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TEM/HREM characterization of self-organized (In,Ga)As quantum dots

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ABSTRACT: The morphology evolution of molecular beam epitaxy grown InAs and $In_{0.5}Ga_{0.5}As$ layers as a function of deposition thickness, ranging from 1 to 10 monolayers (ML), is studied by transmission electron microscopy (TEM) to characterize the formation and the self-organization of pseudomorphic quantum dots. The luminescence of all samples with coherent dots exhibits a high quantum efficiency.

1. INTRODUCTION

Currently, self-organization phenomena in crystalline semiconductors have increased in research interest (see e.g. Leonard et al. 1993, Nötzel et al. 1994, Ledentsov et al 1994) as a way possible to form nanoscale islands, namely quantum dots (QDs), without any patterning process. QDs are expected to exhibit unique properties like δ-function density of states leading to novel and/or strongly improved properties for photonic and electronic devices, e.g. lasers. Recently, this δ-function density of states (Ledentsov et al. 1994, Grundmann et al. 1995) and QD lasers (Kirstaedter et al. 1994) with unique properties were demonstrated.

Islanding growth in the (In,Ga)As-GaAs system is considered to result from a morphology evolution of the two-dimensional (2D) layer after the growth of 1 - 2 monolayers, explained by a Stranski-Krastanov model of coherent island growth. Either the elastic relaxation of the strain in heterostructures, i.e. a local minimum in the total energy (Eaglesham & Cerullo 1990), or the kinetics of strain-induced surface roughness (Snyder et al. 1991, Madhukar et al. 1994) has been assumed to be responsible for the self-organized formation of nm-size 3D islands. However, a conclusive picture for the 2D-3D morphology transformation has not yet been established. The results of the present work demonstrate the importance of both strain and temperature, which implies that there are energetic and kinetic effects.

Here we report on the nucleation of (In,Ga)As QDs on GaAs during the growth of thin pseudomorphic layers. Our experimental results as to dot size and shape differ from those reported in other papers on InAs-GaAs (Moison et al. 1994)) and InGaAs-GaAs dots (Leonard et al. 1993, Mo et al. 1990) grown under similar conditions. Here pyramid-shaped dots, as known for Ge on Si (Mo et al. 1990), are reported for InAs and $In_{0.5}Ga_{0.5}As$ for the first time. In addition, a self-organized short-range ordering of dots into rows along <100> is revealed, forming a primitive two-dimensional cubic lattice.

2. RESULTS AND DISCUSSION

InAs and InGaAs dots were grown by molecular beam epitaxy (MBE) on GaAs (001) using an EP1201 system (see, e.g., the details in Ledentsov et al. 1994). Their evolution was studied *in situ* by reflection high energy electron diffraction (RHEED), and *ex situ* (after the deposition of a cap layer) by transmission electron microscopy (TEM) using JEOL JEM1000 (1MV) and JEM4000EX (400kV) microscopes. Low-temperature (T = 8 K) photoluminescence (PL) and cathodoluminescence (CL) were used to characterize the optical properties of the dots.

In situ RHEED experiments show that the critical thickness for the formation of 3D islands (quantum dots) depends on the layer composition, i.e. elastic energy. Indeed, the critical thickness of $In_xGa_{1-x}As$ increases from 1.7 to 3 ML with decreasing In content x from 1.0 to 0.5. TEM studies generally confirm this observation. Figs. 1 (a)-(d) show typical plan-view TEM images of InGaAs and InAs layers grown at T_d=480°C and differing in thickness (which is above the critical value in all four cases). At the initial stages of (In, Ga)As dot formation with slashes in the RHEED pattern just appearing, the corresponding TEM images (see, e.g., Fig. 1 (a) for 3.3 ML of $In_{0.5}Ga_{0.5}As$) demonstrate a fine-scale, black-and-white granular contrast composed of round-shaped dots having a diameter of approxiamtely 6 nm. Locally connected, the dots appear in agglomerates. Depositing further material causes to the formation of well-developed coherent islands of increasingly uniform size, most probably owing to the higher growth rate of relatively small islands (see Figs. 1 (b); (d)). At that stage diffraction spots appear in the RHEED pattern. At a coverage of 5.3 ML of $In_{0.5}Ga_{0.5}As$, coherent islands of about 5-15 nm in size are observed. Further increasing the nominal thickness of $In_{0.5}Ga_{0.5}As$ up to 7.3 ML results in the formation of well-developed dots with size and spacing less varing (Fig. 1 (b)).

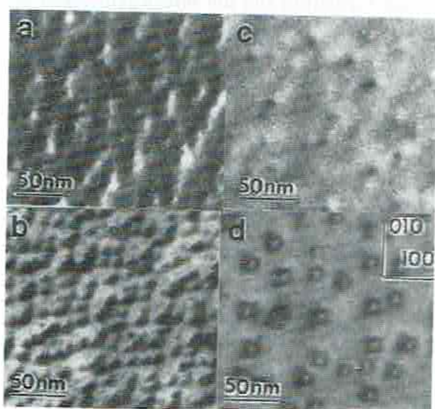

Fig.1 a-d. Plan-view bright-field electron micrographs of (In,Ga)As quantum dots corresponding to different nominal thicknesses of In-containing layers: 3.3 ML (a) and 7.3 ML (b) of $In_{0.5}Ga_{0.5}As$, and 2 ML (c) and 4 ML (d) of InAs. Images (a)-(c) are taken at g=220, Fig. (d) is a symmetrical [001] zone-axis image.

The formation of well-developed InAs dots (at T_d = 480°C) already begins at an average thickness of less than 2 ML. Figs. 1 (c) and (d) show dots formed during InAs deposition of nominal thicknesses of 2 and 4 ML, respectively.

Comparing the results of the InAs and $In_{0.5}Ga_{0.5}As$ deposition indicates that the onset of dot formation is governed by the strain energy per interface area. Figs. 1 (b) and (d) indicate that the dots are of pyramidal shape with the square base of those principal axes being close to the two orthogonal <100> directions and the average length being about 12±1 nm.

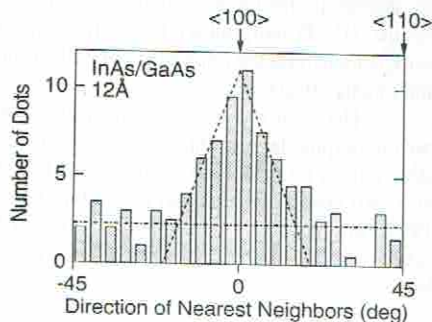

Fig.2. Histogram of next neighbour dot direction (modulus 90°) for 4 ML InAs, obtained from a TEM image as, e.g., Fig. 1 (d). The dashed line denotes the partially disordered square lattice, the dash-dot line illustrates the superimposed random dot distribution.

The size of our InAs dots is similar to that reported by Leonard et al (1994) but about half of that reported by Moison et al. (1994), obtained by atomic force microscopy (AFM), for dots of an average coverage of 2.3 ML grown at 500°C, under rather similar growth conditions. Thus either AFM overvalues the size of the dots, or the dot formation is extremely sensitive to the growth conditions. The size distribution of the dots in InAs samples of 2 and 4 ML is rather narrow (< 20%) and similar to results reported by Leonard et al (1994). The variations of dot size and interdot distance at the initial stages of dot formation for $In_{0.5}Ga_{0.5}As$ deposition are larger than that for InAs deposition.

The dots align in rows along <100>, demonstrating the symmetry of a two-dimensional primitive cubic lattice as Fig. 1 (d) reveals. A histogram of the direction of adjacent dots (Fig. 2) was obtained from a statistical analysis of a TEM image. It clearly has a maximum in <100> direction confirming the impression from the image of Fig. 1 (d). The dashed line in Fig. 2 marks the dependence expected of dots in a partially disordered square lattice. The self-ordering is superimposed by some randomness, resulting in an offset (dash-dot line) independent of the azimuth.

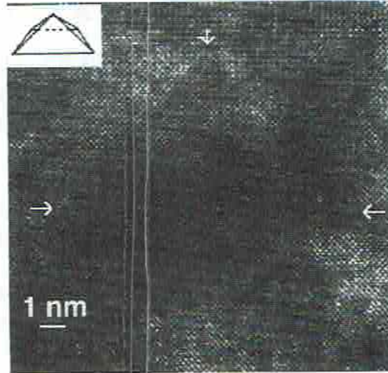

Fig. 3. Experimental 400 kV HREM micrograph of 3 ML InAs QD in GaAs. Arrows indicate the boundary facets.

The dot geometry is additionally confirmed by cross section HREM images as in Fig. 3. The height of the dots is about 6 nm. The sidewalls are close to {110}. Weakly pronounced is the top of the pyramid which together with the strong strain-induced contrast around the dot is shown in the simulated HREM micrograph of Fig.4 calculated by use of CERIUS Programme package (Molecular Simulation Inc.), with the pyramid always appearing to be truncated.

Being rather typical dot size and shape seem to be energetically favourable (Tersoff & Tromp 1993), corresponding to the energy decrease arising from the strain relaxation similar to surface faceting (Marchenko 1981). Growth kinetics are likely to strongly affect the initial stage of layer transformation either at extremely low substrate temperatures or under the changing the optimal As pressure. The regular distribution of the dots may be due to two alternative processes, viz. the ordering of nucleation centres on the surface followed by equalization of the dot sizes during further material deposition and/or by strain field induced ordering with increasing coverage.

Indeed, periodic lateral modulations of the layer composition have been observed in highly strained heterostructures (Glas et al 1990) resulting from the spinodal decomposition of ternary or quaternary solid solutions. Similar effects may also result in the formation of quasi-periodic dot arrays.

Fig. 4. Simulated HREM image of pyramidal InAs QD in GaAs (400 kV, C_s=1 mm, a_o=0.6 A^{-1}, def=-70 nm, foil thickness t=22.6 nm).

The optical properties of the dots are correlated with the morphology revealed by TEM. In general, at low temperatures the dots exhibit a high internal quantum efficiency close to 100% . Fig. 4 depicts PL spectra of the structures with 2 ML and 4 ML of InAs. They differ in peak position and intensity in accordance with the TEM data on size and distribution of the dots. The smaller dots with larger dispersion give rise to a weaker and broader PL line which is also closer to the GaAs bandgap energy as compared to the PL peak of the larger dots showing a more homogeneous size distribution.

The low density of quantum-dot induced states in 2 ML InAs dots is not distinguishable in the absorption spectrum, which is dominated by the large density of states in the residual non-transformed 2D layer. For a dense array of uniform InAs dots (Fig. 1 (d)), however, the absorption peak of the dot ground state is clearly identified, and coincides with the PL maximum (the absorption of the wetting layer additionally occuring here).

The corrugation of the $In_{0.5}Ga_{0.5}As$ layer (Fig. 1 (a)) revealed by TEM at the initial stage of layer transformation has also a strong effect on luminescence properties resulting in an intense PL peak which is shifted from the position expected for a uniform InGaAs layer (~1.4 eV for a 1 nm thick $In_{0.5}Ga_{0.5}As$ layer) to lower energies. The structures with 3.3 to 4 ML of $In_{0.5}Ga_{0.5}As$ deposited at 400 - 450°C exhibit the PL peak at 1.23-1.26 eV with 40 - 60 meV as the full width at half maximum (FWHM).

Here we observe a large Stokes shift between the PL line (1.24 eV) and the absorption (1.31 eV) indicating a non-uniform size distribution of InGaAs dots.

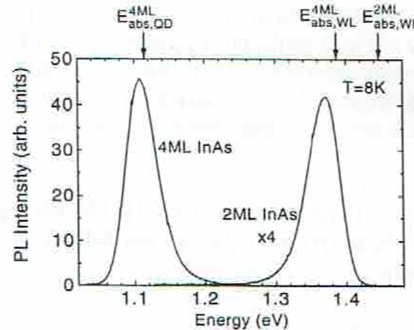

Fig. 4. Low-temperature (T=8 K) PL spectra of quantum dots in samples with 2 ML and 4 ML of InAs of an excitation density D=1 Wcm^{-2}. The peak energies in the absorption spectra of the wetting layer (WL) in both samples and the quantum dots (QD) in the 4 ML sample are marked by arrows.

In conclusion, the formation of ordered arrays (forming a primitive two-dimensional cubic lattice) of pseudomorphic dots of pyramid-like shape, typically 6 nm in height and about 12 nm in base diameter, has been observed on a residual two-dimensional layer above a nominal coverage of 2 ML for InAs, and of 6 ML for $In_{0.5}Ga_{0.5}As$ on GaAs. The dispersion of dot size and interdot distance was remarkably low (< 20 %). Efficient luminescence from dot states has been observed for all samples. For dense dot arrays (~10^{11} dots/cm^2) grown under optimum conditions, the quantum dot ground state photoluminescence and absorption were found to coincide energetically.

ANKNOWLEDGEMENT

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REFERENCES

Eaglesham D J & Cerullo M 1990 Phys. Rev. Lett. 64 1943
Glas F, Gors C & Henoc P 1990 Phil. Mag. B62 373
Grundmann M, Ledentsov N, Christen J et al 1995 phys. stat. sol. *in press*
Kirstaedter N, Ledentsov N N, Grundmann M et al 1994 Electr. Lett. 30 1416
Ledentsov N N, Grundmann M, Kirstaedter M et al 1994 Proc. of the 22nd Int. Conf. Phys. Semicon., (Vancouver, Canada, 1994) *in press*.
Leonard D, Krishnamurthy M, Reaves C M, Denbaars S P & Petroff P M 1993 Appl. Phys. Lett. 63 3203
Leonard D, Krishnamurty M, Fafard S, Merz J L & Petroff P M 1994 J. Vac. Sci. Technol. B12 1063
Madhukar A, Xie Q, Chen P & Konkar A Appl. Phys. Lett. 1994 64 2727
Marchenko V I 1981 Sov. Phys. JETP 54 605
Mo Y-W, SavageD E, Swartzentruber B S and. Lagally M G 1990 Phys. Rev. Lett. 65 1020
Moison J M, Houzay F, Barthe F, Leprice L, Andre E & Vatel O 1994 Appl. Phys. Lett. 64 196
Nötzel R, Temmyo J & Tamamura T 1994 Nature 369 131
Snyder C W, Orr B G, Kessler D & Sander L M 1991 Phys. Rev. Lett. 66 3032
Tersoff J & Tromp R M 1993 Phys. Rev. Lett. 70 2782