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Full Length Article

Size and shape tunability of self-assembled InAs/GaAs nanostructures through the capping rate

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ABSTRACT

The practical realization of epitaxial quantum dot (QD) nanocrystals led before long to impressive experimental advances in optoelectronic devices, as well as to the emergence of new technological fields. However, the necessary capping process is well-known to hinder a precise control of the QD morphology and therefore of the possible electronic structure required for certain applications. A straightforward approach is shown to tune the structural and optical properties of InAs/GaAs QDs without the need for any capping material different from GaAs or annealing process. The mere adjust of the capping rate allows controlling kinetically the QD dissolution process induced by the surface In-Ga intermixing taking place during overgrowth, determining the final metastable structure. While low capping rates make QDs evolve into more thermodynamically favorable quantum ring structures, increasing capping rates help preserve the QD height and shape, simultaneously improving the luminescence properties. Indeed, a linear relationship between capping rate and QD height is found, resulting in a complete preservation of the original QD geometry for rates above ~2.0 ML s^{-1}. In addition, the inhibition of In diffusion from the QDs top to the areas in between them yields thinner WLs, what could improve the performance of several QD-based optoelectronic devices.

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1. Introduction

Since the experimental realization of semiconductor quantum dots (QDs) and the observation of their unique properties arising from the zero-dimensional confinement, increasing efforts have been made in order to take advantage of such structures for real applications. Epitaxial QDs, and more in particular InAs QDs, have aroused many expectations not only for the development of more efficient optoelectronic devices, but also as a key element in quantum information applications [1–3]. However, the achievement of small fine-structure splitting energies depends on strict geometrical requirements [4] and further exploration of QD growth conditions may help reduce the dependence on external means to produce entangled photon pairs [5,6]. Indeed, geometry and composition determine the properties of QDs [7,8], strongly affecting also the performance of lasers, solar cells or any other optoelectronic devices based on QDs. Most efforts on controlling QD properties have commonly been focused on the growth conditions leading to a good understanding of the QD growth mechanism. The growth rate [9–12] and the growth temperature [13], as well as the time interval between the interruption of the QD growth and the capping process [14] play an essential role in determining the morphology of InAs QDs during their initial formation. Less attention has been paid to the capping process, which is necessary for the implementation of QDs in almost any device application and can significantly affect the final QD properties. Indeed, capping with GaAs entails a heteroepitaxial process which alters the thermodynamic equilibrium structure, leading to a considerable dissolution of QDs and altering their structural properties [15–21]. Fig. 1(a) shows reported heights of buried InAs QDs, which commonly range within 3–4 nm and whose base is typically near 20 nm width [16,22–29]. Remarkably, this is observed under standard capping conditions (~1 ML s^{-1}, 450–530 °C) for very different initial QD heights (QD height before capping), from 4.2 nm up to ~10 nm. Therefore, the overgrowth process seems to impose a generalized evolution

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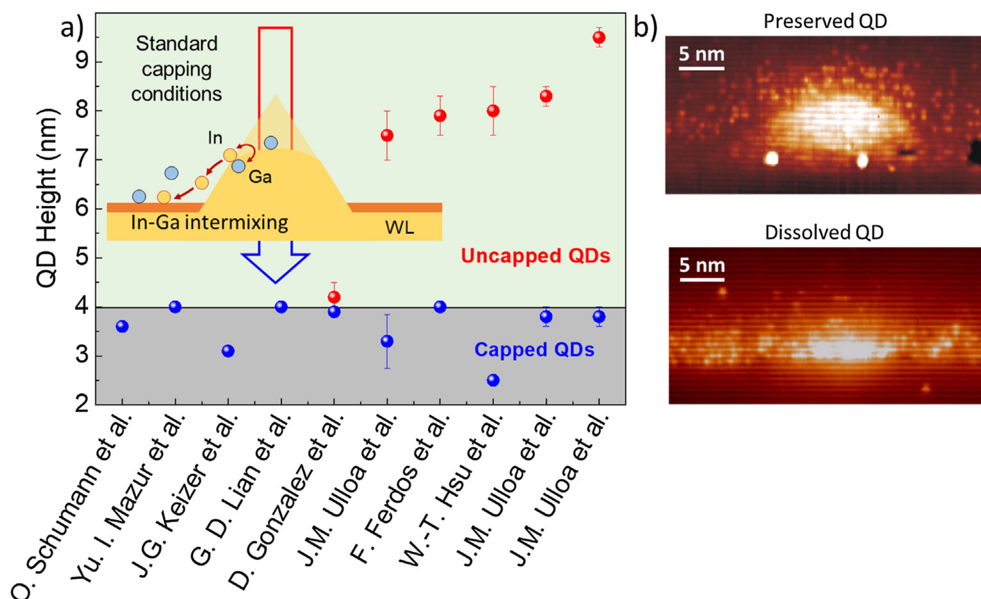

Fig. 1. (a) Average height of InAs/GaAs QDs reported by different authors before (when available) and after the capping process (red and blue dots, respectively). A schematic of the QD dissolution process during capping is also represented in the inset, whereby the QD top flattens through surface In-Ga intermixing and surface diffusion of In leads to the WL thickening (orange layer). (b) The upper image shows a completely preserved QD, achieved by the application of a GaAsSb capping layer (from Ref. [25]). This QD is therefore identical to the uncapped ones. A typical QD capped with GaAs is shown on the lower image (from Ref. [26]), where a flatter QD top and thicker WL are observed. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

towards thermodynamic equilibrium whereby QDs are similarly levelled, regardless of the initial QD size. This means that, however accurate the QD growth conditions are, the QD size and shape will ultimately be affected by the capping process, reducing the QD height to ~3–4 nm and limiting to a high extent the tunability of the optical properties.

A minimization of the additional strain introduced in the QD by the capping layer through In-Ga intermixing is generally believed to be behind this dissolution process. Therefore, most efforts aiming to preserve the QDs have been focused on minimizing strain-driven QD dissolution by using strain-reducing layers made of alternative capping materials [24,29–40]. This means that, unfortunately, even in the event that the strain-field surrounding the QD be sufficiently reduced as to completely preserve the QD, this will occur at the expense of significant and inevitable additional electronic structure modifications. For instance, although the use of GaAsSb capping layers strongly limits the QD dissolution (Fig. 1(b)), a high Sb content yields a transition of the band alignment into a type-II structure [26]. Likewise, a band alignment transition may also be induced by the introduction of N into the capping layer [28]. The application of other strain-reducing layers such as InGaAs may even lead to a QD shape evolution into columnar QDs [41]. Therefore, strain engineering approaches may give rise not only to a complete modification of the QD band structure, but also to strong structural transitions.

Here we propose a very straightforward way by means of which the metastable state of capped QDs can be controllably tuned farther or closer to the thermodynamic equilibrium state without the need for strain engineering approaches. The simple tune of the capping rate allows us to control the size and, therefore, the optical properties of standard GaAs-capped InAs QDs. Besides, the modified intermixing process also affects the final wetting layer (WL) structure, whose impact on the performance of optoelectronic devices is recently gaining an increasing interest [42–45]. The impact of the capping rate on the properties of InAs/GaAs QDs and WLs are both analyzed in this work.

2. Experimental methods

2.1. Growth details

The samples were grown by solid source molecular beam epitaxy on n^+ (100) GaAs substrates. A series of three samples containing a single QD layer was grown for photoluminescence (PL) studies. In all these samples, 2.8 ML s of InAs were deposited at 450 °C and 0.04 ML s^{-1} on an intrinsic GaAs buffer layer. The QD layer was subsequently capped by a 30 ML-thick GaAs layer grown at 0.5, 1.0, and 2.0 ML s^{-1} in samples S_0, S_1, and S_2, respectively, followed by a 250 nm-thick GaAs layer grown at 580 °C and 1.0 ML s^{-1}. An additional sample for transmission electron microscopy (TEM) and cross-sectional scanning tunneling microscopy (X-STM) measurements, labeled as S_3, was grown containing three QD layers equivalent to those of S_0, S_1, and S_2 (L_0, L_1, and L_2 QD layers, respectively, grown in this sequence), separated from each other by a 50 nm-thick GaAs spacer grown at 580 °C and 1 ML s^{-1}. Finally, a 200 nm-thick GaAs layer was deposited, on top of which a fourth layer of uncapped QDs was grown for reference (L_S).

2.2. Optical and structural characterization

The PL was measured at 15 K using a closed-cycle He cryostat. A He-Ne laser with a power of 3 mW was used as the excitation source and the emitted light was dispersed through a 1 m spectrometer, being detected with a liquid N-cooled Ge detector. The chopped signal was detected with a lock-in amplifier in order to enhance the signal to noise ratio. Cross-sectional TEM measurements were conducted using a JEOL-2100 LaB6 microscope operating at 200 kV. Chemical sensitive dark-field TEM images were taken at the [1 1 0] pole axis. Standard tapping mode atomic force microscopy (AFM) measurements with antimony-doped Si tips were also used to characterize surface QDs. X-STM measurements were performed at liquid nitrogen temperature (77 K) on a (1 1 0) surface plane of the sample, which was obtained by cleavage in situ under ultra-high vacuum conditions ($p < 4 \cdot 10^{-11}$ Torr).

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