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Three dimensional atom probe imaging of GaAsSb quantum rings

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ABSTRACT

Unambiguous evidence of ring-shaped self-assembled GaSb nanostructures grown by molecular beam epitaxy is presented on the basis of atom-probe tomography reconstructions and dark field transmission electron microscopy imaging. The GaAs capping process causes a strong segregation of Sb out of the center of GaSb quantum dots, leading to the self-assembled $GaAs_xSb_{1-x}$ quantum rings of 20–30 nm in diameter with $x \sim 0.33$.

1. Introduction

Ring-shaped semiconductor nanostructures have generated great interest over the past decade due to their unique properties, such as quantum mechanical Aharonov–Bohm oscillations that are a function of the magnetic flux trapped by carriers confined in the quantum rings [1–3]. This true quantum effect with no classic analog is also present in excitons confined in electronic rings defined by Coulomb interaction in type II nanostructures such as InP/GaAs or ZnTe/ZnSe QDs [4,5]. The main interest for the GaSb–GaAs system lies in the fact that it also achieves type II staggered band alignment [6,7] and is suitable for the observation of Aharonov–Bohm related magnetic behavior with holes confined in nanoscale rings and spatially separated electrons. More generally, GaSb–GaAs nanostructures are very promising for single charge carriers, opto-electronic devices operating in the infra-red range [8], memory devices [9] because of the long hole lifetime inside the nanostructures [10] as well as for photovoltaic solar cells [11,12].

Although high quality GaSb–GaAs heterostructures have been achieved by molecular beam epitaxy (MBE) [13,14], metal organic chemical vapor deposition (MOCVD) [15,16], or organometallic vapor phase epitaxy (OMVPE) [17], there are still difficulties related to the control of GaSb growth. Growth temperature, thickness of the capping layer and growth rate during capping have crucial effects on the resulting structure. In particular, intermixing phenomena, such as segregation or diffusion during the growth of multilayers of GaSb on GaAs(0 0 1), can cause changes in film morphology and composition gradients. Evidence for the self-assembly of quantum rings during capping of GaSb quantum dots has been hypothesized by various authors who used cross-sectional electron microscopy specimens [18–22]. We present unambiguous evidence of ring-shaped self-assembled GaAsSb nanostructures based on atom-probe tomography imaging. The self-assembled formation of such nanostructures depends critically on growth temperature, deposition rate and partial pressures of the III–V elements. Similarly to the case of self-assembled QDs, nano-rings possess atom-like properties that make them promising for new device applications in optics, optoelectronics, quantum cryptography and quantum computing [23]. InAs self-assembled quantum rings (QRs) grown on GaAs [24] have been widely explored. They are formed during growth interruption of partially capped QDs. A drastic redistribution of material occurs from "lens-shape" to "volcano-shape" with an increased lateral size, a reduction of height and a well-defined center hole of around 20 nm [25–28]. This method has also been successfully applied for the fabrication of self-assembled rings in other materials systems, such as SiGe [29].

Accurate atomic scale characterization of size, shape and composition of buried nanostructures is of great interest to understand the effects of growth conditions and to optimize

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device properties. Scanning tunneling microscopy (STM) [30–32], transmission electron microscopy (TEM) [33–34] and scanning transmission electron microscopy (STEM) [35], both in cross-section and plan-view orientations, have been typically used to characterize capped nanostructures. Nevertheless, they mainly provide a two-dimensional (2D) projection of the nanostructures and assumptions have to be made to infer their three-dimensional topology, as shown in Ref. [36]. On the other hand, electron tomography that combines TEM images acquired by tilting the sample by small angular increments can be used to construct a 3D representation [37]. Electron tomography has mainly been applied to biological, geological and functional material samples, in spite of limitations especially for crystalline materials such as diffraction contrast effects, limitation of specimen thickness or poor signal-to-noise ratio [38].

Atom-probe tomography (APT) is a very good alternative to perform 3D atom-by-atom chemical characterization, and, in most cases, it provides the highest available spatial resolution for chemical analysis [39]. The very first analyses of GaAs structures were performed using nanosecond pulse duration lasers but yielded very limited data partially due to unstable lasers and difficulty in preparing specimens [40]. Significant hardware improvements and laser developments over the last decade have contributed to the expansion of this technique for 3D quantitative characterization of an extended range of materials (including semiconductors [41] and oxides [42]). In this work we show the application of APT as a powerful approach to achieve the atom-by-atom chemical and structural analysis of GaAs/Sb quantum structures.

2. Experimental section

The heterostructure has been grown by solid source MBE on GaAs (0 0 1) substrate n-type (Si: 1×10^{18} cm^{-2}). 2 monolayers (ML) of GaSb were first deposited at 480 °C, exposed to Sb flux for 20 s, and finally annealed for 20 s without a Sb flux. A 50 nm GaAs layer was then grown over the GaSb wetting layer in two steps: a 10 nm thick GaAs layer was deposited at the temperature of QD nucleation (480 °C) followed by a 40 nm thick GaAs layer deposited at 570 °C. This procedure was repeated six times and finally, the multilayer was capped with a 300 nm thick GaAs layer (Be: 1×10^{18} cm^{-2}, growth details and photoluminescence spectra of this heterostructure can be found in Ref. [43]).

Cross-section TEM specimens were prepared following standard techniques (mechanical grinding to below 20 μm and polishing using 1 μm diamond suspension on a soft nap pad). Special care during the ion milling process was applied to obtain a uniform specimen thickness over the six layers. This multilayer heterostructure was studied by CTEM using a JEOL-JEM 1200 EX microscope operating at 120 kV. High resolution transmission electron microscopy (HRTEM) was carried out in a JEOL 2011 microscope operating at 200 kV. APT specimens were prepared following the method described in Ref. [44] using a Zeiss NVision 40 dual beam focused ion beam instrument equipped with a Kleindiek micromanipulator to perform the lift-out procedure. APT was performed using a Cameca LEAP-3000XHR microscope. Specimens were held at 15 K and field evaporation was performed in voltage pulsing mode at 150 kHz using a ratio of pulse amplitude to standing voltage equal to 15%. Note that voltage pulsing was preferred over laser pulsing, the latter yielding non-uniform evaporation as described in Ref. [45].

3. Results and discussion

For the structural characterization of the heterostructure, CTEM dark field (DF) $g=002$ images of the six GaAs capped GaSb layers were acquired as shown in Fig. 1. These CTEM observations show two types of nanostructures: (i) pairs of bright regions as shown in Fig. 1b, and (ii) randomly distributed bright regions. The 2D high angle annular dark field scanning transmission electron microscopy (HAADF–STEM) analysis of the randomly distributed features can be found in Ref. [21] where the authors concluded that this contrast originated from very small nanostructures, with heights in the order of 1 nm or below. The nanostructures are formed by a significant Sb segregation process that occurs after GaAs capping of 2 ML of GaSb deposited by MBE.

The pairs of nanostructures were observed by HREM as shown in Fig. 2. The two adjacent lobes seen in the cross-sectional images are consistent with ring shape, which can only be confirmed by three-dimensional imaging. Plan-view TEM images would not allow to determine the real morphology of the nanostructures because of the superposition of the six layers and the thickness of the intermediate layers.

The nanostructure was therefore analyzed by APT and a 3D reconstruction of the regions between the self-assembled nanostructures (usually named wetting layer) is shown in Fig. 3a. A representative 1D concentration profile is shown in Fig. 3b. The

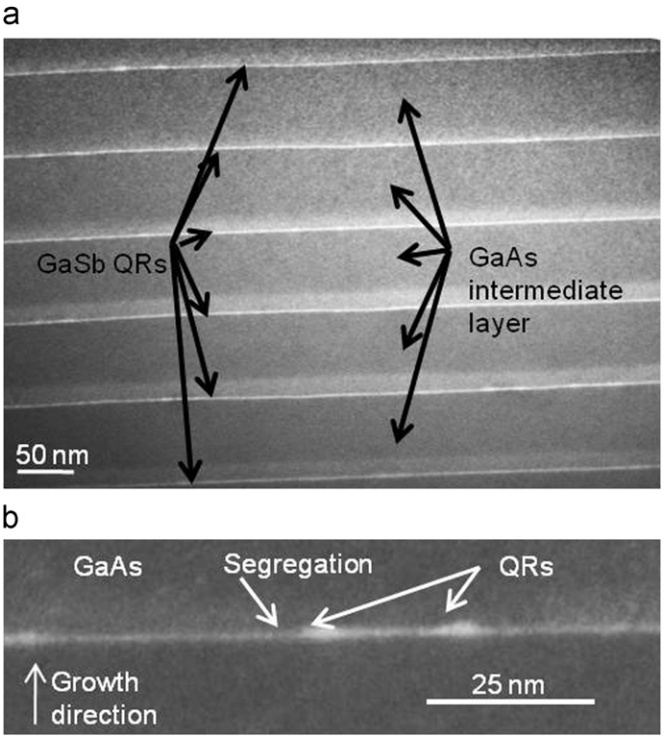

Fig. 1. (a) Dark field 002 cross-section TEM image and (b) enlarged view of one of the layers.

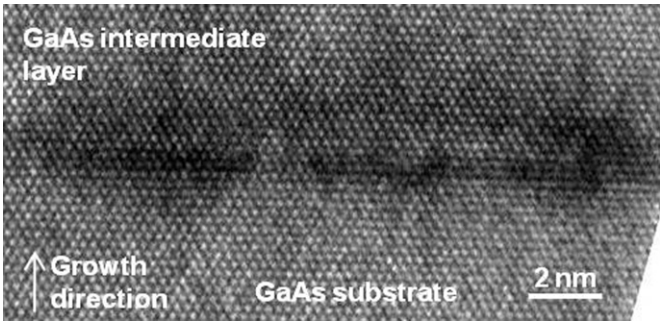

Fig. 2. HRTEM image acquired on [1 1 0] zone axis showing one of the studied nanostructures.