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Effects of Annealing on Self-Assembled InAs Quantum Dots and Wetting Layer in GaAs Matrix

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ABSTRACT

Post-growth thermal annealing effects on InAs/GaAs quantum dots (QDs) near Stransky-Krastanow transformation were investigated. Self-assembled QDs of average size of about 10 nm were grown by metalorganic vapour phase epitaxy. The photoluminescence (PL) due to emission from QDs as well as two peaks due to emission from the strained InAs wetting layer (WL) were observed in as-grown samples. Bimodal structure of the WL PL was attributed to WL regions of different thickness. There was almost no difference in the PL spectrum after 30 s annealing at 600°C. However, annealing at temperatures in the range between 700°C and 950°C resulted in quenching of the PL from QDs and the thinner WL. The PL peak from the new, thicker WL blue-shifted and narrowed with increasing annealing temperature. This behavior was in agreement with TEM observations. Complete dissolution of the QDs and substantial broadening of the WL was observed. All our results indicate that thermally induced modifications of the WL rather than QDs can be responsible for the blue-shift and narrowing of the PL peaks in structures containing InAs QDs.

INTRODUCTION

Thermally induced intermixing of semiconductor low-dimensional structures attracts an attention as a way of tuning their properties [1]. In recent years this technique has been applied to semiconductor quantum dots (QDs), which appear to be promising in many optoelectronic applications [2-4]. Although there are many reports dealing with this subject, some questions still remain. In particular, there is a debate over the character of carrier confinement in intermixed QDs. In this study we will show that the thermal annealing of QDs at relatively high temperatures can lead to their disolution and that the photoluminescence (PL) blue-shift and narrowing in such structures can be due to wetting layer (WL) modification.

EXPERIMENTAL PROCEDURE

In this study a layer structure grown by metal organic vapor phase epitaxy (MOVPE) was investigated. It contained self-assembled InAs QDs grown at 470°C in a Stranski-Krastanow growth mode on a GaAs (001) substrate. These QDs were then overgrown by a 100 nm thick GaAs layer. Six samples cut from this wafer were annealed using rapid thermal annealing (RTA) under nitrogen ambient for 30 seconds at 600°C, 700°C, 800°C, 850°C, 900°C and 950°C. Samples were covered with GaAs substrate during annealing. Surface morphology of such samples was not affected during RTA procedure. All annealed samples as well as the as-grown one were studied by PL technique. Measurements were performed at liquid helium temperature in a continuos flow CF-1204 Oxford cryostat with excitation from a semiconductor laser (λ= 780

nm). The signal was dispersed by a 0.5m monochromator and collected using a liquid nitrogen cooled germanium detector. In order to correlate changes in PL spectra with sample microstructure as-grown sample and samples annealed at the four highest temperatures were studied using transmission electron microscopy (TEM). Cross-sectional TEM specimens were prepared from these samples by the standard method of mechanical thinning followed by ion milling. In addition, plan-view specimens were prepared from the as-grown sample. Wet etching in a 5 % methanol solution of Br was applied.

EXPERIMENTAL RESULTS

The PL spectrum of the as-grown sample contains three main features as can be seen in Fig.1. A broad peak at 1.155 eV is due to the optical recombination within the InAs QDs. Its broadening reflects an inhomogeneous size distribution of QDs. Two other PL features observed at 1.42 eV and 1.37 eV were due to the InAs WL. It is known that both the QD and WL PL peaks can be simultaneously observed from QDs near Stransky-Krastanow transformation density [5]. The bimodal structure of the WL-related feature is probably due to different thickness of the WL in different sample regions. The lower energy PL is due to emission from a thicker WL whereas the higher energy PL peak is due to emission from a thinner WL.

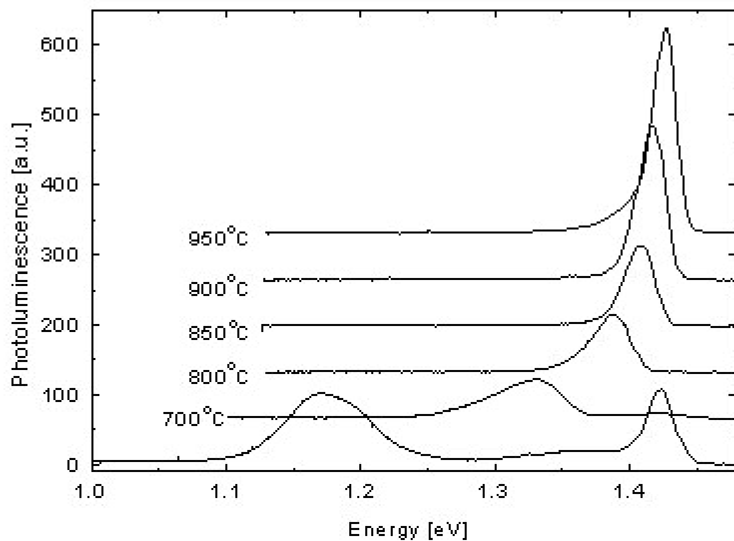

Fig.1. Low temperature photoluminescence from as grown and annealed samples.

TEM results obtained for as-grown sample are in agreement with this interpretation of the PL spectrum. In the, as-grown sample a high density of self-assembled QDs was present. They are clearly visible in bright field TEM images taken from plan-view samples under two-beam (0,g) condition, where g was of the (220)-type reflection (Fig.2). The average size of these QDs was about 10 nm. Their density, as estimated form plan-view observation, was of the order of 10^{11} cm^{-2}. TEM results obtained for this plan-view sample indicated that QDs present in as-grown samples were fully strained. No indication of any strain relaxation such as the presence of big, dislocated islands or dislocations threading through the capping layer was observed. The presence of self-assembled QDs in as-grown sample was also confirmed by studies of cross-

sectional specimens. This can be seen from the high-resolution electron microscopy (HREM) image shown in Fig. 3a. Well-defined InAs QDs can be seen in this image.

The HREM studies of as-grown sample also confirmed the presence of a WL as suggested by the features measured in the PL spectrum. One can notice in Fig. 3a a thin WL connecting the bases of adjacent QDs. The thickness of this WL is not uniform. We found that it can change significantly in neighboring areas. Fig. 4 shows such a situation. HREM images of the WL presented in Fig. 4a and Fig. 4b are magnified images of areas A and B shown in Fig. 4c. One can notice that the WL in area A, located farther from the QDs is about half the thickness of that in area B, located closer to the QD.

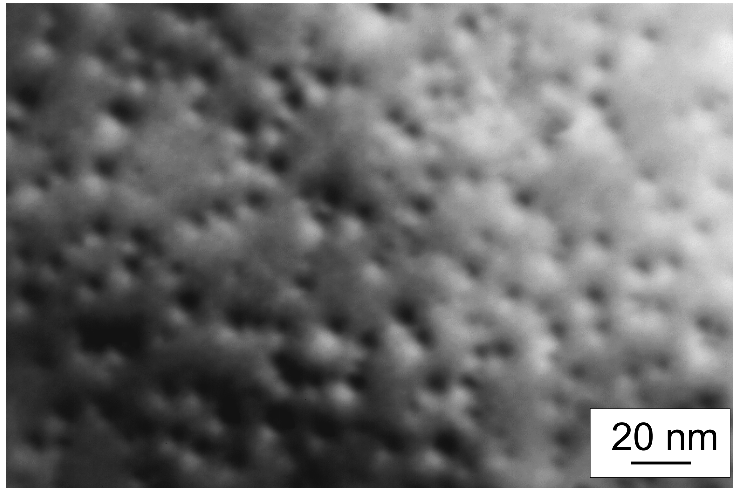

Fig. 2. Bright field TEM image of the plan-view as grown sample.

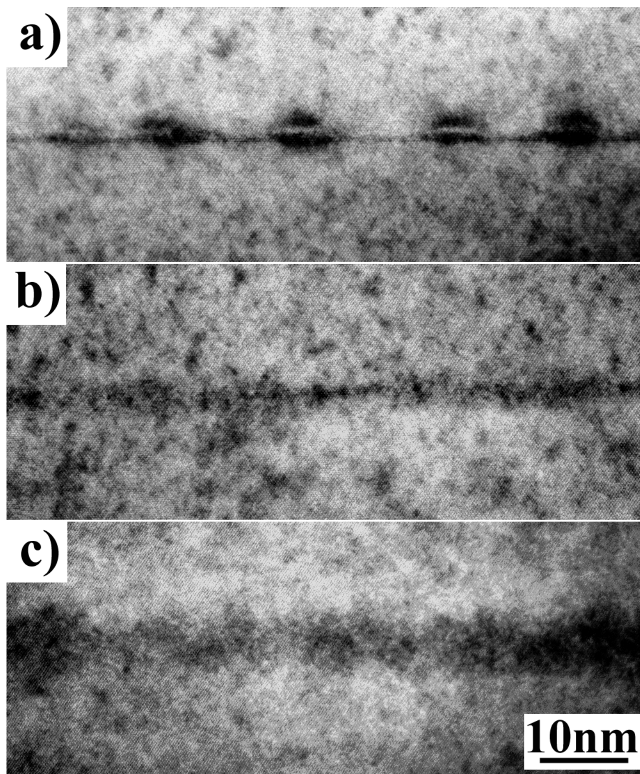

Fig. 3. HREM images of samples: as-grown (a) and annealed at 800°C (b) and 950°C (c).

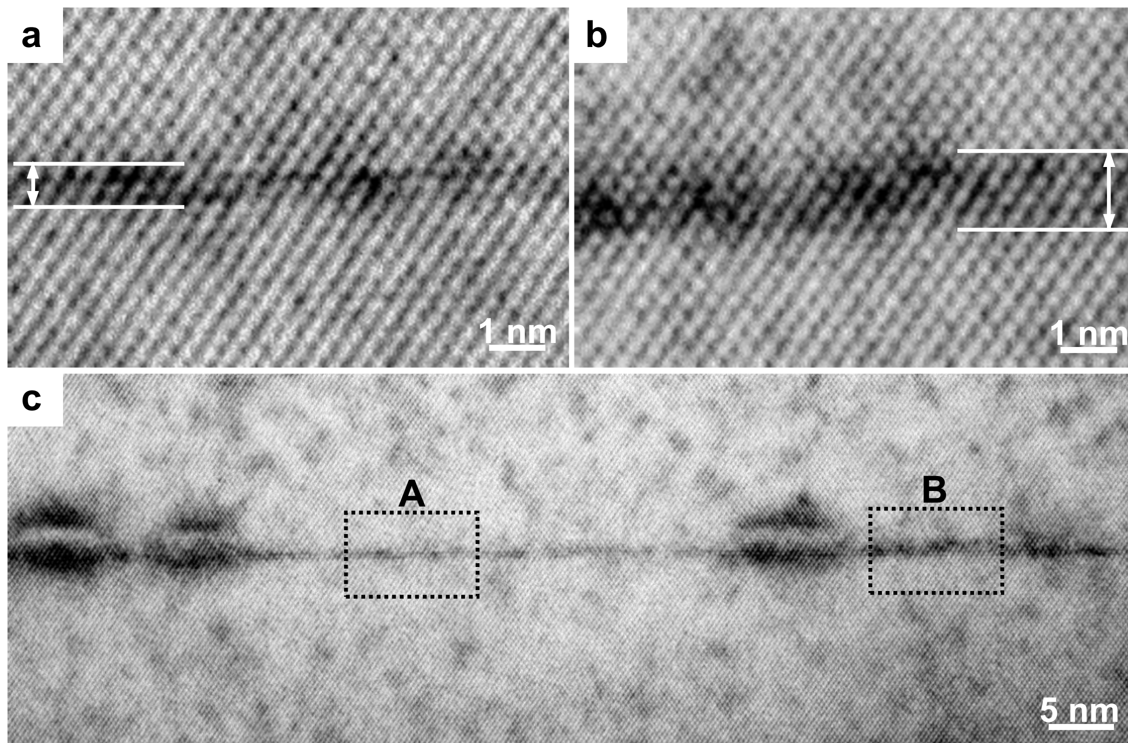

Fig.4. HREM images of InAs WL in area A (a) and B (b) shown in (c). Notice that WL thickness further from QDs in area A is almost twice thinner than that in area B closer to QD.

Whereas, annealing at 600°C (for 30 s) did not result in any significant changes in the PL spectrum, that spectrum changed drastically after thermal treatment at 700°C or higher temperature (Fig.1). Only one PL peak can be observed for samples annealed at such temperatures. The PL peak energy position changed in these samples from 1.33 eV (700°C) to 1.427 eV (950°C). In addition to the blue-shift of this peak significant narrowing and intensity increase occurred with increase of annealing temperature.

A PL blue-shift and narrowing in thermally treated QDs was previously reported for similar InAs QDs grown by molecular beam epitaxy (MBE) [6]. However energy shift of the PL peaks in our annealed samples is significantly higher than reported previously. This suggests that the processes observed in our experiment can be of different origin. One possible explanation is the effect of capping. The samples studied in [6] were capped with SiO_2. It is known that such a capping creates group III point defects due to Ga outdiffusion [7] and promotes the In-Ga interdiffusion across the dot-cap interface. Another possible explanation is the morphology of the as-grown sample. We are dealing with QDs near the Stransky-Krastanow transformation characterized by simultaneous WL and QDs emission [5], whereas the study reported in [6] deals with saturated QDs with no WL PL.

RTA treatment of our sample results in quenching of the PL from the thinner WL regions [2]. This may be due to diffusion of In out of the QDs, which thickens surrounding WL regions. Simultaneously the thicker WL undergoes an intermixing, which results in a blue-shift and narrowing of its PL peak. Therefore in our experiment the PL seen from annealed structures is attributed to the WL and not to the QDs. This interpretation is confirmed by our TEM studies of annealed samples.

Drastic changes of microstructure were found after high-temperature treatment. In all annealed samples which were investigated by TEM (samples annealed at 800°C and higher temperatures) QDs were no longer observed. There were still some indium fluctuations visible in the HREM images of the sample annealed at 800°C (Fig.3c), however there were no well defined QDs as in as-grown samples (Fig.3a). HREM studies of annealed samples showed also that disapearence of QDs was asocciated with drastic broadening of the WL (see Fig.3). The layer thickness was found to increase to approximately 3 nm after annealing at 800°C (Fig.3b) and to about 8 nm after annealing at 950°C (Fig.3c). WL thickness measured from TEM images ploted *vs.* annealing temperature is shown in Fig.5a.

The thickness of the WL in annealed samples, measured from TEM was used to estimate relative changes of indium content present in these layers. A simple model was employed. It was based on the assumption that all the indium present before anneling mainly in the form of InAs QDs was uniformly redistributed during thermal treatment and that for a specific annealing temperature the WL had a constant indium composition: $In_xGa_{1-x}As$. It can be shown that under such an assumption indium content (x) depends on the layer thickness (t) according to the formula:

$$x = (a_{InGaAs}/a_{InAs})^3 \rho V/t$$

where, a_{InGaAs} and a_{InAs} are lattice parameters of $In_xGa_{1-x}As$ and InAs, and ρ and V are areal density and average volume of QDs, respectively. According to Vegard's law [8] the lattice parameter of $In_xGa_{1-x}As$ also depends on indium composition. This dependence is described by the following formula:

$$a_{InGaAs} = x a_{InAs} + (1-x) a_{GaAs}$$

where, a_{GaAs} is a lattice parameter of GaAs.

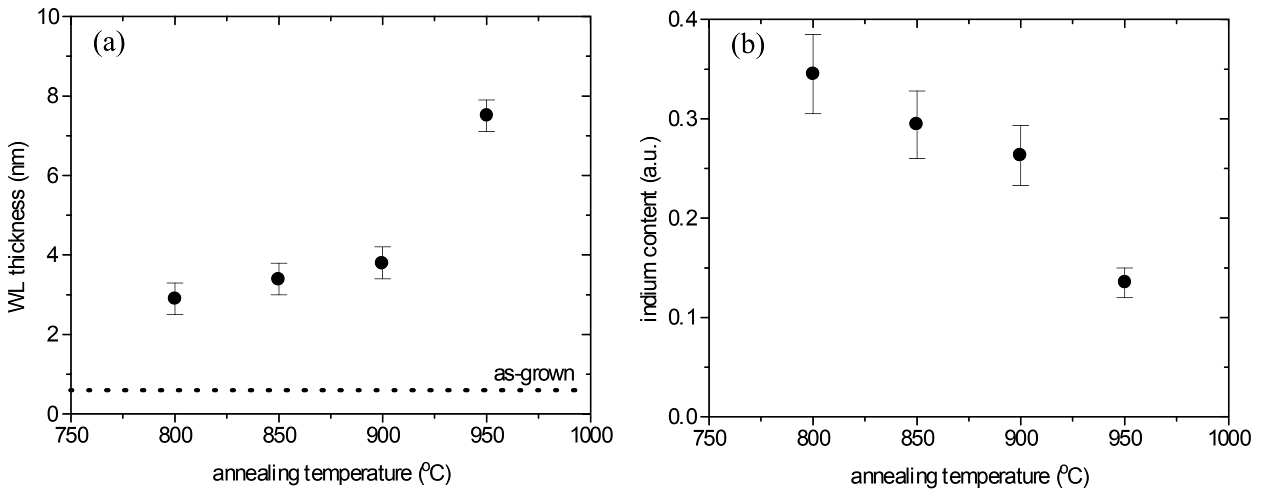

Fig. 5. WL thickness measured from TEM (a) and estimated relative changes of indium content in WL (b) plotted vs. annealing temperature.

Relative changes of indium content estimated from TEM data using this simple model are plotted vs. annealing temperature in Fig. 5b. One can notice that annealing at higher temperature leads to larger interdiffuison within the WL. In our opinion this interdiffusion is responsible for the blue-shift of the PL peak observed in spectra measured for samples annealed at higher temperatures.

CONCLUSIONS

In conclusion, we studied the effect of thermal treatment on the structures of InAs/GaAs QDs grown by MOVPE. PL and TEM studies of samples annealed at temperatures up to 950° C strongly suggest a complete disappearance of QDs and substantial thickening of the WL. It is proposed that the thermally induced modification of the WL rather than changes of QDs can be responsible for the blue-shift and narrowing of PL peaks in our structures originally containing InAs/GaAs QDs.

ACKNOWLEDGEMENTS

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