InGaAs/GaAs hybrid quantum well-dot nanostructures: Impact of substrate orientation and recombination mechanisms

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Abstract. We study the impact of substrate orientation on growth of quantum well-dots (QWD) hybrid low dimensional structures that are intermediate in properties between quantum wells and quantum dots. QWD are formed by thickness and composition modulations of InGaAs quantum well, which localize electrons and holes. We show that in case of growth on 6 degree misoriented substrates such modulations are much more pronounced than in case of growth on exact oriented substrate. The modulations result in efficient strain relaxation, which allows vertical stacking of at least fifteen 8 ML-thick In\textsubscript{0.4}Ga\textsubscript{0.6}As QWD layers without dislocation formation. Fitting of experimental dependencies of integrated PL intensity on excitation power by using ABC model allowed us determining relative contribution of different recombination processes.

1. Introduction

Semiconductor quantum wells (QWs) and quantum dots (QDs) have been intensively studied all over the world owing to their unique fundamental properties as well as advantages for device applications \cite{1-2}. In an ideal QW, the carriers are confined in the direction perpendicular to the QW but can freely move in the QW plane. In an ideal QD, electrons and holes are strongly confined in all three spatial directions and there is no carrier exchange between individual QDs within the ensemble. However, even using modern epitaxial technologies such as MBE or MOCVD it is difficult to fabricate ideal semiconductor QWs and QDs. For instance GaAs/AlGaAs QWs usually contain monolayer thickness fluctuations that confine carriers in lateral direction and play the role of shallow QDs at helium temperatures. In case of InGaAs/GaAs QWs there are fluctuations of In composition that act as localization potential for electrons and holes. In a typical array of self-organized In(Ga)As/Al(Ga)As QDs there is temperature escape of electrons and holes from QDs to wetting layer or GaAs matrix and recapture by other QDs. This carrier exchange is more pronounced for QD array with small localization energy and is getting more intensive as temperature increases. In a very dense QD array,
carriers may tunnel in lateral direction between neighboring QDs. These processes result in carrier lateral transport in realistic QD array making it “non-ideal”. Technologists usually try to fabricate as “ideal” as possible QWs or QDs. However, nanostructures that are intermediate in properties between QWs and QDs may attract special fundamental interest and be beneficial for certain practical applications. Recently we have developed novel quantum well-dots hybrid nanostructures (hereinafter QWD structures) that show some advantages over both QWs and QDs [3-4]. This hybrid type of the active region represents a QW, containing ultra-dense array of narrow-gap In-rich regions with increased thickness, which are capable of localizing electrons and holes. Using “QD language” the QWD structures can be considered as a high-density array of shallow In_{x}Ga_{1-x}As QDs inside thin In_{y}Ga_{1-y}As layer (x>y). In this paper, we focus on the influence of substrate orientation on structural and optical properties of QWDs and consider mechanisms of carrier recombination in QWD structures.

2. Experimental

Epitaxial structures were grown on either exact oriented or vicinal (100) GaAs substrates using a MOCVD installation with horizontal reactor. Metal alkyls (trimethylgallium, trimethylaluminum, trimethylindium and diethylzinc) as well as hydrides (arsine and silane) were used as precursors. The QWDs were formed using 3D growth mode at low growth temperature by the deposition of In_{x}Ga_{1-x}As with In concentration x from 30% to 50% and average thickness D (3-27 monolayers) on exact oriented as well as on 6° and 10° misoriented GaAs substrates.

Structures for studying photoluminescence (PL) and structural properties by transmission electron microscopy (TEM) contained one or several InGaAs QWD layers in a GaAs matrix surrounded with 50 nm-thick Al_{0.3}Ga_{0.7}As barrier layers. PL spectra were recorded with the use of a 2nd harmonics of YAG:Nd laser (532 nm) and cooled Ge photodiode employing a conventional lock-in technique.

Photovoltaic structures represented GaAs p-i-n-diodes with a 3 μm-thick base, a 0.5 μm-thick emitter and contained 15 InGaAs QWD layers in the i-region. Al_{0.3}Ga_{0.7}As and Al_{0.8}Ga_{0.2}As layers were utilized for window and back surface field layer, respectively. Photovoltaic characteristics were studied under illumination with terrestrial and space spectra.

A Titan 80–300 (FEI, USA) transmission electron microscope (TEM) with resolution of 0.136 nm and with 0.1 nm information limit in scanning TEM (STEM) and high resolution TEM mode, respectively, was used for structural and compositional characterization at nanometer and atomic levels. The Titan TEM was equipped with an energy dispersive x-ray spectrometer (EDS, Oxford Inca) with a spectral energy resolution of 130 eV. Cross-sectional TEM samples were prepared with a focused ion beam technique using a FEI Helios SEM/FIB Nanofactory system.

3. Results and discussion

Cross-sectional TEM images of QWD InGaAs layers grown on both exact (001) oriented (a, c) and vicinal (b, d) GaAs substrates are shown in Figure 1. The images were taken along <110> zone at medium (a, b) and high (c, d) magnifications to demonstrate the structure difference associated with substrate orientation. QD-like features are seen in both QWD InGaAs layers as thickness and compositional fluctuation, but they are much more pronounced in case of vicinal substrates due to presence of surface steps on the substrate. Compositional fluctuation result in contrast variations inside the QWD layers. Thicker areas have darker contrast indicating the presence of composition modulations along the QWD layers.

The thickness modulations may result from 3D growth mode employed for the growth of QWD layers being further enhanced by surface migration of In atoms due to lateral strain. High-resolution TEM images (figure 1, c) reveal that the thickness modulations often follow
the atomic steps on the vicinal surfaces. This explains stronger modulations of the thickness as well as larger lateral sizes (~20 nm) of QD-like features grown on vicinal surface (figure 1, c). Large lens-shaped islands on top of QW-like InGaAs layers are also seen in case of the sample grown on vicinal surface (figure 1, a) resembling QDs formed in Stranski-Krastanow growth mode. Such QDs cover more than one atomic step and may appear due to coalescence of individual QWDs. In case of exact (001) oriented surface, the thickness/composition modulations have smaller lateral size ~10 nm being more periodic (figure 1, d).

Figure 1. Cross-sectional TEM images of QWD InGaAs layers taken along <110> zone at medium (a,b) and high (c,d) magnifications. InGaAs layers were grown on both exact <001> oriented (a,c) and vicinal (b,d) GaAs substrates. One can see much higher variations in both thickness and composition of InGaAs layers in case of vicinal substrates due to presence of surface steps on the substrate. The compositional fluctuation are visible as changes in the gray scale variations.

The thickness and composition modulations in QWD structures that result in partial strain relaxation in the layers provide an opportunity to deposit more layers without formation of misfit dislocations in comparison with uniform QW of the same thickness and composition.

Figure 2 demonstrates the impact of substrate orientation on PL spectra. The QWDs were formed by the deposition of 8.1 ML of In$_{0.4}$Ga$_{0.6}$As on exact oriented as well as on 6° and 10° misoriented GaAs substrates. Samples grown on misoriented substrates show significantly broader PL spectra. This fact is in agreement with stronger modulation of QWD thickness and composition revealed by TEM studies. The inset of figure 2 shows the dependence of integrated PL intensity on substrate orientation for QWDs formed by the deposition of 7, 8, 9 and 10 ML of In$_{0.4}$Ga$_{0.6}$As. The highest PL intensity is achieved in case of 6° misoriented GaAs substrates and In$_{0.4}$Ga$_{0.6}$As effective thickness of 8-9 ML.
Substrate orientation significantly influences properties of photovoltaic QWD structures. GaAs solar cells (SCs) were based on 15 QWD layers grown under optimal conditions (8.1 ML-thick In$_{0.4}$Ga$_{0.6}$As QWDs separated by 40 nm thick GaAs spacers) on exact oriented and 6° or 10° misoriented GaAs substrates. Insertion of QWD layers in the SC grown on vicinal surface results in a broadening of SC sensitivity range from 900 nm to 1050 nm (figure 3) without any change (for 6° off) or with minimal changes (for 10° off) of the spectrum in the GaAs photosensitivity range (less than 900 nm). Thus, incorporation of 15 strained QWD layers (in case of 6° misoriented substrate) does not deteriorate crystal quality of GaAs p-n-junction. For the SC grown on exact GaAs substrate the form of QWD related part of QE spectrum looks similar to that for SC on vicinal surface (figure 3) but the QE in entire spectral range is about 30% lower. This fact can be explained by non-elastic strain relaxation accompanied by formation of misfit dislocations in case of QWDs grown on exact GaAs substrate. In contrast, more pronounced thickness and composition modulations in QWD structures on vicinal surface (figure 1, a) provide better elastic strain redistribution and correspondingly, high crystalline quality.

Figure 2. PL spectra of QWD structures formed by the deposition of 8.1ML of In$_{0.4}$Ga$_{0.6}$As on exact oriented as well as on 6° and 10° misoriented GaAs substrates. Inset: integrated PL intensity of QWDs formed by the deposition of 7, 8, 9 and 10 ML of In$_{0.4}$Ga$_{0.6}$As vs substrate orientation.

Figure 3. Internal quantum efficiency for reference GaAs SC (green) as well as GaAs SCs with 15 QWD layers grown on vicinal (6° off – green, 10° off – blue) and exact oriented (red) substrates.
4. Recombination mechanisms in QWD structures

Analysis of recombination process was done by fitting logarithm of excitation level as a function of logarithm of square root of integrated PL. The recombination rate of carriers is given by the so-called ABC model [5].

\[ P \sim A_n + B_n^2 + C_n^3 \]  

where \( P \) is carrier generation rate (excitation power density), \( n \) is carrier concentration (we assume that electron and hole concentrations are equal), \( A_n \) is the Shockley-Read-Hall (SRH) recombination rate (non-radiative recombination on defects), \( B_n^2 \) is the rate of radiative recombination, \( C_n^3 \) is the Auger recombination rate. It is obvious that at very low excitation conditions SRH mechanism dominates the others. As the excitation increases, an increasing number of high-order terms come into action. At high excitation densities the Auger mechanism becomes the dominant recombination process. Depending on relations of recombination coefficients in the intermediate excitation range one can expect radiative recombination process to be the most intensive one.

Fitting of dependencies of integrated PL intensity on excitation power by equation (1) allowed us determining relative contribution of different recombination processes (figure 4). Experimental dependencies are well fitted without consideration of Auger recombination (\( P \sim A_n + B_n^2 \)). Obviously, investigated range of excitation densities is insufficient for arising intensive Auger recombination.

Figure 4. Experimental data and fits of logarithm of excitation level as a function of logarithm of square root of integrated PL for QWD structures with In content of 40% and thicknesses 7, 8, 9 and 10 ML (a), temperature dependent experimental data and fits for QWD structures with In content of 40% and thicknesses of 8 ML (b). The points corresponding to equal SRH and radiative recombination rates are marked by stars.

Since we measured PL in arbitrary units we can estimate only interrelation of recombination constants or, that is more informative, estimate excitation level at which rates of non-radiative and radiative recombination become equal. Such inflection point on the dependency of integrated PL intensity on excitation power is marked by star in figure 4, a. The higher is the excitation density corresponding to inflection point the stronger is contribution of SRH (non-radiative) recombination. For the 10 ML-thick QWDs the inflection point is located at the highest excitation density as compared to the other samples and thus, the contribution of SRH (non-radiative) recombination is the most intensive. We attribute this to the fact that deposition of too thick QWD layer results in the formation of misfit dislocation. 7 ML-thick and 9 ML-thick QWDs show small and nearly the same excitation levels corresponding to the inflection point that implies the weakest non-radiative
recombination. In case of 7 ML-thick QWDs this fact can be explained by the low total strain. In case of 9 ML-thick QWDs we assume effective strain relaxation without creation of dislocations via formation of QWDs. Slightly higher excitation density corresponding to the inflection point for the 8 ML-thick QWDs may be due to some fluctuation of technological parameters during the MOCVD deposition.

As temperature decreases, a dramatic increase of radiative recombination rate in relation to SHR one (figure 4, b) occurs. Such behaviour is typical of different semiconductor systems [6-8]. Temperature lowering from 300 K to 150 K results in decrease of excitation density corresponding to the inflection point by more than an order of magnitude. Dependence taken at 80K shows no SHR as well as no Auger non-radiative recombination in the entire investigated range of excitation densities (4 orders of magnitude).

5. Conclusions
We described structural and optical properties of novel quantum well-dots hybrid nanostructures grown on exact oriented and vicinal (100) GaAs substrates using MOCVD. Thickness and composition modulations along the QWD layers are resolved by TEM for the structures grown on both exact oriented and vicinal substrates but for the latter ones they are much more pronounced. The thickness modulations appear due to 3D growth mode used to form the QWD structures. TEM images at high magnification reveal that the thickness modulations often follow the atomic steps existing on the vicinal surface. Effective strain relaxation in QWD provides an opportunity to deposit more layers without formation of misfit dislocations in comparison with uniform QW of the same thickness and composition. Structures grown on 6° misoriented GaAs substrates show higher PL intensity and quantum efficiency as compared to the ones grown on exact oriented substrate. Relative contribution of different recombination processes is evaluated by fitting logarithm of excitation level as a function of logarithm of square root of integrated PL with ABC model.

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