast and high-resolution imaging (HREM) modes were exploited. TEM specimens were prepared in conventional manner by grinding, dimpling and final 4 keV Ar⁺ ion milling.

3. Results and discussion

Varying the growth temperature in the range from 425 °C to 440 °C, two types of the InSb QDs, big and small, were found to be present simultaneously on the binary InAs surface (Fig. 1a). The small InSb quantum dots had the height 2–5 nm, the lateral size 20–40 nm and exhibited a high surface density of around 1×10^{10} cm⁻² in the order of magnitude, while the big QDs of which the height was 12–14 nm and the lateral size 50–60 nm appeared to be at a much lower density of approximately 3×10^8 cm⁻². Increase in the growth temperature led to reduction in density of the small QDs up to their complete vanishing at the critical temperature T=445 °C whereas the big ones remained on the matrix surface and increased their density up to 10^9 cm⁻² (see Fig. 1b).

It should be noted that the bimodal size distribution appears for the InSb quantum dots grown by different growth techniques such as metal-organic vapor phase epitaxy [9] and it can be considered as an evidence of competition between two growth modes (Stranski–Krastanow and Volmer–Weber) [18].



Fig. 1. AFM topography images of 1 μ m × 1 μ m square surface of the samples with InSb QDs grown on InAs(001) substrate at (a) *T* = 430 °C (QD density 1 × 10¹⁰ cm⁻²) and (b) *T* = 445 °C (QD density 1 × 10⁹ cm⁻²).

Transmission electron microscopy was used to investigate the InSb QD morphology in more details. Fig. 2 exemplifies plan-view TEM images of two individual small dots showing moiré fringes running along [1–10] direction. The fringes are known to appear in the image of a free-standing QD due to difference in interplanar distances in the dot and in the substrate. For a coherent free-standing QD this difference results from Poisson effect.

While the coherent InSb QD is compressed at its base to latticematch with the InAs substrate, the strain gradually relives to the dot top. The behavior of the strain inside the free-standing InSb QD was previously calculated using finite element method and reported elsewhere [19]. It was shown there is very weak influence of the dot shape on the displacements at the center of the quantum



Fig. 2. 220 bright-field plain-view TEM images of two individual small InSb quantum dots grown at T = 430 °C on InAs(001) surface with moiré fringes running along [1–10] direction.

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