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LOW-DIMENSIONAL SYSTEMS

Optical and Structural Properties of InAs Quantum Dot Arrays Grown in an $In_xGa_{1-x}As$ Matrix on a GaAs Substrate

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Abstract—Structural and optical properties of InAs quantum dots (QDs) deposited on the surface of a thick InGaAs metamorphic layer grown on a GaAs substrate have been studied. The density and lateral size of QDs are shown to increase in comparison with the case of QDs grown directly on a GaAs substrate. The rise of In content in the InGaAs layer results in the red shift of the photoluminescence (PL) line, so that with 30 at % indium in the metamorphic layer the PL peak lies at 1.55 μm. The PL excitation spectroscopy of the electronic spectrum of QDs has shown that the energy separation between the sublevels of carriers in QDs decreases as the In content in the InGaAs matrix increases. © *2004 MAIK "Nauka/Interperiodica".*

1. INTRODUCTION

The effect of the substrate material on the details of the formation of quantum dots (QDs) in the Stranski–Krastanow growth process is one of the most interesting and important problems in modern semiconductor technology. Despite significant progress in experimental [1] and theoretical [2] studies of the physics of self-organized formation of QDs, the dependences of density, lateral size, and height and shape of QDs on the lattice constant and surface energy of the substrate material are not clearly understood. Experimental study of these processes is hindered by the fact that only certain types of substrates are available. For example, the use of InGaAs substrates to grow structures in the InGaAs–InAlGaAs system has not become widespread. Furthermore, systematic experiments of this kind would require InGaAs substrates with a varied In content.

We recently demonstrated the possibility of growing thick InGaAs layers with quite high crystalline and optical quality on GaAs substrates [3–6]. To ensure the transition from the lattice constant of GaAs to that of the $In_xGa_{1-x}As$ layer, a metamorphic $In_xGa_{1-x}As$ buffer layer was deposited between them. In the growth of the $In_xGa_{1-x}As$ buffer, a special technique that reduces the density of dislocations was used [7], which provided a sufficiently low density of dislocations penetrating into the optically active region and high planarity of all the heterointerfaces. Thus, the stress-free $In_xGa_{1-x}As$ layer plays the role of the substrate material for further growth, and the In content in this layer can be varied in a wide range. This technique was used in [4], and 1.52 μm lasing was obtained in a laser based on QDs that were grown on a thick metamorphic $In_{0.2}Ga_{0.8}As$ layer on a GaAs substrate. We now report on a detailed study of the effect of the composition of an $In_xGa_{1-x}As$ ($0 \le x \le 0.30$) layer on which InAs QDs are grown on the density, size, shape, and energy spectrum of the QDs.

2. EXPERIMENTAL

The structures were MBE-grown on (100) *n*-GaAs substrates in a Riber 32P system with a solid-state As source. First, the intermediate 0.5-μm-thick $In_xGa_{1-x}As$ ($x = 0$–0.30) buffer layer intended for ensuring the transition from GaAs to InGaAs lattice constant was grown at a temperature of 400°C. The growth temperature was then raised to 500°C, and the active region of the structure was grown; it was a 0.2-μm-thick $In_xGa_{1-x}As$ layer (hereinafter the matrix) confined on both sides by $In_{0.2}Ga_{0.8}As/In_{0.2}Al_{0.8}As$ superlattices to prevent the leakage of carriers to the surface and the buffer layer. QDs were located in the middle of the $In_xGa_{1-x}As$ layer; their growth sequence was as follows. First, seed islands were formed by depositing 2.6 monolayers (ML) of InAs; then they were overgrown with an $In_yGa_{1-y}As$ ($y = x + 0.2$) layer with an effective thickness of 5 nm.

A study by transmission electron microscopy (TEM) in the diffraction-contrast mode was performed on a Philips CM microscope with a 100-kV acceleration voltage. Dark-field images of a structure were obtained in (200) reflection directed in parallel to the growth surface.

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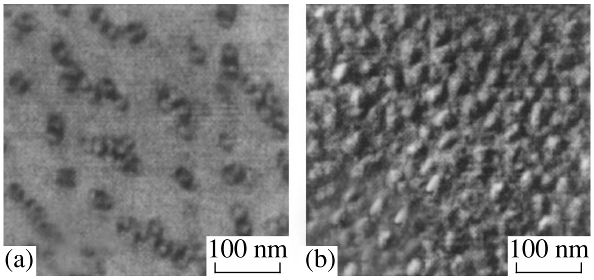

Fig. 1. Planar TEM images of QDs for structures with (a) GaAs and (b) $In_{0.2}Ga_{0.8}As$ matrices.

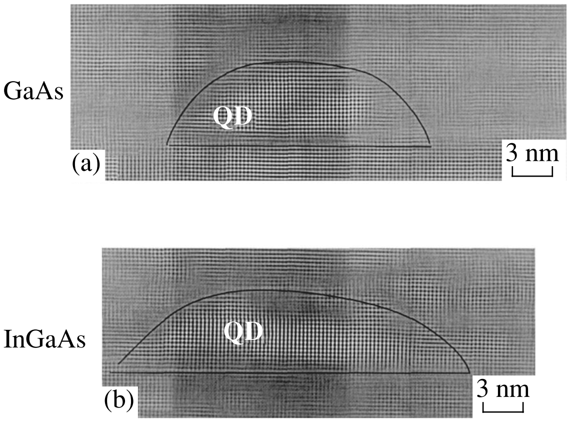

Fig. 2. Cross-sectional HRTEM images of QDs for structures with (a) GaAs and (b) $In_{0.2}Ga_{0.8}As$ matrices.

A JEM 4010 microscope with a 400-kV acceleration voltage was used for the high-resolution transmission electron microscopy (HRTEM). The Fourier filtration was applied in the analysis of the images obtained. In the reconstruction of images, (200) reflections and the transmitted beam were used.

Photoluminescence (PL) of the structures was excited by an Ar-ion laser ($W = 1500$ W/cm^2, $\lambda = 514$ nm) and studied in the temperature range 10–300 K. The PL excitation spectra were recorded under excitation from the light of a halogen incandescent lamp that was passed through a monochromator. The signal was detected using a monochromator and a cooled Ge diode.

3. RESULTS AND DISCUSSION

Figures 1a and 1b show TEM images of QD arrays grown on GaAs and $In_{0.2}Ga_{0.8}As$ surfaces, respectively. As can be seen from these figures, the surface density of QDs is significantly higher in the case of deposition on an $In_{0.2}Ga_{0.8}As$ layer than on GaAs. This increase in the QD density with a constant scatter in size is desirable and opens the way to attaining a higher gain on the ground state in QD lasers [8].

HRTEM data (Figs. 2a, 2b) showed that the growth of QDs on a metamorphic $In_xGa_{1-x}As$ layer also causes an increase in the lateral size of islands with the retention of their height. The lateral size is 19 nm for GaAs and 26 nm for the $In_{0.2}Ga_{0.8}As$ matrix (Figs. 2a and 2b, respectively). Note that RHEED patterns, which were recorded during the growth of the structure, indicate that the formation of QDs (the transition to 3D growth) occurs after the deposition of InAs with an effective thickness of 1.7 ML on a GaAs matrix and 1.5 ML for the case of $In_{0.2}Ga_{0.8}As$. Thus, in the latter case the formation of QDs consumes more material. One may assume that the increase in the density of QDs and their lateral size can be caused both by the activated decomposition of $In_xGa_{1-x}As$ with a high InAs content ($x = 0.4$) at the overgrowth stage and by possible segregation of In on the surface of the $In_xGa_{1-x}As$ buffer layer.

When the material of the matrix is changed from GaAs to $In_xGa_{1-x}As$, the PL line corresponding to the emission associated with the QD ground state (asterisked in the spectra) is red-shifted (Fig. 3). With a 30% InAs content in the matrix material, the PL peak lies at 1.55 μm. This shift of QD PL to lower energies may be caused by several factors. First, the decrease in compressive stress in QDs when the matrix material is changed lowers the energy of the bottom of the conduction band. Second, the increase in the lateral size of QDs also brings down the quantization levels. Furthermore, when the InAs content in the matrix material increases, the band gap of the matrix decreases, and, consequently, the penetration of carrier wave functions into the potential barrier is diminished; thus, the carrier levels in QDs are reduced. At the stage of QD overgrowth, the presence of a stressed $In_xGa_{1-x}As$ layer with large x (tensile-strained in the vertical direction and with lower elasticity) also depresses stress fields in InAs QDs and brings down the levels. All these effects can contribute to the red shift of the PL line [9].

When the InAs content in the matrix is less than 20%, no decrease in the integral efficiency of PL was observed up to high excitation densities, which is consistent with the localization of penetrating dislocations in the buffer layer. When the InAs content in the matrix is raised to over 25%, the PL intensity decreases; this is related to the partial penetration of residual dislocations from the buffer to the upper layers and to the formation of dislocated QDs in the overgrowth of initial islands with an InGaAs layer with a rather high (>45%) In content [10, 11]. Both effects are observed in TEM images of these structures.

The PL spectrum of a sample with QDs in an $In_{0.2}Ga_{0.8}As$ matrix recorded at high excitation density (Fig. 4, dashed line) demonstrates lines corresponding to ground and excited states, as well as the line of PL from a quantum well (QW) formed by the wetting layer and the $In_{0.4}Ga_{0.6}As$ layer used for overgrowth of the seed islands. The peak at 1.185 eV corresponds to the emission from the $In_{0.2}Ga_{0.8}As$ matrix. Owing to inhomogeneous broadening of the ensemble of QDs, it is difficult to determine precisely the peak energies in the

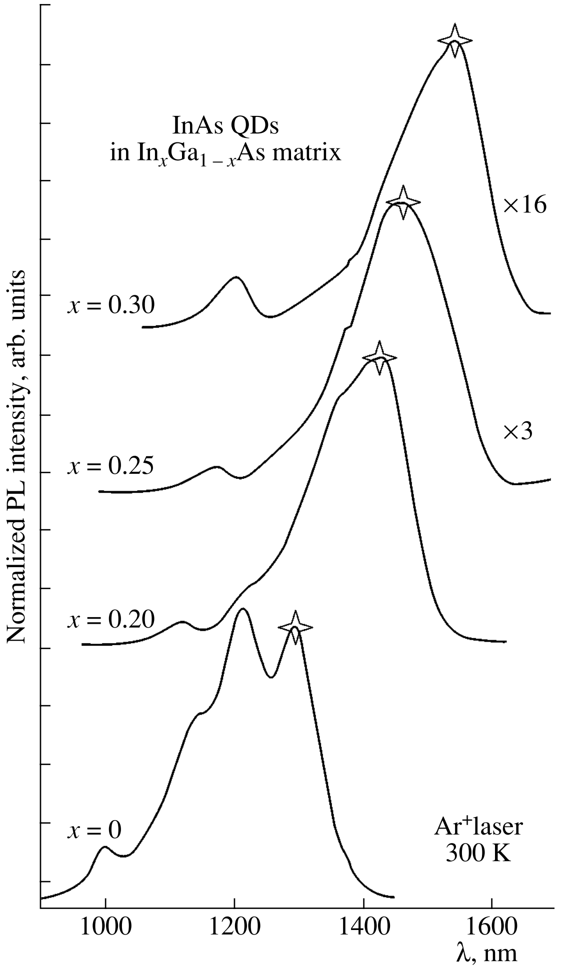

Fig. 3. Normalized PL spectra of InAs QDs deposited on $In_xGa_{1-x}As$ layers with different InAs contents (x).

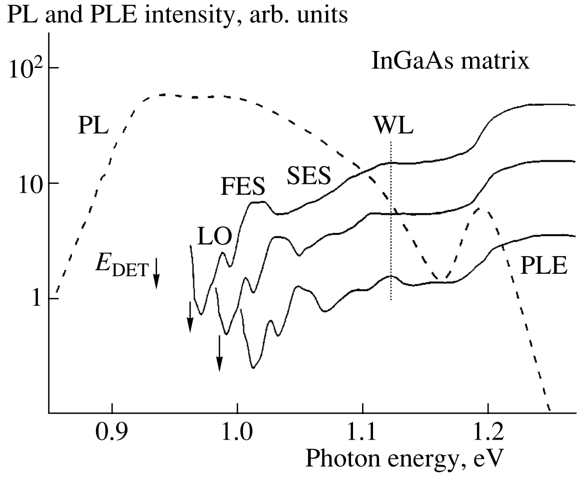

Fig. 4. PL spectrum of InAs QDs deposited on an $In_xGa_{1-x}As$ layer, recorded under excitation with an Ar laser (1.5 kW/cm^2) (dashed line). PL excitation spectra recorded at the detection energies E_{DET} = 0.94, 0.96, and 0.985 eV (solid line).

PL spectrum. A more detailed determination of the energies of optical transitions is possible when luminescence excitation spectroscopy is used, which makes it possible to study the relaxation and recombination processes in QDs with a definite energy of the ground state (to be more precise, in the ensemble of dots for which the ground state energy lies within the spectral resolution of the optical system, typically several millielectronvolts). Since lateral transport between QDs is absent at low temperatures, the PL excitation spectra recorded along the profile of a PL line contain information on the energy spectrum of QDs with different energies of the ground state, i.e. for QDs of different size.

Figure 4 shows PL excitation spectra recorded at the temperature T = 10 K at different detection energies E_{DET} = 0.94, 0.96, and 0.985 eV (marked with arrows). These excitation spectra exhibit peaks corresponding to absorption in the InGaAs matrix and in the QW formed by the wetting layer and covering the $In_yGa_{1-y}As$ layer (Fig. 4, WL) and the absorption associated with the excited states (Fig. 4, SES, FES). The complex structure of the peaks of the excited states can be explained based on the following considerations. As was shown theoretically in [12], several electron and hole levels exist in pyramidal-shaped InAs–GaAs QDs that are quite large. It is well known also that the degeneracy of levels in pyramidal QDs with a square base is completely removed by the Coulomb interaction and piezoelectric effect [12]. Therefore, a great number of levels appear in the QD energy spectrum, although the probabilities of optical transitions between some of these can be quite low (partially forbidden transitions). As a result, optical transitions with similar energies in the ensemble of QDs can behave as a single "effective transition," and the absorption line of this "effective excited state" will be a superposition of inhomogeneously broadened lines of transitions involving the excited states with similar energies.

The peaks of separate transitions in PL excitation spectra, which lie within the limits of the "second effective excited state," are resolved much worse. This may be related to the great number of these closely spaced transitions and to their stronger inhomogeneous broadening. Moreover, because of the possible fluctuations of the QD shape, several excited states may, in general, correspond to a single ground state.

Note that carrier relaxation processes must be considered in the interpretation of peaks in the PL excitation spectra [13]. The high relative intensity of peaks associated with certain excited states is caused by the fact that the energy difference between these states and the ground state is a multiple of the energy of longitudinal optical (LO) phonons, which facilitates the relaxation of carriers from these excited states [14]. In the case under study, the PL excitation spectrum also shows a peak related to the phonon relaxation; this peak is located 35 meV from the detection energy and corresponds to the LO phonon energy in the GaAs–InAs system.

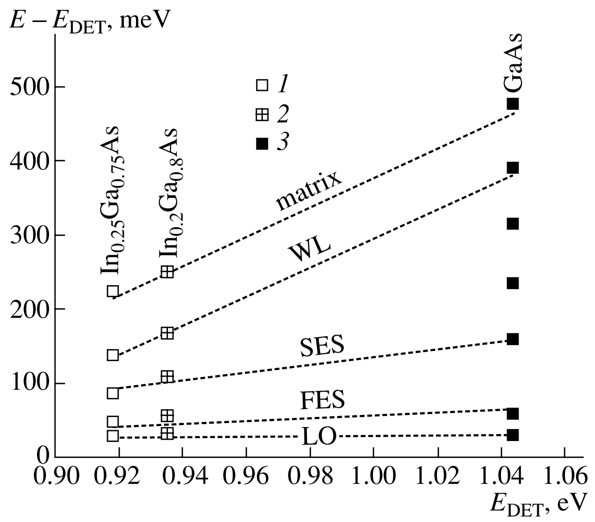

Fig. 5. Energies of peaks in the PL excitation spectra for QDs in (*1*) $In_{0.25}Ga_{0.8}As$, (*2*) $In_{0.2}Ga_{0.8}As$, and (*3*) GaAs matrices. For convenience, the detection energy E_{DET} is subtracted.

Figure 5 shows the energies of peaks in the PL excitation spectrum of QDs in $In_{0.25}Ga_{0.75}As$, $In_{0.2}Ga_{0.8}As$, and GaAs matrices; for convenience they are shifted by the detection energy. In all cases, the detection of the PL excitation spectra was performed at the energy of the PL peak corresponding to transitions from the QD ground state. It can be seen that, in the case of QDs deposited on InGaAs layers, the increase in the In content causes a decrease in the energy of carrier localization, defined as the difference between the matrix band gap and the energy of optical transition from the QD ground state. The energy separation between the energies of excited states and the ground state also decreases. This fact can be attributed to a decrease in the energy separation between the sublevels of electrons (holes) in a QD as its lateral size increases, while the number of sublevels seems to increase [15].

4. CONCLUSION

Structural and optical properties of InAs QDs in an $In_xGa_{1-x}As$ ($0 \le x \le 0.3$) matrix have been studied. The dots were formed on the surface of a thick $In_xGa_{1-x}As$ metamorphic layer grown on a GaAs substrate. The density and lateral size of QDs is higher when they are grown on an InGaAs metamorphic layer than when they are grown on GaAs. The height of dots remains approximately constant. The peak wavelength in the QD emission spectrum depends on the composition of the matrix material; it can be varied in a controlled manner from 1.3 μm (at $x = 0$) to 1.55 μm ($x = 0.3$), and a high efficiency of luminescence is retained. The energy spectrum of QDs was studied by means of PL excitation spectroscopy. When the InAs content in the matrix is increased, the localization energy of QDs (the difference between the energy of QD ground state and the matrix band gap) decreases, and the energy separation between the ground and excited states also decreases.

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