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InGaAs/GaAs Quantum Dots (~15nm) Grown by MOCVD

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We report the fabrication of InGaAs-dots with diameters of about 15 nm on GaAs-surfaces by metal organic chemical vapor deposition growth. Scanning electron micrographs and atomic force images show highly uniform dots formed by the Stranski-Krastanow growth mode. The photoluminescence spectra of buried dots indicates efficient carrier capture at room temperature. The average area dot density can be controlled from 10^8cm^{-2} up to more than 10^{11}cm^{-2} by substrate off-angle orientation, InGaAs deposition thickness, and growth temperature, respectively.

1. Introduction

The formation of quantum dots has recently received great attention for optoelectronic device applications as well as for basic physics[1]. To exhibit zero-dimensional confinement with high quantum efficiency, highly uniform quantum-size dots with a large area dot density are desirable. For the realization of these dot structures, usually a pattern is defined by high resolution lithography and transferred to a semiconductor quantum well substrate, e.g., by deep dry chemical etching[2, 3] or impurity induced disordering using ion beam implantation[4]. The physical properties of dots defined in this manner often are determined by surface effects, process induced damage and rough heterointerfaces. Alternative methods which fabricate the dot structures directly during the growth are the growth of microclusters on terraced surfaces[5] or selective growth on patterned substrates[6].

A promising path for the clean and defect free formation of dot structures directly on the epilayer surface is the Stranski-Krastanow growth method[7]. This growth mode is started with an initial two dimensional layer deposition on the substrate material. After a critical layer deposition thickness is achieved, the surface transforms into three dimensional highly strained dots that grow coherently on the heterostructure interface. The advantages of this dot fabrication technique are that no nanolithography and etch or implantation induced process are necessary. The dots are grown *in-situ* without breaking the growth run, maintaining a homogeneous surface morphology and avoiding defect creation. Previous studies of InGaAs/GaAs-dots by the Stranski-Krastanow growth method show the accumulation of nearly defect free dot structures on GaAs surface[8-11]. Leonard et al. demonstrated impressive formation of optically active InGaAs-quantum-size dot structures on GaAs by molecular beam epitaxy (MBE)[11]. InP-dots on a InGaP/GaAs-substrate were also successfully produced by using hydride vapor phase epitaxy (VPE)[12] and metal organic chemical vapor deposition (MOCVD)[13]. However, up to now all of the studies which realized a dot size smaller than 50 nm have used MBE growth technique: the dot sizes ranged from about 18 nm up to more than 50 nm. Moreover, the accurate controlling of dot diameter and area dot density by growth conditions has been an unsolved question.

In this paper, we report the successful fabrication of InGaAs-dot structures with the diameter of about 15 nm on GaAs-surfaces by MOCVD growth technique. The dot structures are produced highly uniformly by the Stranski-Krastanow growth method. Furthermore, strong and narrow photoluminescence (PL) emission spectra of the dots even at room temperature demonstrates efficient carrier capture and homogeneously heterostructure interface. In addition, the average area dot density and diameter can be controlled accurately by substrate off-angle orientation, InGaAs deposition thickness, and growth temperature, respectively.

2. Experimental

The samples were grown by MOCVD growth technique, which was carried out on GaAs-substrates in a low-pressure (76 torr) horizontal quartz reactor. During growth the partial pressure of arsine (AsH_3), trimethylgallium (TMG), and trimethylindium (TMI) was kept at 2.2 x 10^{-4}, 2.2 x 10^{-6}, and 2.2 x 10^{-6}atm, respectively. The dots were grown with a composition of $In_{0.5}Ga_{0.5}As$ using hydrogen as a carrier gas, with a total gas flow rate of 9 liter/min. The group V/III ratio was 50 for $In_{0.5}Ga_{0.5}As$-, and GaAs-growth, respectively. Due to the complicated surface diffusion kinetics during the growth of the highly strained dot structures, a deviation from the expected $In_{0.5}Ga_{0.5}As$-composition is possible[13].

A 200 nm GaAs-epilayer was initially grown at the temperature of 500 ^oC and a rate of about 0.2 monolayers (ML) per second. The InGaAs-dots were formed with an InGaAs deposition thickness between 1.5 and 5 ML, corresponding to InGaAs deposition times between 3 and 10 s. To study the dot formation as a function of substrate off-angle, vicinal (100) GaAs-wafers tilted between 0^o and 41^o towards [111] B-orientation were used. For the scanning electron microscopy (SEM) and atomic force microscopy (AFM) investigations of the dot formation, the growth was stopped at this point and the sample was cooled down to room temperature under arsine pressure. For PL measurements the dots were immediately covered with a 30 nm GaAs-cap barrier layer. To avoid the aggregation or degradation of the quantum-size dots, the GaAs-cap layer was deposited at the growth temperature of 400 ^oC. Reference samples without InGaAs-dots and sample structures including an InGaAs-quantum-well (QW) layer of 5 nm thickness sandwiched between a 100 nm GaAs-layer were grown under similar conditions.

The successful fabrication of 15 nm InGaAs-dots on AlGaAs-surface is described elsewhere[14].

3. Results and Discussion

Figures 1(a) and 1(b) show high resolution SEM-micrographs of an InGaAs-dot on a GaAs-surface in plane view, and cross-section, respectively. The dot has a highly uniform shape without facets. The absolute diameter of the dot is about 15 nm and the dot height is about 6 nm.

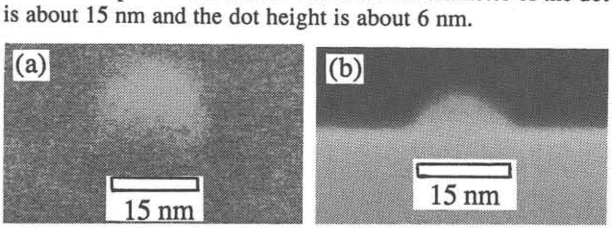

Fig. 1: High resolution scanning electron micrographs of an InGaAs-dot on a GaAs-surface in (a) plane view and (b) cross-section.

This structure is the first InGaAs-dot ever prepared by the MOCVD in the Stranski-Krastanow growth mode. This dot formation occurs for an InGaAs deposition thickness of 1.5 ML (3 s) at growth temperature of 500 ^{o}C.

Figure 2 show the AFM-micrograph of the InGaAs-dots on GaAs for an average dot density of 10^{11}cm^{-2} obtained for an InGaAs deposition thickness of 2.5 ML (5 s). The substrate angle is 0^{o} and the growth temperature is 500 ^{o}C.

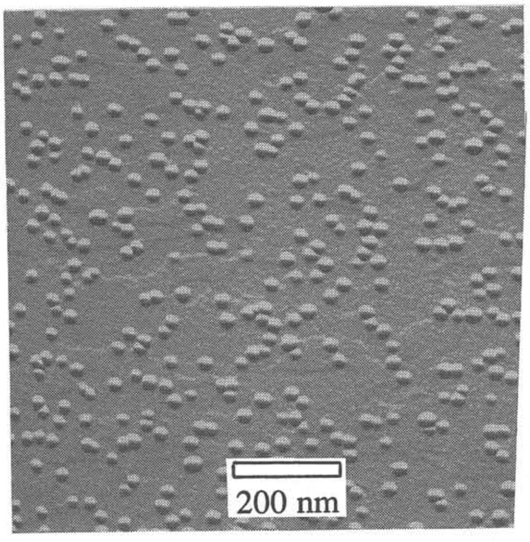

Fig. 2: Atomic force image of the InGaAs-dots on (100) GaAs-surface for high area dot density of about 10^{11}cm^{-2}.

The dots show a highly uniform diameter of about 15 nm for the whole sample area. The smooth and homogeneous surface layer between the dot structures demonstrates the high quality growth process by MOCVD technique. Furthermore the growth of many dot structures under same growth conditions yields similar results for average dot diameter and area dot density, demonstrating the high reproducibility of the MOCVD process.

Figure 3 shows PL-emission spectra of the buried InGaAs-dots measured for temperatures between 15 and 300 K.

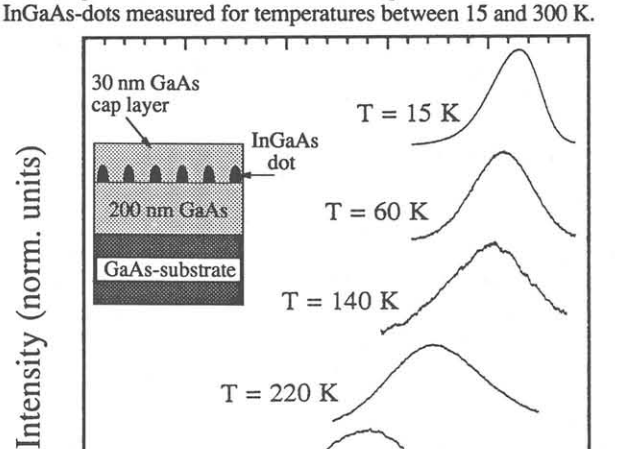

Fig. 3: Temperature dependence of photoluminescence emission spectra of the dots according to a sample structure including dots with a diameter of about 15 nm and an average area dot density of 10^{9}cm^{-2}. The laser excitation power is 12 mW.

The narrow dot emission linewidth indicates highly uniform dots with negligible size fluctuations and a homogeneous heterostructure interface. The emission band shifts about 80 meV from 15 to 300 K. This dot emission energy shift is similar to the emission energy shift of an InGaAs-reference QW of 5 nm thickness grown under same growth conditions, indicating that the gap energy of the dots is not affected by defects. In addition the absolute emission intensity of dots and QW-layer is similar at 15 and 300 K, demonstrating efficient carrier capture of the quantum size dots. We would like to point out that the measured dot emission has not been scaled with area filling factor. (Inclusion of a filling factor corresponding to a dot density of 1 x 10^{9}cm^{-2} and dot diameter of 15 nm would lead to a dot intensity which is a factor of about 500 larger compared to the QW-layer intensity). For a laser excitation power of 0.12 mW a sharp emission linewidth of 14 meV (T = 15 K) is obtained. Furthermore the emission intensity of the dots shows linear response with the laser excitation power ranging between 0.12 and 55 mW. There is no indication of defect formation, which demonstrates the high potential of MOCVD growth process for the fabrication of dot structures.

To verify that the emission comes from the dot structures, samples with dots but no GaAs-top barrier layer and sample structures without InGaAs-dots but with cap-layer deposition were fabricated under similar growth conditions and investigated by PL-spectroscopy. No luminescence is detectable from either structure, demonstrating that a buried dot structure is necessary for optical activation.

Figure 4 displays the average dot density as a function of substrate off-angle for InGaAs monolayers deposition thickness of 1.5, 2.0 and 2.5 ML at growth temperature of 500 ^{o}C. The average dot density can be tuned from 10^{9}cm^{-2} up to more than 10^{11}cm^{-2} by increasing the off-angle from 0^{o} to 27^{o}, and the InGaAs deposition from 1.5 to 2.5 ML, respectively.

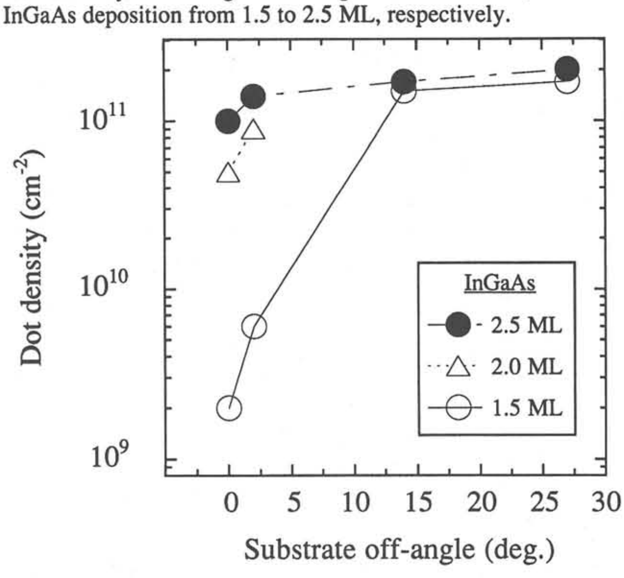

Fig. 4: Average dot density of InGaAs/GaAs-dots as a function of substrate off-angle for different InGaAs deposition thickness between 1.5 and 2.5 monolayers. The growth temperature is 500 ^{o}C.

A low dot density of 2 x 10^{9}cm^{-2} is formed by growing InGaAs for a low deposition thickness of 1.5 ML on 0^{o} off-angle substrate. High dot density structures with an average density up to 2 x 10^{11}cm^{-2} were obtained for a large InGaAs deposition thickness of 2.5 ML and a large substrate off angle of 27^{o}. The high dot density is most likely correlated to the larger substrate surface step density for larger off-angle misorientation. For a high surface step density a low surface diffusion and an enhancement of area dot density is plausible.

For a very large substrate off-angle orientation of 41^o (not shown) large InGaAs-island structures with facets appear and only a few small-dot structures are found among the surface layer, indicating the almost complete coalescence of small-size dots.

It should be noted that for the low dot density structures with an average InGaAs deposition thickness of 1.5 ML only quantum-size dots with a diameter of 15 nm appear. No larger dot structures are found on the whole sample surface. For the high dot density samples corresponding to an average InGaAs deposition thickness of 2.0 and 2.5 ML, a few mid-size and large-size dots with an absolute diameter of about 30 and 50 nm were found among the small-size dots. The average area dot density of the mid- and large-size dots (about $10^7 cm^{-2}$) is four orders of magnitude smaller than that of the small-size dots. For a very large InGaAs deposition thickness of 5 ML (10 s), the average dot density of the mid- and large-size dots is increased to $10^8 cm^{-2}$. Only a few small-size dots of about $10^9 cm^{-2}$ are observed, suggesting the coalescence of small-dots to mid- and large-size dots. For InGaAs deposition thickness below 1.5 ML no dot formation is observable, most likely due to the desorption of InGaAs-dots below a critical coverage thickness.

In order to study the dot diameter and density formation for growth temperatures above 500 ^oC, sample structures with a fixed InGaAs-dot deposition of 2.5 ML were grown on 0^o substrate. As described above, for this InGaAs coverage only a few larger dots with a diameter around 50 nm and a density of about $10^7 cm^{-2}$ occur. The average dot diameter and density for the small-size dots as a function of growth temperature is shown in Fig. 5.

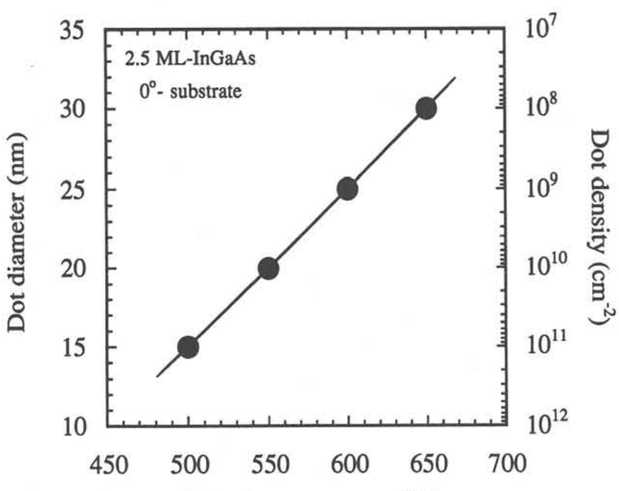

Fig. 5: Average diameter and density dependence of the quantum-size InGaAs/GaAs-dots versus growth temperature. The InGaAs deposition thickness is about 2.5 monolayers and the substrate off-angle 0^o.

Dot diameter and density can be changed linearly with growth temperature. By increasing the growth temperature from 500 to 650 ^oC, the dot diameter is increased by a factor of two and the dot density is decreased up to about three orders of magnitude. At 650 ^oC InGaAs-dots with a diameter of about 30 nm and an average dot density of $10^8 cm^{-2}$ are obtained. Large dot diameter and low dot density indicating high surface diffusion for high growth temperatures.

4. Conclusion

We have realized quantum-size InGaAs-dots with a diameter of about 15 nm by MOCVD growth technique. These dots were formed highly uniformly among the GaAs-epilayer. Strong and narrow photoluminescence emission lines of the dot structures demonstrates efficient carrier capture and indicates small dot-size fluctuations and a homogeneous heterostructure interface even at room temperature. The average area dot density can be tuned from $10^8 cm^{-2}$ up to more than $10^{11} cm^{-2}$ and the dot diameter from 30 to 15 nm by substrate off-angle orientation, InGaAs deposition thickness, and growth temperature, respectively. The results indicating high dot density and small dot diameter for low surface diffusion. We hope our approach for the formation of high quality InGaAs-dot structures stimulates further progress in the definition of dots based on MOCVD crystal growth process.

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