Radiation Anisotropy and Ordering Effects Inherent to Quantum Dots and Wires in (In,Ga)As/GaAs Nanostructures

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Abstract

Atomic force microscopy and polarised luminescence is used for studying interrelations between the surface morphology and the optical properties of multilayer (In,Ga)As/GaAs(100) nanostructures grown with molecular-beam epitaxy technique, which possess self-organised quantum dots and quantum wires. With increasing number of periods in the structure, aligning of the quantum dots in rows parallel to the crystal direction $[01\bar{1}]$ is observed. The improvement of lateral ordering correlates with increasing radiation anisotropy of the structure. A possible ordering mechanism is discussed.

Key words: quantum dot, quantum wire, polarization photoluminescence, AFM.

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I. Introduction

Self-organised growth of quantum dots (QDs) and quantum wires (QWRs) owing to Stranski-Krastanov mechanism, occurring in molecular-beam epitaxial heterostructures with strains, is one of the most promising nanotechnological methods. Prospects of applications of these systems in optoelectronics have caused a permanent interest in them. When one deals with the practical applications, it is rather important to obtain uniform sizes and shapes of QDs and QWRs, as well as their ordered arrangement. If the thickness of spacers in these multilayer structures is small, then a vertically correlated QD arrangement (i.e., one under another) takes place in all the layers [1,2]. In this case, the QD size dispersion is decreased, whereas the mean QD size is somewhat increased. At the same time, the lateral arrangement of QDs in each layer still remains an actual problem.

Recently we have shown that employing a special technology for growing the multilayer (In,Ga)As/GaAs(100) structures enables one to achieve a tendency to aligning these QDs in rows [3]. In that work, the laterally ordered multilayer nanostructures have been studied, using the methods of atomic force microscopy (AFM) and polarised photoluminescence (PL).

Polarization properties of QD radiation have been studied relatively seldom. Some results have been obtained earlier with the methods of cathodoluminescence [4], excitation spectra [5] and PL [6]. The radiation anisotropy has been associated with a geometrical anisotropy of the QD shape. In particular, the PL polarization of InAs QDs, grown on the GaAs (311) surface, has been found to be in a good agreement with the lengthened shape of QDs positioned along $[\bar{2}33]$ direction [7].

In the present paper, we represent the results of investigations aimed at finding effects

of period number, GaAs spacer layer thickness and $In_xGa_{1-x}As$ layer composition on both the surface morphology and the optical anisotropy of self-organised, laterally ordered arrays of QDs and QWRs.

II. Experimental

All the structures were grown, using the method of molecular-beam epitaxy on GaAs substrates with (100) orientation. The nominal In concentration in $In_xGa_{1-x}As$ layers was $x = 0.30 \div 0.50$. The number of $In_xGa_{1-x}As$/GaAs periods for various samples varied from 2 to 17. The thickness of the layers, their composition and period number are summarized in Table. The growth regimes have been earlier described in [3].

Transition from the pseudomorphic 2D growth regime to creation of 3D islands was controlled using the high-energy electron diffraction. Surface morphology of the samples was studied with the scanning atomic force microscope Nanoscop IIIa. Photoluminescence of the structures was excited at low temperatures close to 8 K, using the second harmonics of Nd:YVO_4 laser radiation (the wavelength of 532 nm). The spectra were detected with the aid of spectrometer TRIAX-550 and a special system based on the cooled CCD camera (OMA V:512-1.7). Finally, the PL polarization was analysed using the Glan-Thomson polarizer.

III. Results and discussion

Shown in Fig. 1 (left) are the AFM images of the upper uncovered layer for the structures with 2, 7 and 9 $In_{0.5}Ga_{0.5}As$(9.8 ML)/GaAs(60ML) periods. The averaged sizes of dots and their surface densities determined with AFM are summarized in Table.

The two following features seem to be worth noticing. First, the lateral QD sizes along the directions $[0\bar{1}1]$ and [011] differ a little. In the first direction, these are slightly larger, i.e., the QDs possess somewhat lengthened shape along the $[0\bar{1}1]$ direction. Second, the covering of the surface with QDs is rather dense (up to 30%). Characterising it with the ratio *w/d* (with *w* being the diameter of QDs and *d* the mean distance between them), one can see this ratio to exceed 0.5. In case of 9-period sample, it reaches 0.71. As a consequence, one can expect an effective interaction between the adjacent QDs [8] and a related correlation in their mutual arrangements.

Table. Data of AFM and Fourier analysis for the multilayer $In_xGa_{1-x}As$/GaAs structures.

Technological parameters of sample				Direction	Parameters of QDs (QWRs) obtained with AFM				Fourier analysis results for AFM data		w/d
$In_xGa_{1-x}As$		GaAs	Number of periods		Lateral size, w (nm)	Density (μm^{-2})	Height, (nm)	Coverage of surface, %	Halfwidth of Fourier maximum, (μm^{-1})	Average distance between QDs (QWRs), d (nm)	
x	Thickness (ML)										
0.5	9.8	60	2	[011]	41.10	300	5	33	8.90	74.67	0.55
0.5	9.8	60	7	[011]	50.50	286	8	29	8.20	77.41	0.65
0.5	9.8	60	9	[011]	55.50	133	11	*25*	6.93	78.13	0.71
0.4	7.6	67	17	[011]	48.20	190	7	13	2.72	110.65	0.44
				$[0\bar{1}1]$	49.70				4.10	51.11	0.97
0.3	11.5	67	17	[011]	77.20	9	7	38	2.31	101.16	0.76
0.3	16.3	67	17	[011]	78.04	11	8	43	2.98	89.7	0.87

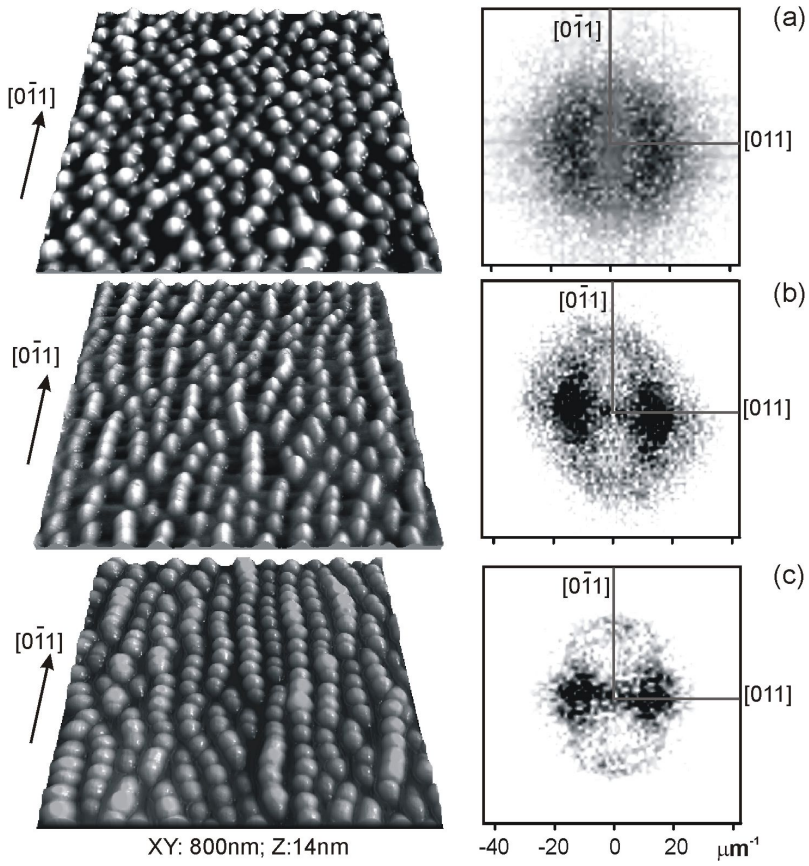

Fig. 1. 3D AFM images of surface fragments for the multilayer $In_{0.5}Ga_{0.5}As/GaAs$ structures (left) and their 2D Fourier transforms (right): (a) 2, (b) 7 and (c) 9 periods.

The effect of QD ordering occurring under increasing period number has been analysed using the Fourier analysis of AFM images for the squares with 5×5 μm^2 dimensions. The results are depicted in Fig. 1 (right side). The fact that the 2-period sample exhibits the peaks, which are blurred substantially stronger in the direction $[0\bar{1}1]$ than in [011] one, indicates that a weak tendency to periodic distribution of QDs along [011] direction is evident even for this sample. With increasing period number, a higher sharpness of the peaks localised in [011] direction (see Fig. 1, b and c) reflects the improvement of periodicity in the QD disposition along this direction. The AFM images are also indicative of the fact that it is related to the tendency of QDs to align in chains along $[0\bar{1}1]$ direction. The latter effect cannot be explained using the known data for anisotropy inherent to elastic constants of the separating GaAs spacer layer that is covered with the next layer of growing InGaAs QDs. The matter is that the minima of the elastic tensor describing GaAs(100) layer determine the minima of elastic energy, related to incompatibility of deformations between the layers, which predict possible ordering along <100> directions. Therefore, some different factors should be considered as controlling the mentioned effect:

First, the fields of elastic strains from the QD boundaries decay considerably faster in elastically "hard" <011> directions, when compare to the «soft» <100> ones. It can promote more intensive mass transfer along <011> directions.

Second, the dominant role can be played by the difference in surface diffusion flows forming these QDs. The lengths of the surface diffusion for cation adatoms along $[0\bar{1}1]$ direction can exceed those referred to [011] by an order of magnitude [9].

And finally, third, a lateral superposition of elastic strain fields of the adjacent barrier QDs

can be decisive in such a multilayer structure with a high density of QDs, which can enhance ordering of the spatial QD distribution in the next layer.

Our further investigations have shown that the improvement of the lateral structure ordering correlates with the growth of anisotropy in InGaAs QD radiation. Fig. 2 (a and b) depicts the PL spectra of 2- and 11-period structures for the two polarizations parallel to $[0\bar{1}1]$ and [011] directions. The PL is excited by the radiation with the energy E_{exc}=2.33 eV and is usually associated with exciton recombination in (In,Ga)As QDs.

Though the exciting radiation is linearly polarised, the PL polarization in our case does not depend on the polarization of excitant radiation. It is associated with the fact that, under the excitation with the quantum energy of 2.33 eV, which exceeds considerably the band gap in GaAs, photogenerated carriers thermalise at the quantum levels of QD and lose a polarization "memory".

It is seen that the polarization degree for the PL band peak increases from P≈5% up to P≈8%, when passing from 2 - to 11-period structure. In the latter case, the P value grows at the low-energy side of the emission band up to P≈17%. This fact can be understood while taking into account that the above-mentioned anisotropy of the QD shape becomes larger, when going from the lower structure layers to the upper ones. However, as far as the averaged QD sizes are larger for the upper layers than those for the lower ones, this fact results in the low-energy asymmetry of the PL band.

Fig. 2c represents the dependence of the PL band intensity on the azimuth of linear polarization with respect to [011] axis. This dependence is well approximated by the relation

$$I = I_{[0-11]} \sin^2\theta + I_{[011]} \cos^2\theta . \qquad (1)$$

The ratio $I_{[0\bar{1}1]}$ / $I_{[011]}$ is equal to 1.13 and 1.20 for 2- and 11-period structures, respectively.

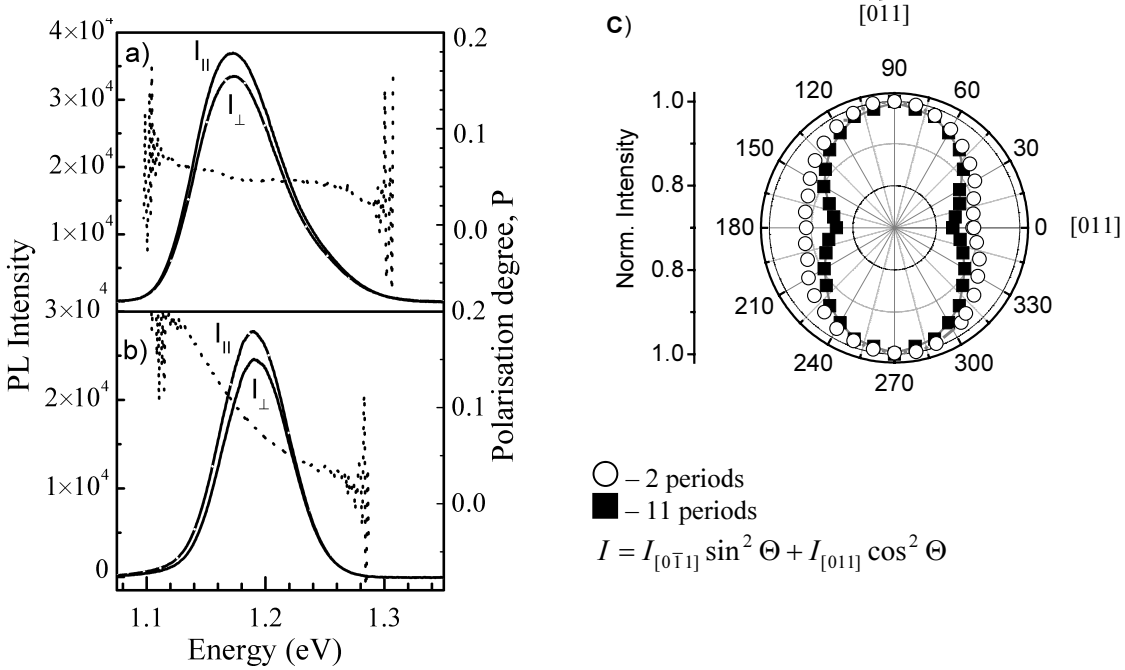

Fig. 2. PL spectra for 2-period (a) and 11-period (b) $In_{0.5}Ga_{0.5}As/GaAs$ structures. The emission is linearly polarised parallel to $[0\bar{1}1]$ (I_{II}) and [011] ($I_{\perp}$). The dotted line indicates the linear polarization degree $P = (I_{II} - I_{\perp})/(I_{II} + I_{\perp})$; (c) angular distribution of linearly polarised light intensity. E_{exc} = 2.33 eV, T = 8 K and P_{exc} = 0.1 W/cm^2.

In order to study profoundly the mechanism of self-organisation of QD chains and anisotropy effects, we have grown 17-period structures with various thickness of the $In_xGa_{1-x}As$ layer and different In concentrations in it. In doing so, we have kept the thickness of the separating spacer GaAs layer the same (67 ML) as in all the above cases (see Table).

2D Fourier transforms of the AFM images peculiar for these 17-period structures are shown in Fig. 3. It is seen that, for x=0.4 and d(InGaAs) = 7.6 ML, the surface morphology of the sample is an array of periodic QD chains (Fig. 3a). Two peaks at the corresponding Fourier transform are indicative of an essential short-range correlation in the mutual QD arrangement along [011] direction (at least, up to the second neighbour). Blurred peaks for $[0\bar{1}1]$ correspond to weaker periodicity for this direction. The mean distance between the chains is 110 nm along [011] direction, and it is close to 51 nm between the QDs along the chain in $[0\bar{1}1]$ direction (see Table).

The situation is cardinally changed when the In concentration goes down to x = 0.30, and the nominal thickness of InGaAs layer increases up to 11.5 and 16.3 ML (see Fig. 3, b and c). In this case, we observe formation of the array consisting of QWRs laterally ordered along $[0\bar{1}1]$ direction. As one can assume, this qualitative difference in the surface morphology might be associated with decreasing In concentration. The latter would result in decrease of elastic energy of the system, which, in turn, contributes to growth of the critical thickness for surface morphological instability, when 3D islands are created due to 2D-3D transition. At the same time, a considerable difference between the adatom mobilities along $[0\bar{1}1]$ and [011] directions remains an essential factor in providing anisotropy of the mass transfer, as well as resulting the strain anisotropy.

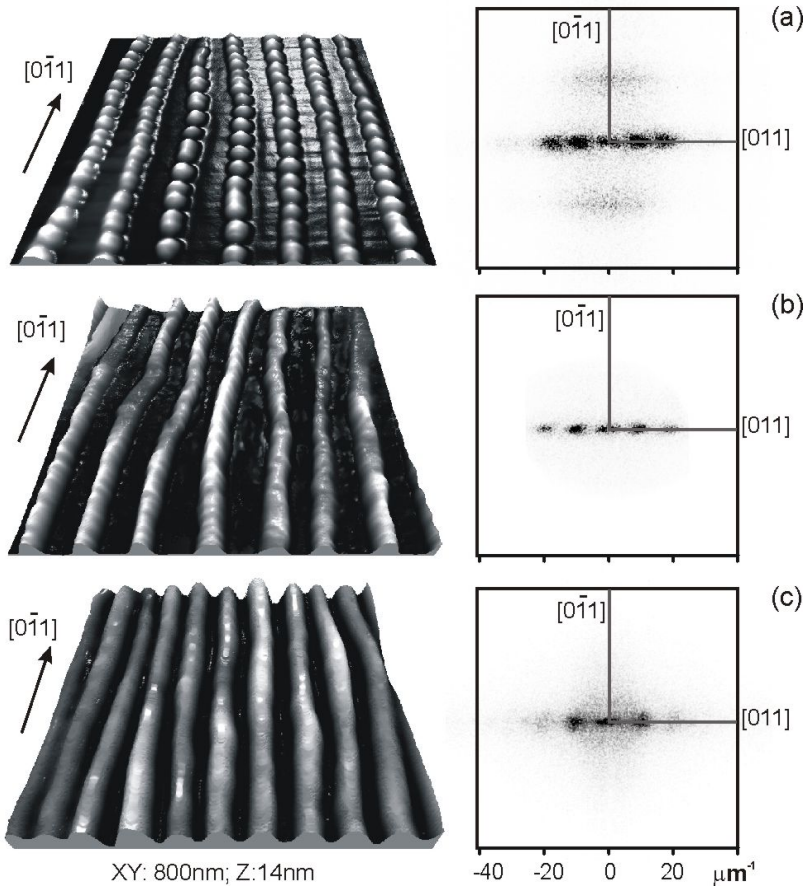

Fig. 3. 3D AFM images of surface fragments for 17-period $In_xGa_{1-x}As/GaAs$ structures (left) and their 2D Fourier transforms (right): (a) x = 0.40, d_{InGaAs} = 7.6 ML; (b) x = 0.30, d_{InGaAs}=11.5 ML, (c) x = 0.30, d_{InGaAs} = 16.3 ML.

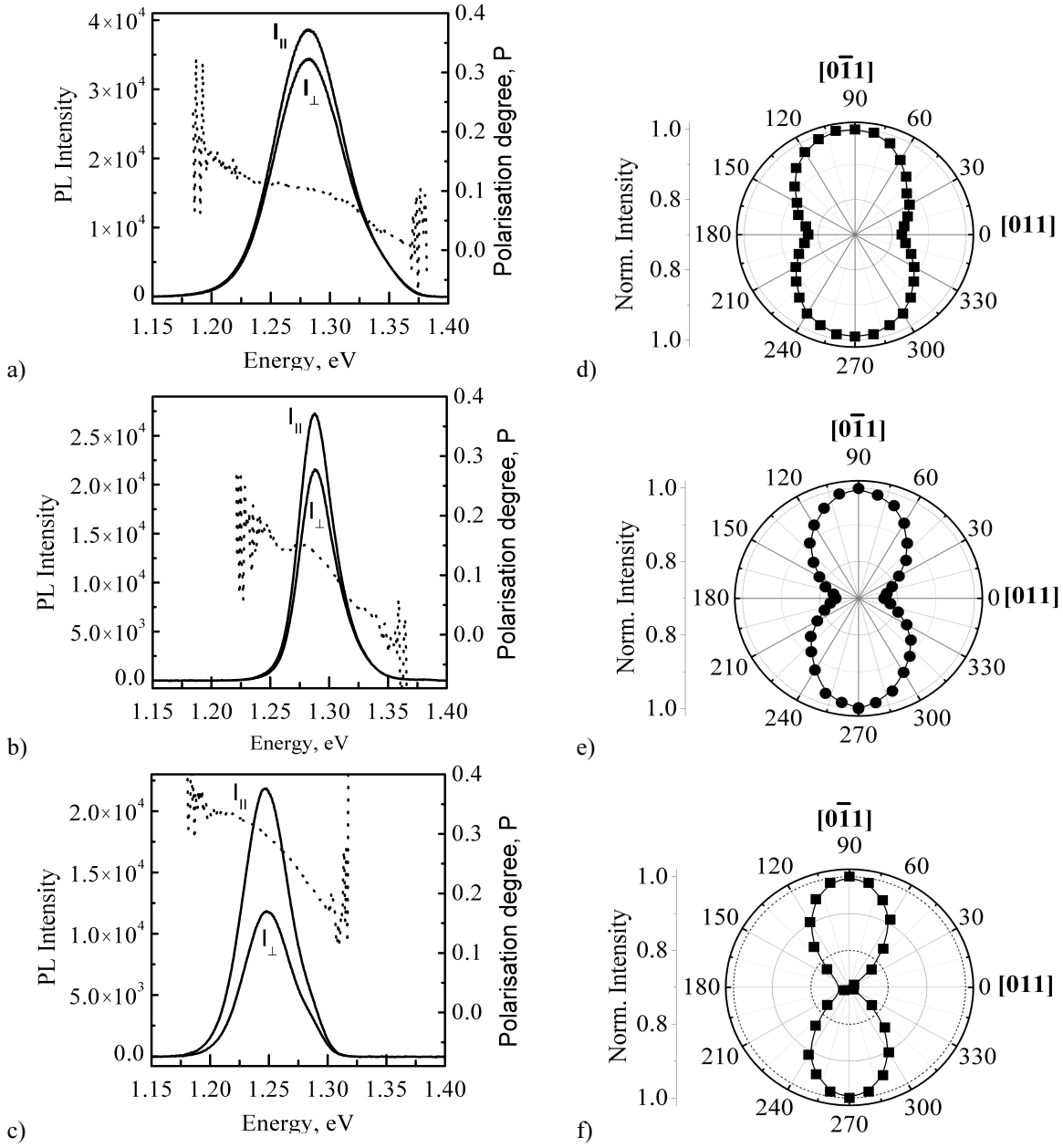

Fig. 4. PL spectra of 17-period $In_xGa_{1-x}As/GaAs$ structure for x = 0.40 (a), x = 0.30 (b), (c) and the nominal thickness 7.6 ML (a), 11.5 ML (b) and 16.3 ML (c). The emission is linearly polarised along $[0\bar{1}1]$ (I_{II}) and [011] ($I_{\perp}$), respectively. The dotted lines in (a), (b) and (c) indicate the degree of linear polarization and the solid ones in (d), (e) and (f) show the angular dependence of the approximated emission intensity $I = I_{[0\bar{1}1]}\sin^2\Theta + I_{[011]}\cos^2\Theta$. E_{exc} = 2.33 eV, T = 8 K and P_{exc} = 0.1 W/cm^2.

It seems reasonable that the structure with QWRs is also characterised by more pronounced anisotropy observed in the measurements of luminescence polarization. The relevant results are depicted in Fig. 4. The polarization degree for the emission peak of the QD structure (Fig. 4a) is close to 10%, but it increases up to 14% (Fig. 4b) and 30 % (Fig. 4c) for the structures with QWRs. The angular dependence of the polarised emission intensity (see Fig. 4, d, e and f) is well approximated by Eq. (1) and the relation $I_{[0\bar{1}1]}/I_{[011]}$ is equal to 1.21 for the structures with QD chains (Fig. 3a) and 1.32 and 1.65 for the two structures with QWRs (see Fig. 3, b and c).

IV. Conclusions

Summarising the results obtained, one can conclude that, when using definite growth conditions, it is possible to provide ordering QDs in the heteroboundary plane already under deposition of the first periods of the multilayer (InGa)As/GaAs structure. Lateral ordering and size uniformity characteristic of QDs and QWRs are improved with increasing period number. Moreover, the changes in both the thickness of the separating layer and the In concentration enable one to provide creation of uniform periodic chains of QDs and QWRs. The process of lateral ordering of these QDs and QWRs in multilayer structures is determined by competing effects of anisotropy in elastic properties of the separating layer, anisotropy of surface adatom diffusion and elastic interaction between the QDs. Optical anisotropy of the QD and QWR emission is determined by the effects of anisotropic relaxation of elastic strains, together with the shape of the nanoobjects.

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