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Long wavelength (>1.55 μm) room temperature emission and anomalous structural properties of InAs/GaAs quantum dots obtained by conversion of In nanocrystals

A. Urbańczyk,[1,a)] J. G. Keizer,[1] P. M. Koenraad,[1] and R. Nötzel[2]
[1]*COBRA Research Institute on Communication Technology, Department of Applied Physics, Eindhoven University of Technology, 5600 MB Eindhoven, The Netherlands*
[2]*Institute for Systems Based on Optoelectronics and Microtechnology (ISOM), ETSI Telecommunication, Technical University of Madrid, Ciudad Universitaria s/n, 28040 Madrid, Spain*

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We demonstrate that molecular beam epitaxy-grown InAs quantum dots (QDs) on (100) GaAs obtained by conversion of In nanocrystals enable long wavelength emission in the InAs/GaAs material system. At room temperature they exhibit a broad photoluminescence band that extends well beyond 1.55 μm. We correlate this finding with cross-sectional scanning tunneling microscopy measurements. They reveal that the QDs are composed of pure InAs which is in agreement with their long-wavelength emission. Additionally, the measurements reveal that the QDs have an anomalously undulated top surface which is very different to that observed for Stranski-Krastanow grown QDs. © *2013 American Institute of Physics*. [http://dx.doi.org/10.1063/1.4792700]

Molecular beam epitaxy (MBE) growth of InAs/GaAs based materials is a very well established technology capable of producing high-quality low-dimensional structures such as quantum wells and quantum dots (QDs) which constitute essential building blocks of modern optoelectronic devices. One of the long pursued research goals is the extension of the QD emission wavelength up to or even beyond 1.55 μm, where the absorption of silica optical fibers is lowest. This would allow for a cheaper alternative to the presently used InP-based devices for long-distance optical communication. The QDs are usually obtained by the Stranski-Krastanow (SK) growth mode. It is well known that at typical QD growth temperatures significant mass transport occurs resulting in pronounced In/Ga intermixing.[1] This increases the band gap energy and, hence, is clearly not desirable when long-wavelength emission is to be obtained. Intermixing can be minimized by growth at very low temperature,[2] however, at the cost of introducing non-radiative recombination centers, degrading the optical quality of the QDs. Another approach to achieve long emission wavelengths is embedding the QDs in InGaAs strain reducing layers.[3–5] But this reduces the depth of the confinement potential having adverse effects on the temperature characteristics of lasers containing such structures.[6] Therefore, we consider an alternative approach to grow high In-composition QDs which is derived from droplet epitaxy (DE).[7–9] DE is an inherently low growth temperature process made possible by the decoupling of the group III and group V fluxes. We recently found that in order to obtain small and uniform InAs QDs following DE, it is actually necessary to begin with solid In nanocrystals (NCs)[9] and not with liquid droplets. In this case the initial exposure to As_4 flux and conversion process take place when the NCs are still solid, allowing in principle for extremely In-rich QDs due to the lack of intermixing with the substrate. Obviously some intermixing does occur during the necessary subsequent annealing and capping steps; however, it can be minimized when those steps are performed at as low as possible temperature.

Here we report the growth of InAs/GaAs QDs by conversion of epitaxial, solid In NCs that emit over a wide wavelength range extending beyond 1.55 μm. This result is complemented with atomic force microscopy (AFM) investigations of the morphology of uncapped QDs, as well as measurements of the shape and composition of buried QDs by means of cross-sectional scanning tunneling microscopy (XSTM) with atomic-scale resolution. The latter measurements reveal that the QDs are composed of pure InAs and have a complex undulated top surface which is qualitatively different to that of SK-grown QDs. The analysis of both the AFM and XSTM data reveals that there is a significant change of the QD shape and large material redistribution during the capping process.

All samples were grown by solid-source MBE on (100)-oriented GaAs substrates. The growth commenced with a 100-nm-GaAs buffer layer at 580 °C with As_4 beam-equivalent pressure of 5×10^{-6} mbar. The samples were subsequently cooled down to 450 °C, and the As_4 flux was switched off. This established a (2×4) surface reconstruction, monitored by reflection high-energy electron diffraction (RHEED), which was frozen-in during cooling down to 80 °C (thermocouple reading). After 30 min temperature stabilization 0.5 nm In was deposited at a growth rate of 0.045 nm/s in order to obtain In NCs. The background pressure during that process was lower than 2×10^{-9} mbar allowing the (2×4) surface reconstruction to be preserved during the long temperature stabilization step. Then the As flux was turned on, and the substrate temperature was increased to 450 °C at a rate of 25 °C per minute. After additional 5 min annealing the QDs were capped with 20-nm-GaAs at the same temperature. Next, the temperature was further increased to 580 °C and 180-nm-GaAs was deposited. For surface morphology studies

a)Author to whom correspondence should be addressed. Electronic mail: adam.jan.urbanczyk@gmail.com.

by AFM (tapping-mode in air) a sample was grown under the same growth conditions without capping. The structure and composition of the capped QDs were accessed by XSTM at room temperature (RT) with atomic-scale resolution.[10,11] The STM was operated in constant current mode (I = 40 pA) on *in situ* cleaved (011)-surfaces at negative bias voltage ($U_{bias} = -3$ V). Under such conditions InAs (GaAs) rich regions appear bright (dark) in the topographic XSTM images. The optical properties of the capped QDs were investigated by RT photoluminescence (PL) spectroscopy. A green 532-nm solid-state laser was used for excitation. The PL was dispersed by a quarter-meter monochromator and detected by a liquid nitrogen cooled InGaAs photodiode array detector.

Figure 1(a) shows the AFM image of the uncapped QDs, and Fig. 1(b) presents a histogram of the measured QD heights. The QDs are dome-shaped with density of 160 per μm^2, which is a moderate value despite the low deposition temperature of the In NCs. This is the consequence of high In ad-atom mobility on GaAs in the absence of As flux.[12] As evident from Fig. 1(b) the average QD height is 6.5 nm, and the half-width of the height distribution equals to 4 nm. Such large heights are maintained due to the relatively short and low-temperature annealing.

Most relevant, however, is the final QD shape and composition after capping, which is known to cause significant material redistribution to flatten the QDs.[13] Figure 2(a) shows the XSTM image of an individual QD after capping with GaAs. The QD is on average 3.5 nm high and 15 nm wide and has an undulated top surface with height maxima in the center and at the periphery. This is in distinct contrast to SK-grown QDs which maintain a lens or truncated pyramid like shape.[13,14] Such modulated QD shape is commonly found in the studied sample, as shown in Fig. 2(b). Moreover, the QDs are composed of pure InAs. There is no signature of individual Ga atoms inside the QDs which would appear as dark spots. This is in agreement with the initial low-temperature growth of the In NCs and their immediate conversion to InAs QDs, preventing intermixing with the GaAs substrate as well as etching into the underlying layer which happens for GaAs/AlGaAs QDs grown by droplet epitaxy at higher temperatures.[15]

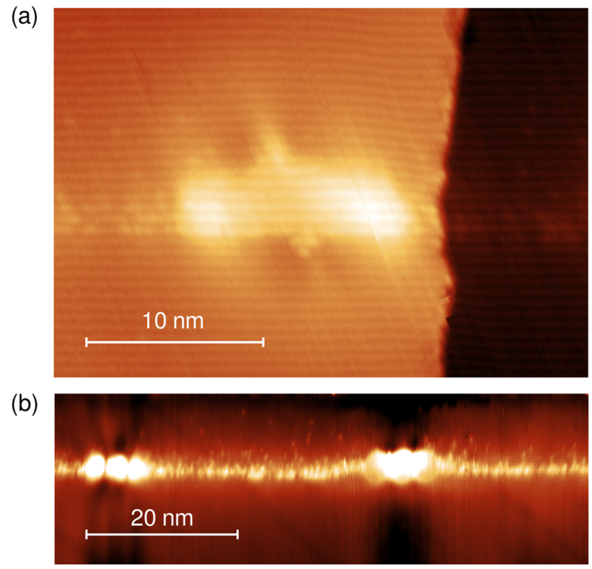

FIG. 2. (a) XSTM image of an individual InAs QD obtained by conversion of an In NC. (b) XSTM image of two InAs QDs confirming the characteristic wavy shape. The dark contrast on the right side visible in (a) is an artifact due to the presence of a step edge.

We attribute the anomalously undulated shape of the buried QDs to significant morphological changes occurring during the capping process. Such changes are evident from the large difference in height of the QDs observed with XSTM compared to the average height of the surface QDs investigated by AFM. A possible explanation of such behavior is as follows: due to the pure In content, the freshly converted QDs are highly strained, preventing complete overgrowth to leave the central part of the QDs exposed during the initial capping. This induces significant material redistribution during heating up for the final thick GaAs layer growth at 580 °C. At this temperature, the partially exposed top of the QDs readily decomposes and In desorbs, leaving the final buried QDs greatly flattened. Similar to the case of SK-grown QDs, and intentionally enhanced by the so called In-flush technique,[16] this process certainly takes place to reduce the overall height of the QDs, but does not explain the modulated shape of our QDs. Therefore, we believe that the final shape of the QDs is additionally determined by the formation process of the QDs itself. For QDs formed by DE it has been found that the recrystallization process starts from the side of the droplets, proceeding to the top. This

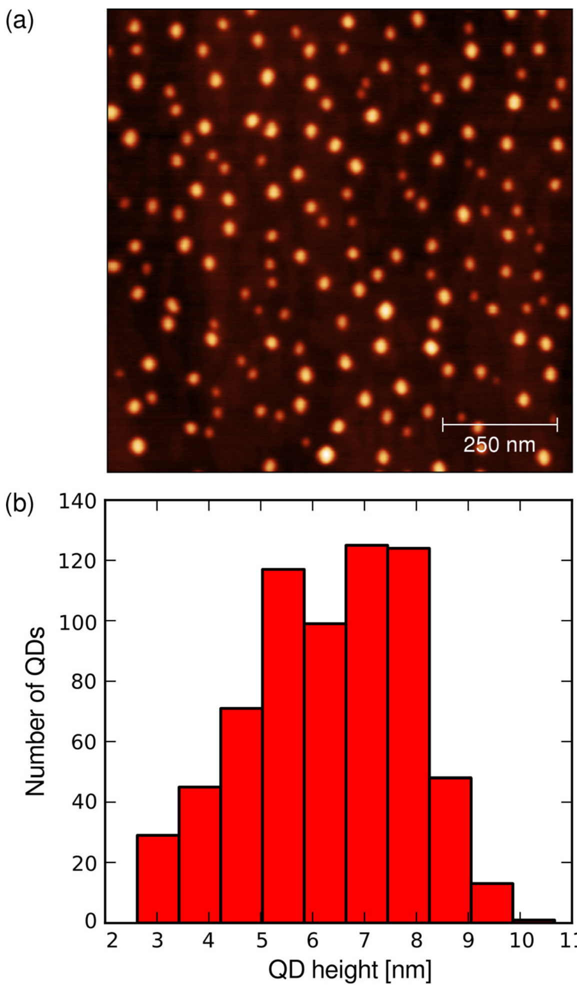

FIG. 1. (a) AFM image of uncapped InAs QDs and (b) histogram of the QD heights. The AFM scan area is 1 × 1 μm^2 and the full height contrast is 12 nm.

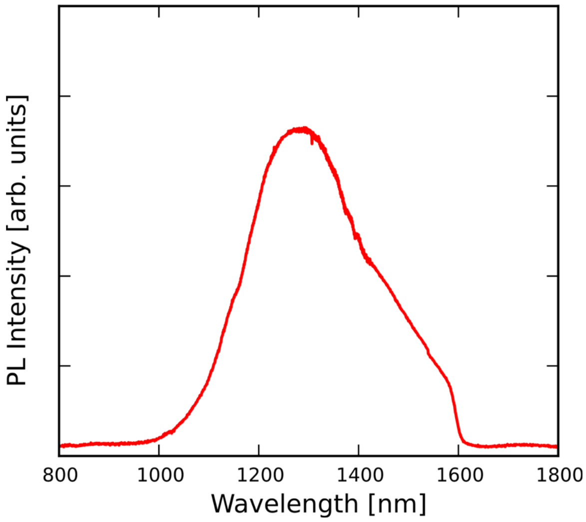

FIG. 3. RT PL spectrum of the InAs QDs. The detector cut-off is at 1.6 μm.

suggests a better crystal quality at the side and, hence, higher stability during overgrowth. Together with the initially highest part in the center this might lead to a buried QD with the observed shape, i.e., highest in the center and the periphery. Of course, other mechanisms leading to undulated topography of strained layers should not be excluded.[17,18]

The result of the RT PL measurement is shown in Fig. 3. The QDs exhibit a very broad emission band with intensity maximum at 1285 nm and half-width of 320 nm. Such a large inhomogeneous broadening is most likely a consequence of the broad distribution of initial QD heights and the complex overgrowth process. What is most important, the emission extends significantly beyond 1.55 μm, even beyond the detector cut-off at 1.6 μm, notably without the need for strain reducing layers. This long emission wavelength is mainly attributed to the QDs being composed of pure InAs which has proven to be difficult to obtain for SK-grown QDs of high optical quality. Along these lines, further optimization of the QDs for specific applications is straight-forward due to the excellent control of size and density of the epitaxial In NCs,[19] much better than the control achievable for directly grown SK QDs.

To conclude, we have demonstrated room-temperature long-wavelength emission beyond 1.55 μm of InAs QDs on GaAs obtained by conversion of epitaxial In NCs. We also presented the results of AFM and XSTM measurements for structural analysis. These measurements showed that the investigated QDs are composed of pure InAs and exhibit an undulated top surface, very different to that of SK-grown QDs, which has been related to the distinct QD formation process. We believe that the presented findings are a stepping stone for the usage of the InAs/GaAs material system for optoelectronic devices operating in the second telecommunication window and beyond. Furthermore, we believe that our findings can be directly applied to other material systems, such as InAs/InP, for extending the emission into the mid-infrared.

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