The effects of quantum dot coverage in InAs/(In)GaAs nanostructures for long wavelength emission

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Abstract

We present a study on the effects of quantum dot coverage on the properties of InAs dots embedded in GaAs and in metamorphic In0.15Ga0.85As confining layers grown by molecular beam epitaxy on GaAs substrates. We show that redshifted emission wavelengths exceeding 1.3 µm at room temperature were obtained by the combined use of InGaAs confining layers and high quantum dot coverage. The use of high InAs coverage, however, leads to detrimental effects on the optical and electrical properties of the structures. We relate such behaviour to the formation of extended structural defects originating from relaxed large-sized quantum dots that nucleate in accordance to thermodynamic equilibrium theories predicting the quantum dot ripening. The effect of the reduced lattice-mismatch of InGaAs metamorphic layers on the quantum dot ripening is discussed in comparison with the InAs/GaAs system.

Keywords

Quantum dots; long wavelength emission; molecular beam epitaxy; quantum dot ripening.
Introduction

In recent years, great interest has been devoted to optoelectronic devices based on quantum dot (QD) structures [1,2] and one of the major concerns is actually related to the preparation of long wavelength (> 1.3 µm) emitting QD structures grown on GaAs substrates. To this aim, different approaches have been used, among which the growth of QDs on metamorphic buffers [3,4] proved to be useful for the achievement of room temperature (RT) emission wavelengths exceeding 1.5 µm [3,5]. While redshifted emissions are obtained by decreasing the band discontinuities between QDs and confining layers (CL), it has been shown that the activation energy for thermal quenching of luminescence inevitably decreases when such band discontinuities are reduced [6]. As a consequence, QD structures engineered to emit at long wavelength may suffer from poor emission efficiency at RT. Redshifting emission wavelength by increasing the QD sizes does not have such a drawback and, since it can be simply obtained by increasing the QD coverage (θ), it is usually combined with other more sophisticated approaches in order to reach the 1.3 – 1.5 µm spectral window. While several experimental results report on the detrimental effects of large-sized QDs on the properties of QDs embedded in GaAs CLs [7,8], such studies have not yet been reported for QD structures grown on InGaAs metamorphic buffers. This work is aimed at studying the effects of θ on the properties of InAs quantum dots embedded in GaAs and in In_{0.15}Ga_{0.85}As metamorphic CLs in structures grown by molecular beam epitaxy (MBE) on (001) GaAs substrates. Capped QD structures were characterised by photoluminescence (PL), capacitance-voltage (C-V), deep level transient spectroscopy (DLTS), transmission electron microscopy (TEM) and X-ray diffraction, while the morphological and structural properties of uncapped QD structures were studied by atomic force microscopy (AFM) and plan view TEM.

Experimental

InAs/GaAs QD structures were prepared by MBE with a coverage θ in the 1.5 – 2.9 ML range
according to Ref. [9]. InAs/In$_{0.15}$Ga$_{0.85}$As metamorphic QD structures with $\theta$ ranging between 2.0 – 3.0 ML were grown by MBE on GaAs semi-insulating substrates and consist of a single layer of InAs QDs grown by Atomic Layer MBE [10] embedded in a 20 nm-thick undoped In$_{0.15}$Ga$_{0.85}$As layer. Such a layer was grown on a 0.8 $\mu$m n-doped In$_{0.15}$Ga$_{0.85}$As lower CL (LCL) and it was capped by an In$_{0.15}$Ga$_{0.85}$As upper CL (UCL) consisting of a 0.3 $\mu$m n-doped layer followed by 10 nm $p^+$-doped cap.

Contact-mode AFM measurements were carried out under ambient conditions with allowance made for QD-tip convolution effects. 10 K and RT PL was excited by a 532 nm laser source with a 5 W/cm$^2$ excitation power density and measured by means of a fast Fourier transform spectrometer fitted with a cooled Ge detector. For C-V and DLTS measurements, planar Schottky barriers were prepared by evaporating Au rectifying dots, 400 $\mu$m in diameter, and AuGeNi ohmic contacts on the cap layer. C-V measurements were performed by a 4192 HP impedance analyser while DLTS measurements were carried out by a high sensitivity lock-in type spectrometer. Plan view and cross section TEM investigations were performed by a Jeol 2000FX microscope working at 200 kV.

**Results**

By increasing $\theta$ beyond 1.55 ML in the InAs/GaAs QD system, we observe the nucleation of three-dimensional (3D) islands that undergo a fast increase in density, until a saturation value of $\sim2\times10^{11}$ islands/cm$^2$ is reached when $\theta$ is 2.2 ML. The island heights and base diameters, given by the most frequent values of the corresponding AFM size distributions, increase with increasing $\theta$ and reach the maximum values of 2.7 nm and 15 nm, respectively. Noticeably, while $\theta$ increases in the density-saturation regime, a second family of 3D islands shows up as evidenced in Fig. 1(b) for $\theta = 2.9$ ML. The heights and density of these large-sized islands increase with $\theta$, up to values of 6 nm and $1.5\times10^9$ islands/cm$^2$, respectively, while their base diameters remain almost constant at 25 nm. As shown in Fig. 1(c) and (d), the metamorphic In$_{0.15}$Ga$_{0.85}$As layer surfaces underlying the 3D islands are characterised by a peculiar
morphology consisting of cross-hatch patterns along the ⟨110⟩ directions [11]. Such a morphology hinders the detailed observation of the properties of 3D island distributions. However, similarly to what observed on GaAs surfaces, by increasing the InAs coverage on metamorphic In₀.₁₅Ga₀.₈₅As layers (Fig. 1(d)) we were able to distinguish, beside the family of small-sized islands, some large-sized 3D islands with density lower than 1x10⁸ islands/cm², for θ = 3.0 ML. At this coverage the small-sized islands have a surface density of 9x10¹⁰ islands/cm², while their size is larger as compared to the size of the InAs/GaAs islands at a coverage of 2.9 ML. The nucleation of large-sized islands observed on both GaAs and InGaAs surfaces is consistent with thermodynamic equilibrium theories of lattice-mismatched heteroepitaxy [12] which show that, independently of lattice-mismatch and for high enough coverage, unstable ripened islands coexisting with the stable ones are going to form. Being energetically unstable, the size of ripened islands is expected to increase indefinitely, giving rise to strain relaxation through the formation of structural defects that may lead to detrimental effects on the properties of the structures. According to Monte Carlo simulations [13,14], the larger size and lower density of 3D islands nucleated on In₀.₁₅Ga₀.₈₅As surfaces as compared to GaAs ones may stem from the lower lattice-mismatch existing between InAs and In₀.₁₅Ga₀.₈₅As metamorphic LCL (6.3%, calculated on the basis of Ref. [15] and validated by Ref. [16], instead of 7.2%). An accurate study of the QD properties in connection with the formation of large-sized QDs is thus needed in order to verify the anticipations of equilibrium theories.

Consistently with the increase in QD size observed by AFM, the emission energy of InAs/(In)GaAs QD structures is redshifted by increasing θ as represented in Fig. 2(a). More interestingly, we note that in the whole 2.0 – 3.0 ML coverage range, the emission energy of InAs QDs embedded in In₀.₁₅Ga₀.₈₅As metamorphic CLs is redshifted by more than 150 meV with respect to the emission energy of the InAs/GaAs QD structures. Such a result may be attributed to: i) the reduced QD-to-CL band discontinuities provided by the In₀.₁₅Ga₀.₈₅As CLs as compared to the GaAs ones, ii) the decreased lattice-mismatch between InAs QDs and the
partially relaxed $\text{In}_{0.15}\text{Ga}_{0.85}\text{As}$ CLs and, then, the decreased QD strain, and iii) the larger size of 3D islands grown on $\text{In}_{0.15}\text{Ga}_{0.85}\text{As}$ surfaces with respect to GaAs ones, as observed by AFM. As a consequence, the combined use of high-coverage QDs and $\text{In}_{0.15}\text{Ga}_{0.85}\text{As}$ metamorphic layers led to 1.32 $\mu$m RT emission from the 3.0 ML InAs/GaAs QD structure. We note that the integrated intensity of the 10 K PL spectra of the InAs/(In)GaAs QD structures decreases concomitantly with the nucleation of the large-sized QD family (Fig. 2(b)). As explained in Ref. [9], such a behaviour is not consistent with the decrease in the oscillator strength of redshifted QD emission and, then, suggests the existence of non-radiative recombination processes that compete with the QD emission. Similarly to the integrated PL intensities, the electrical properties of the InAs/(In)GaAs QD structures show different characteristics in the low- and high-$\theta$ regime. In Fig. 3 we report the profile of apparent carrier concentration for InAs/GaAs and InAs/$\text{In}_{0.15}\text{Ga}_{0.85}\text{As}$ structures with coverages of 2.0 and 2.9 ML and of 2.0 and 3.0 ML, respectively. Both low-$\theta$ structures show a clear accumulation peak centred around the depth of QDs and wetting layers that can be related to quantum confined carriers. On the contrary, the samples with high coverages (2.9 and 3.0 ML), exhibit: i) a large carrier depletion close to the interface between the UCL and the QDs and ii) a reduction, for the InAs/GaAs structure, or the disappearance, for the InAs/$\text{In}_{0.15}\text{Ga}_{0.85}\text{As}$ one, of the accumulation peak. DLTS measurements show the occurrence of traps that we relate to extended defects in the QD region owing to the logarithmic dependence of the DLTS signal amplitude on the filling pulse width [17]; in InAs/GaAs structures such traps have been observed for $\theta \geq 2.4$ ML, while in the InAs/$\text{In}_{0.15}\text{Ga}_{0.85}\text{As}$ counterparts they are already present at $\theta = 2.0$ ML and their concentration increases with increasing $\theta$ from 2.0 to 3.0 ML. On the basis of the C-V and DLTS results, we suggest that the free carrier depletion observed for high QD coverage could be attributed to acceptor-like defects produced by extended defects. The occurrence of extended defects is confirmed by TEM measurements: as for the InAs/GaAs QD structures, TEM plan view and cross section analyses showed that strain relaxation of large-sized QDs occurs by the formation
of pure edge-type misfit dislocations which propagate towards the surface forming V-shaped defects having the apex at the QDs/GaAs interface [18]. Their density increases from 6.1x10^7 cm^-2 to 1.2x10^9 cm^-2 in the 2.4 ≤ θ ≤ 2.9 ML coverage range. Independently of the InAs coverage, the InAs/In_{0.15}Ga_{0.85}As metamorphic QD structures exhibit a network of misfit dislocations at the InGaAs/GaAs interface accompanied by threading dislocations propagating through the whole thickness of the structures with a density of 1x10^8 cm^-2. In addition to these dislocations, V-shaped defects nucleating at the QD plane are revealed in the θ = 3.0 ML structure with a density of 4x10^7 cm^-2. Such a density is consistent with the density of large-sized 3D islands revealed by AFM, thus suggesting that the ripening could be responsible for their formation, similarly to what previously found in the InAs/GaAs QD structures.

**Discussion and conclusions**

The effect of QD coverage on the properties of InAs/(In)GaAs QD structures has been presented. We have shown that high QD coverages lead to the nucleation of large-sized ripened QDs that coexist with the small-sized ones, consistently with the thermodynamic theory of lattice-mismatched growth by Daruka and Barabasi [12]. Such ripened QDs undergo relaxation through the formation of V-shaped defects that we relate to: i) the decrease in the low temperature integrated PL intensity, ii) the depletion of carriers from QD levels and iii) the presence of deep levels associated to extended defects in the QD region. Very interestingly, we observed a reduced density of large-sized QDs, and of the corresponding V-shaped structural defects when InAs QDs were grown on the In_{0.15}Ga_{0.85}As metamorphic layers. This difference can be ascribed to the reduced lattice-mismatch experienced by the QDs grown on metamorphic CLs, and thus to the different critical coverages for the ripening onset [12]. In addition, the presence of the pre-existing threading dislocations, which are not taken into account in the thermodynamic equilibrium approach presented by Ref. [12], can also play an important role in the delay of the ripening onset in the InAs/InGaAs system. Such threading dislocations, being related to strain relaxation in In_{0.15}Ga_{0.85}As metamorphic LCL and not to the coverage θ, may be
responsible for the deep levels detected in the low-θ InAs/In_{0.15}Ga_{0.85}As QD structures.

We have shown that the combined use of high θ and metamorphic CLs is a viable approach to redshift the emission wavelength of QD structures and that, in spite of the occurrence of extended defects originated from ripened QDs, the PL intensities decrease by less than the 50% with respect to the low-θ structures. Our results suggest that such a drawback should be addressed by selecting a suitable set of growth conditions that allow not only the optimisation of the structural properties of metamorphic layers [3] but also the kinetic limitation of the QD ripening.
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References


Figure captions

Fig. 1  1 µm² AFM images of the uncapped: i) InAs/GaAs QD structures with (a) $\theta = 2.0$ ML and (b) $\theta = 2.9$ ML and ii) InAs/InGaAs QD structures with (c) $\theta = 2.0$ ML and (d) $\theta = 3.0$ ML.

Fig. 2  (a) 10 K emission energies as functions of the QD coverage for the InAs/GaAs (closed circles) and InAs/InGaAs (open circles) structures; (b) 10 K PL spectra of the InAs/InGaAs (continuous lines) and InAs/GaAs (dashed lines) QD structures; the high-$\theta$ PL spectra are normalised to the low-$\theta$ ($\theta = 2.0$ ML) ones of each series.

Fig. 3.  Apparent free carrier concentration profiles of InAs/GaAs (closed symbols) and of InAs/In$_{0.15}$Ga$_{0.85}$As (open symbols) QD structures with coverages $\theta$ of 2.0 and 2.9 ML and of 2.0 and 3.0 ML, respectively.
Fig. 2
Fig. 3