Optical and structural properties of InAs quantum dots in a GaAs matrix for a spectral range up to 1.7 μm

M. V. Maximov,[a] A. F. Tsatsul'nikov, B. V. Volovik, D. A. Bedarev, A. Yu. Egorov, A. E. Zhukov, A. R. Kovsh, N. A. Bert, V. M. Ustinov, P. S. Kop'ev, and Zh. I. Alferov
A. F. Ioffe Physico-Technical Institute, St.-Petersburg 194021, Russia

N. N. Ledentsov[b] and D. Bimberg
Institut für Festkörperphysik Technische Universität Berlin, 10623 Berlin

I. P. Soshnikov[c] and P. Werner
Max-Planck-Institut für Mikrostrukturphysik, Weinberg 2, D-06120 Halle, Germany

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We demonstrate the possibility of extending the spectral range of luminescence due to InAs quantum dots (QDs) in a GaAs matrix up to 1.7 μm. Realization of such a long wavelength emission is related to formation of lateral associations of QDs during InAs deposition at low substrate temperatures (~320–400 °C). © *1999 American Institute of Physics.* [S0003-6951(99)00242-9]

In recent years self-organized quantum dots (QDs)[1] have found significant interest. One of the important advantages of QDs is their potential to shift the emission wavelength beyond the range available for quantum well (QW) structures on the same substrate.[2–4] Recently 1.3 μm GaAs-based InAs QD lasers are shown to have low threshold current densities (<100 A/cm^2).[2,3] High temperature stability of threshold current, low internal losses, and good differential efficiency are also demonstrated.[3] For applications in long-haul transmitters, cost-effective 1.55 μm emitting GaAs-based lasers are desirable. High-power 1.48 μm GaAs-based devices could serve as pumps for optical-fiber amplifiers.

In this letter we show that a spectral range up to 1.75 μm can be covered by InAs QDs in a GaAs matrix. The main approach to reach long wavelength emission is using lateral associates of QDs which were earlier observed in the upper stacks of vertically coupled QDs.[4]

Samples are grown by elemental-source molecular-beam epitaxy (MBE) on GaAs (001) substrates using a Riber-32 MBE system. The evolution of the surface morphology is studied *in situ*, employing reflection high energy electron diffraction (RHEED). The growth procedure is as follows. After oxide desorption, a 0.5-μm-thick GaAs buffer is grown at 600 °C followed by a 2 nm/2 nm GaAs–AlAs short period superlattice (ten periods) and a 100 nm GaAs layer. Then the substrate temperature is lowered to 300–480 °C and the QDs are deposited. Afterwards the dots are overgrown by 10 nm of GaAs at the same substrate temperature. The temperature is then increased again to 600 °C and a 20-nm-thick GaAs layer is grown. This layer is followed by six periods of 2 nm/2 nm GaAs–AlAs short period superlattice and 5-nm-thick GaAs cap layer for surface protection. The sample geometry enables optical studies as well as cross-section and plan-view *ex situ* characterization using transmission electron microscopy (TEM) and high resolution electron microscopy (HREM). TEM and HREM studies are performed using a high voltage JEOL JEM1000 (1 MV) microscope. Photoluminescence (PL) is excited by an Ar^+ laser and detected by Ge or InSb photodiodes. The spectra are corrected according to the spectral sensitivity curves. For photoluminescence excitation (PLE) experiments, samples are mounted into a continuous flow He cryostat at 6 K. PLE spectra are recorded using the light of a halogen lamp dispersed through a monochromator.

Structural and optical properties of QDs deposited at temperatures of 460–520 °C were studied in our previous work and by other researchers.[5,6] It was shown that deposition of 4 ML of InAs leads to the formation of a dense array of QDs having pyramidal shape with a square base.[6] The principal axes of the pyramid's base are close to the $\langle 100\rangle$ or $\langle 010\rangle$ directions.[6] For QDs grown at 520 °C the average length of the pyramid base is 18 nm with the dot density about 2×10^{10} cm^{-2}. A reduction of the substrate temperature to 460 °C leads to a decrease of the average length of the dot base to 12 nm and to an increase in the density of QDs to 1×10^{11} cm^{-2}. These QDs demonstrate bright PL emission in the range of 1.07–1.1 eV at 10 K. We note that the density of QDs deposited at 460 °C is so high that lateral interaction effects via the strain fields during the QDs formation become important and govern the QD arrangement.[7] One can expect that a further reduction of substrate temperature during InAs deposition will result in a further increase in the density of QDs making the lateral interaction of QDs more important. Indeed, TEM studies of the samples deposited at 325–350 °C demonstrate that the QD density can increase up to 10^{12} cm^{-2}, while the QD size decreases down to 6–7 nm. However, an increase in the deposited thickness above 2 ML results in a qualitative change in the arrangement of QDs.

Figure 1 shows plan-view (a) and the cross-section (b) TEM images of a sample with QDs formed by 4 ML of InAs deposited at 325 °C. Individual QDs with a size of about 10 nm are seen in this image. Their areal density is about

[a]Also at the Institut für Festkörperphysik Technische Universität Berlin.
[b]Also at the A. F. Ioffe Physico-Technical Institute.
[c]Also at the Institut für Festkörperphysik Technische Universität Berlin; electronic mail: iposh@hotmail.com

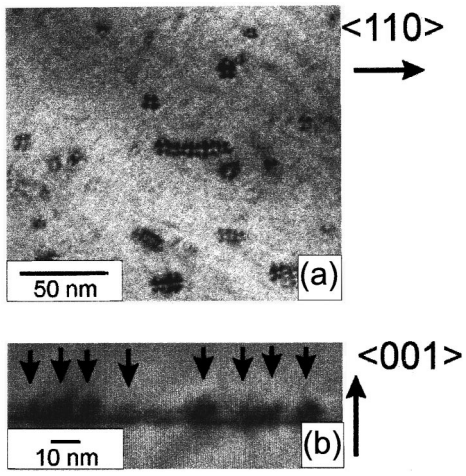

FIG. 1. Bright field plan-view (a) and cross-section (b) TEM images of InAs QDs formed by deposition of 4 ML InAs at 350 °C.

$5\times10^{10}\,\mathrm{cm}^{-2}$. We note that the principal axes of the QD bases are oriented either along ⟨100⟩ or ⟨110⟩ directions. Besides individual QDs, other objects with larger size and a complex shape are revealed in both the plan-view TEM [Fig. 1(a)] and the cross-section HREM [Fig. 1(b)] images. Below, we will refer to these complex objects as lateral associations of QDs (LAQDs). Each LAQD consists of a number of well-defined segments with a lateral size of 6–7 nm, which is clearly seen on the large scale HREM images [Figs. 2(b) and 2(c)]. The density of LAQDs is found to be about $2\times10^{10}\,\mathrm{cm}^{-2}$. Two different types of LAQDs are seen in Fig. 1 and Figs. 2(b) and 2(c). In the first case segments forming a LAQD are attached to one another only in one ⟨110⟩ direction so that the LAQD has a chain-like character [Fig. 2(b)]. In the second case the segments are attached to one another in two directions so that LAQD consists of rectangular arrays of closely located segments aligned along the ⟨110⟩ and $\langle 1\bar{1}0\rangle$ directions.

We note that the lateral size of a segment corresponds to the size of a single QD formed by 2 ML of InAs deposition at the same substrate temperature. One might conclude that the density of QDs formed by the deposition of 2 ML of InAs is already so high that during further InAs deposition an increase in the lateral size becomes energetically unfavorable due to elastic repulsion of the islands.[7] Formation of LAQDs can result in a decrease of the elastic relaxation energy if the shape of the QD is kept constant. However, if the facet angle of the InAs segments is increased, the elastic relaxation energy can be larger in the LAQD case. On the other hand, the QD associations absorb a significant amount of the InAs deposited and allow to keep the density of conventional single QDs low, avoiding their interaction.

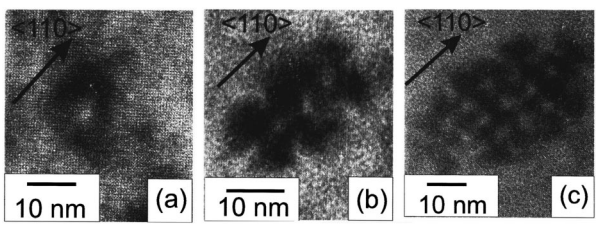

FIG. 2. High resolution electron microscopy images of individual (a) and laterally associated (b,c) QDs.

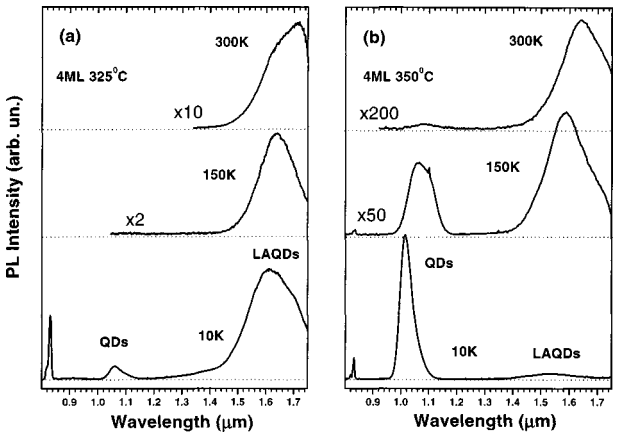

FIG. 3. Photoluminescence (PL) spectra recorded at different temperatures for the samples with 4 ML InAs deposited at 325 °C (a) and 350 °C (b).

The substrate temperature of 320–340 °C seems to be optimal for the formation of LAQDs. InAs deposition at a lower substrate temperature of 350 °C results in a LAQDs density decrease down to $1\times10^{10}\,\mathrm{cm}^{-2}$ and an increase in concentration of individual QDs. Further increase in substrate temperature up to 480–500 °C results in a complete suppression of LAQDs formation with only individual QDs seen in micrographs. On the other hand, decrease of the substrate temperature down to 300 °C suppresses the formation of any kind of QDs, probably due to kinetic limitations.

Formation of LAQDs manifests itself in a pronounced change of the PL spectra. Temperature dependencies of the PL spectra for the samples with 4 ML InAs deposited at 325 and 350 °C are shown in Figs. 3(a) and 3(b), respectively. There are two PL lines in the spectra recorded at 10 K. The short wavelength line at about 1.05 μm is characteristic of samples with QDs formed by the deposition of 4 ML InAs at 480–520 °C where formation of LAQD is not observed. However, samples grown at 325–350 °C have an additional long wavelength PL line at 1.5–1.6 μm[8] (Fig. 3). This emission is not observed for the samples where LAQDs do not form, e.g., in the case when the average thickness of the InAs deposited at low substrate temperature is below the value necessary for the formation of the islands, as controlled by RHEED studies. Thus, we attribute this line to the PL from LAQDs. The temperature shift of the line is in agreement with the temperature dependence of the InAs band gap, which confirms that the emission originates from the nanostructures rather than from dislocations or point defects. The fact that the LAQD luminescence is redshifted with respect to PL from individual QDs agrees well with the formation of nanostructures that strongly coupled in the lateral direction. A similar effect was reported in Ref. 5 for lateral chains of QDs formed in the upper rows of vertically coupled QDs. Each LAQD consists of several closely located segments. The separation between them is very small, electron wave functions of the neighboring islands essentially overlap, and a LAQD represents a single nanostructure with an effectively increased lateral size. Lateral coupling effects significantly decrease the energy of charge carriers, increase their localization, and lead to a redshift of the PL line.

At room temperature, the wavelengths of the peak

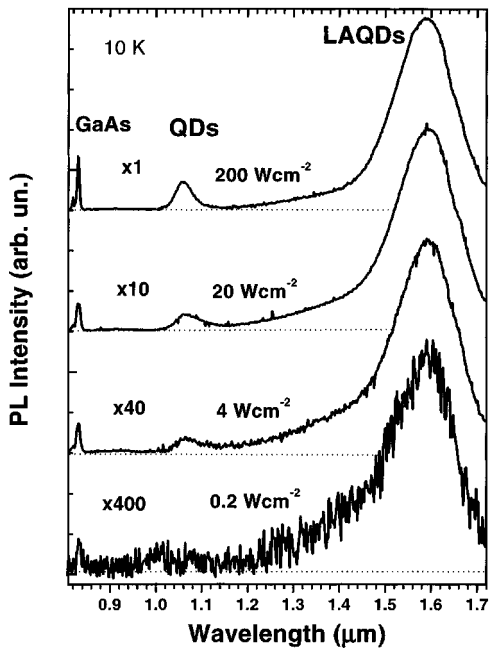

FIG. 4. PL spectra for the sample with 4 ML InAs deposited at 325 °C recorded at different excitation densities.

maxima are about 1.72 and 1.65 μm for the samples grown at 325 °C [Fig. 3(a)] and 350 °C [Fig. 3(b)], respectively. The width of the PL lines is about 100 meV. The LAQDs PL intensity drops by only one order of magnitude when the observation temperature increases from 10 to 300 K.

Figure 4 shows the excitation density dependence of PL for the sample with 4 ML InAs deposited at 325 °C. The integral intensity of the LAQD luminescence increases linearly with excitation density up to high excitation densities. It is important to note that the intensity of the QD emission increases superlinearly with excitation density, pointing to saturation of nonradiative recombination centers at high excitation densities.

To confirm further the QD nature of the observed long wavelength PL line, we performed PLE studies. Figure 5 shows PL and PL excitation (PLE) spectra, recorded at 10 K for the sample with 4 ML InAs deposited at 325 °C. The shape of the PLE spectra is very typical for QD structures.[9] The PL signal appears to be strong only when the excited states of the QDs are populated. Significant intensity of the long wavelength emission under resonant excitation with a photon energy below the GaAs band gap entirely excludes defects formed in the low temperature thin GaAs layer as being the source of the luminescence.

To conclude, we have demonstrated that laterally associated InAs QDs formed in a GaAs matrix emit up to a wavelength of ~1.75 μm. This is important for the future

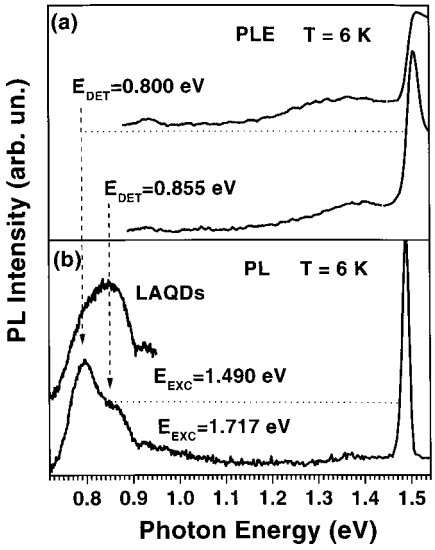

FIG. 5. PL excitation (PLE) spectra (a) for the sample with LAQDs formed by 4 ML InAs deposition at 325 °C. PL spectra (b) are recorded using exciting photon energy above and below the energy of the GaAs band gap. For PLE spectra the arrows indicate the detection energies.

coverage of the Telecom spectral range using GaAs-based devices.

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