Control of density, size and size uniformity of MBE-grown InAs quantum dots by means of substrate misorientation

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Introduction

Realization of the unique electronic properties of semiconductor quantum dots (QDs), resulting from the $\delta$-like energy dependence of the density of electron states in QDs, requires creation of dense arrays of uniform QDs [1]. One of the most promising methods for the preparing such arrays is molecular beam epitaxy (MBE) in Stranski–Krastanov growth mode [2]. Numerous investigations of InAs QDs have shown that their density and size depend on the growth conditions, and QDs arrays in the density range $10^8–10^{11}$ cm$^{-2}$ can be obtained [3, 4]. The formation of InAs QD arrays with a density greater than $3 \times 10^{10}$ cm$^{-2}$ on a correctly oriented GaAs(001) surface is, as a rule, accompanied by coalescence of a noticeable number of QD into large islands, which leads to radical broadening of the QD size distribution [4]. So, independent control of sizes and density of QDs is one of the main problems to be solved in creating QD heterostructures suitable for device application.

We propose that dense arrays of uniform QDs should be created using the conventional MBE growth in Stranski–Krastanov growth mode on a vicinal GaAs(001) substrate misoriented in the [010] direction. On such vicinal surfaces there appears a net of small terraces, bordered by steps from all the sides. The Schwoebel potential barrier at the step edges can considerably suppress the surface diffusion of adatoms between the terraces [5], and the QDs growth on each terrace will occur primarily from the material deposited on the same terrace.

1 Theory

We have performed a Monte–Carlo simulation of terrace formation on vicinal GaAs(001) surfaces misoriented in the [010] direction. We took into account the processes of atom adsorption and desorption on the surface, adatom migration, adatom attachment to, and detachment from a step. The elementary detachment
processes for different kinds of step configuration are shown in Fig. 1(a). A Monte–Carlo simulation of GaAs growth shows that the processes \( n_3 \), \( n_4 \) and \( n_7 \) dominate over the other elementary detachment processes (\( n_1 \)–\( n_{10} \)). The shape of the terraces strongly depends on the atom detachment activation energies (\( E_{nx} \)). The activation energy ratio \( E_{n3}/E_{n4} \) controls the ordering of waves. Parameter \( E_{n7} \) controls the abruptness of the wave. With decreasing \( E_{n7} \), terraces become wave-shaped and connected by gates (see Fig. 1(b)).

In order to reveal the possible role of adatom diffusion between the connected terraces, we estimated the coalescence rate of QDs located on neighbouring terraces. We simulated the motion of adatoms by the Monte–Carlo method and determined the number of adatoms which started to move from the edge of a QD and reached the edge of a neighbouring QD in unit time. The simulation showed that making the constriction stronger linearly retards the coalescence rate.

2 **Technological approach**

Experimentally the effect of substrate misorientation on QDs formation was investigated by atomic force microscopy, photoluminescence and electroluminescence.
The heterostructures used in all kinds of measurements were grown under identical growth conditions. Each growth process included two stages. In first stage terrace net was prepared during GaAs growth in step flow growth mode and in the second—InAs QDs were grown in Stransky–Krastanow growth mode. The effective thickness of InAs coating was 3 monolayers. Each epitaxy process was performed simultaneously on GaAs(001) surfaces misoriented in the [010] direction by 0, 1, 2, 4 and 6°. The comparatively small range of misorientation angle variation was chosen so that the surfaces would differ from one another only in growth-step density without fundamental variation in the surface morphology caused by approach to new singular faces.

3 Results and discussion

When studying the surface structure of GaAs samples by AFM, we revealed on the vicinal surfaces a net of wave-like terraces joined by gates as predicted by the

Fig. 2. AFM image of GaAs surfaces (upper row) and InAs QDs (3 ML InAs coverage thickness) (lower row) MBE-grown on GaAs(001) surfaces exactly oriented and misoriented by 6° in the [010] directions.
Monte–Carlo simulation (Fig. 2, upper row). Increasing the misorientation angle makes higher the terrace density and reduces their mean width from 500 Å for 1° to 250 Å for 6°. These values are considerably larger than the dimensions of the terraces calculated under assumption that the step is one monolayer high. The observed difference can be explained by the phenomenon of step-bunching, which in our case becomes more pronounced with the misorientation angle increasing from 2–3 ML for 1° to roughly 10 ML for 6°. At InAs coverage of 3 ML (Fig. 2, lower row), InAs QDs arrays were found on the terraced surfaces. Also present on the surfaces were large InAs islands formed as a result of coalescence of InAs QDs. Their number falls dramatically with increasing misorientation angle, and on the surface with 6° misorientation there are scarcely any large InAs islands. The terrace density increasing with the misorientation angle leads to higher density of QDs (Fig. 3), decrease in their mean height from 34 Å for 0° to 20 Å for 6° and to more uniform size distribution. The similar variation of the spreads in QD height and terrace width with surface misorientation led us to conclude that the QD size dispersion is controlled by the terrace area distribution.

For PL study two sets of samples were prepared. They differed only in the interruption time ($T_{\text{int}}$) between the end of QD growth and start of GaAs layer overgrowth. For the first set $T_{\text{int}} = 15$ min, and for the second, $T_{\text{int}} = 10$ s. In the grown heterostructures a single-sheet array of InAs QDs was confined between GaAs barriers (200 Å) surrounded by the AlAs/GaAs superlatticies and cladding Al$_0.7$Ga$_0.3$As layers. PL spectra ($T = 77$ K, $\lambda = 514.5$ nm, $P = 200$ Å cm$^{-2}$) taken for both sets of samples are presented in Figs. 4(a) and 4(b). The position of the PL line maximum and the full width at half maximum (FWHM) of the PL line...
are shown in Fig. 5. versus the misorientation angle. For both series of spectra the substrate misorientation leads to a blue shift of the maximum and a decrease in the FWHM of PL lines. These effects can be naturally explained by a decrease in size and better size uniformity for InAs QDs on misoriented surfaces, revealed by AFM for open QD arrays. The observed effects depend on the growth interruption time $T_{\text{int}}$, becoming weaker in samples with longer $T_{\text{int}}$. It is natural to relate the dependence of the InAs QD sizes on the interruption time to the probability of adatom surface migration. On the misoriented surfaces patterned with a dense net of terraces, the adatom surface diffusion between terraces must be much slower than on the exactly oriented surface, since adatom migration between terraces occurs mainly through the gateways connecting the terraces. The increase in the misorientation angle makes terraces and gates smaller and causes progressive suppression of the adatom migration. In this connection, the effect of the interruption time on the PL spectra must be weak for exactly oriented surfaces with fast adatom migration and must become stronger on misoriented surfaces. However, with the misorientation angle increasing further it again may become weaker, with migration completely suppressed on the time scale of the chosen $T_{\text{int}}$. Analysis of the data in Fig. 4 shows that in our studies the effect of the growth interruption time is the strongest for a sample with 2° misorientation.

So, we imply that QD formation on a misoriented surface comprises two main stages with different characteristic times. The first process is very fast. A self-
Fig. 5. Peak position (squares) and FWHM (circles) of the PL line for samples with growth interruption times $T_{int} = 15\,\text{min}$ (full marks) and $T_{int} = 10\,\text{s}$ (open marks).

assembled QD array with high density and high uniformity appears in this stage. In the second, much longer stage QDs increase in size through QD interaction. This interaction occurs via surface migration of adatoms between QDs. Changes in the QDs morphology in this second growth stage determine variations in the position and width of the QD PL lines for samples grown with longer interruption time. The misorientation creates a new, much slower time scale for this stage of QD formation because of the suppression of adatom migration between terraces.

We successfully used misoriented substrates for fabrication of laser heterostructures with a single-sheet QD array. Electroluminescence spectra exhibit the same main features that were observed in PL measurements. As predicted by the theory [6], the threshold current of the “classical” single-sheet QD laser based on these heterostructures shows very strong dependence on their FWHM. The lowest threshold current density at room temperatures ($210\,\text{A cm}^{-2}$) was observed for broad-area ($100\,\mu\text{m}$) lasers with a high-reflecting mirror coating made of $4^\circ$-misoriented substrate.

4 Conclusion

We have found that application of GaAs(001) substrates misoriented in the [010] direction in conventional MBE growth permits one to control InAs QD density and size and to improve the QD size dispersion and heterostructure quality. This possibility originates from the special surface patterning which appears on misoriented substrates and can be used for other semiconductor compounds.
References