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Submicron Raman and photoluminescence topography of InAs/Al(Ga)As quantum dots structures

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ABSTRACT

Two-period structures with and without vertical coupling between indirect and direct bandgap InAs quantum dots (QDs) both with type I band alignment, grown by molecular-beam epitaxy, were investigated by confocal Raman and photoluminescence (PL) microspectroscopy. The observed blue shift of PL band of the indirect (direct) bandgap QD by 20 (80) meV with decrease of thickness of Ga(Al)As intermediate layer between two InAs QD layers from 30 to 8 nm is considered as caused by increase of elastic strains (decrease of QDs sizes) in QD layers and by coupling between QDs electronic states. Scanning confocal resonant Raman microspectroscopy was applied for non-destructive evaluation of composition at various depths along the thickness of vertical coupling of the upper InAs/AlGaAs and lower InAs/AlAs QDs layers of the sandwich structures. Based on the analysis of determined from the in-depth Raman spectra optical phonons frequencies, the depth distribution of composition in InAlAs and GaAlAs alloy layers formed as a result of strain-driven enhanced interdiffusion was determined.

1. Introduction

Heterostructures with the self-organized InAs quantum dots (QDs) grown in the Stranski–Krastanov mode have been extensively investigated because of their potential significance for device application and fundamental physical studies. Due to the unique electronic and optical properties, the Förster resonant exciton energy transfer between two different InAs QDs layers [1] and from the QDs to organic molecules [2] acted as donor and acceptors may be used for fabrication of toxic molecule sensors. At the present time the system of In(Ga)As QDs in GaAs matrix is extensively studied. The system of InAs QDs embedded in AlAs matrix has received much less attention. In comparison with InAs/GaAs QD, the InAs/AlAs QD system is characterized by the increased quantum confinement and the associated longer photoluminescence (PL) decay time and by the PL line shifted into visible spectral range [3]. It is well known that the electron ground state of InAs/AlAs QDs depending on the dot size and the aluminum content may belong to Γ or to X_{xy} valley of the conductivity zone InAs [4,5].

Another feature arises in In(Ga)As/Ga(Al)As heterostructures when the two constituent compounds have different lattice parameters, namely, strong intermixing of InAs and barrier material due to strain-driven InAs segregation [4]. InAs–AlAs intermixing in InAs QD may be considerably reduced only by using the growth at low temperatures and by short-time growth interruptions [4]. The local inhomogeneity of InAs/AlAs distribution across InAs QDs was shown by scanning tunnel electron microscopy [6]. Embedding of Al and Ga from GaAlAs matrix into InAs QD and the lower rate of Al/In intermixing than for Ga/In was shown by Raman spectroscopy [7]. These structure inhomogeneities strongly effect electron and vibration energy structure of InAs QDs, and are of relevance for the fabrication of QD based optoelectronic devices. Despite the number of works devoted to study of composition and strains in InAs/AlAs QDs by transmission electron microscopy (TEM) [4], X-ray diffraction [8] and Raman spectroscopy (RS) [7], so far the impact of submicron scale structural, strain and compositional inhomogeneities on their optical and electronic properties remains unknown. Recently confocal micro-Raman spectroscopy was shown to allow investigations of residual stress depth distribution in strained sapphire crystal with lateral resolution of ≈300 nm and axial depth resolution of ≈600 nm [9], and the technique is non-destructive and non-contact.

In the present paper, the study the spatial inhomogeneity of composition in two-period structures with self-assembled InAs QDs was carried out using scanning confocal Raman and photoluminescence microspectroscopy. To date, such optical studies of InAs/AlAs-QD with submicron spatial resolution are not available.

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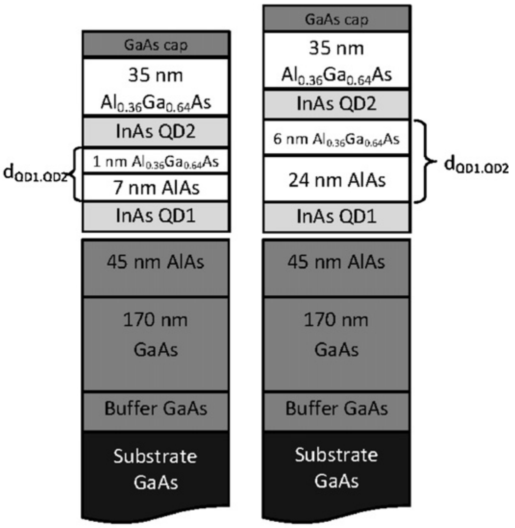

Fig. 1. Two-period structures with coupled InAs QDs. Thickness of the separating layer between the InAs QD layers $d_{(QD1-QD2)}$ = 8 nm (a) and 30 nm (b).

2. Experimental

Two-period structures with InAs QDs grown by molecular beam epitaxy on semi-insulating (0 0 1)-oriented GaAs substrates using Riber-32P system were investigated. The sandwich structures consisted of two vertically coupled InAs QD layers between layers of AlAs (indirect bandgap QDs (QD1)) and $Al_{0.36}Ga_{0.64}As$ (direct bandgap QDs (QD2)) were grown on 170 nm thick GaAs buffer layer. The thickness of Al(Ga)As separating layer between the QD layers was equal to $d_{QD1,QD2}$ = 8 and 30 nm (Fig. 1).

Both QDs layers were deposited at 0.04 ML/s rate to a nominal thickness of 2.5 ML. The QDs were formed at a temperature of 510 °C. A 35 nm $Al_{0.36}Ga_{0.64}As$ layer and 50 nm GaAs cap layer were grown on top of the structure. More detailed description of the growth process was given in [10]. TEM investigations showed the InAs QDs to have lens-like shape with base diameter ~15–20 nm and height ~4–5 nm. Dots density was about 10^{10} cm^{-2}.

Micro-Raman and PL spectra were measured at T = 10/300 K using Horiba Jobin Yvon T64000 spectrometer equipped with confocal microscope and automated piezo-driven XYZ stage. Discrete lines of Ar–Kr ion laser (λ_{exc} = 488.0, 647.0 nm) and He–Cd laser (λ_{exc} = 325.0 nm) with power on sample surface of 1–2 mW were used for excitation. Laser beam was focused on the sample into spot of ~0.2–0.5 μm in diameter. Spatial mapping of the optical spectra was realized by the displacement of the automated stage with spatial step of 0.1 μm.

3. Results and discussion

Fig. 2a and b shows low-temperature micro-PL spectra of the investigated structures with coupled InAs QDs measured at λ_{exc} = 325.0 and 647.0 nm. Ultraviolet excitation is efficiently absorbed by the top direct bandgap $InAs/Al_{0.36}Ga_{0.64}As$ QDs structure, and according to [4], these dots are responsible for low-energy exciton emission band. For the sample with $d_{QD1,QD2}$ = 8 nm, inhomogeneously broadened emission line with the peak energy of 1.55–1.75 eV and full-width on half-maximum (FWHM) of 150–200 meV (QD2 PL band on Fig. 2a) corresponds to the ensemble of direct bandgap InAs QDs. The shape of this band is well approximated with two Gaussian functions with maxima at ≈1.67 eV (Γ = 75 meV) and ≈1.72 eV (Γ = 68 meV), which we attribute to bimodal QDs size distribution, since it does not disappear at low excitation density. At $\hbar\omega_{exc}$ = 3.81 eV, the emission intensity of the bottom layer of indirect bandgap InAs QDs at ≈1.82 eV (QD1 PL band on Fig. 2a) is noticeably less than the intensity of the top layer of direct bandgap InAs QDs, i.e. $I_{QD1} < I_{QD2}$. At $\hbar\omega_{exc}$ = 1.92 eV, the excitation is absorbed in the top and bottom thin direct and indirect bangap InAs QDs layers, and also in 2D AlAs wetting layer (E_{xy}(AlAs) ≈ 2.2 eV [4]). As a result, intensity increase of high-energy PL band at ~1.82 eV (FWHM = 45 eV) corresponding to exciton recombination in indirect bandgap InAs QDs is observed in the PL spectrum. Shape analysis of this band revealed its low-energy asymmetry to be related to radiation with emission of LO(InAs)-like and AlAs-like phonons of In(Al)As [10].

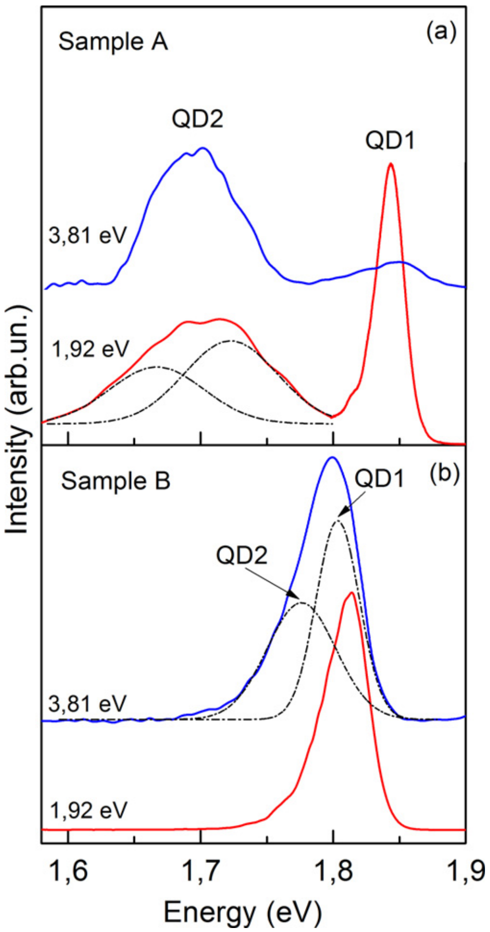

Fig. 2. Low-temperature micro-PL of two-period structures with coupled InAs QDs. Thickness of the separating layer between InAs QD layers $d_{(QD1-QD2)}$ = 8 nm (a) and 30 nm (b). λ_{exc} = 325.0 and 647.0 nm ($\hbar\omega_{exc}$ = 3.81 and 1.92 eV, correspondingly). T = 10 K.

Drastically different situation has place for the sample with thickness of separating Al(Ga)As layer of $d_{QD1,QD2}$ = 30 nm (Fig. 2b). Although, the bottom layer of InAs QDs was grown at the same conditions for both studied indirect bandgap two-period structures, increase of separating layer thickness $d_{QD1,QD2}$ can change their energy structure. It is known [11], that increase of thickness of top barrier layer leads to relaxation of elastic strains in the QDs layer, and correspondingly, to decrease of energy of QD quantum states, and to shift of the PL line towards low-energy side. Indeed, X-ray studies have shown that for the sample with higher thickness of separating Al(Ga)As layer elastic strains of layers an order of magnitude less than for the sample with a thin layer. It follows that, the difference in the energy position of QD1 band in PL spectra of indirect bandgap InAs QDs (ΔE ≈ 20 meV, Fig. 2) is caused by the differences in the elastic strains in the layer of two investigated structures.