

Novel Alternative for the GaAs-based Self-organized Quantum-structure

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***Abstract:** In this work, the evolution of the self-organized III-V based quantum-structures are discussed. These quantum-structures were prepared by droplet epitaxial technique. The droplet-epitaxy is an alternative way for quantum-structure production. Where these structures are formed from Ga droplets during a crystallization process. In all cases, the substrate material was AlGaAs. The different nanostructures such as quantum dot, quantum ring, double quantum ring, or nano hole form similarly from an initial Ga droplet but under different substrate temperature and various arsenic pressure. Started from few atomic courses, we give here a qualitative description of the key processes for all of the forementioned nanostructures.*

***Keywords:** molecular beam epitaxy; III-V materials; nanostructures; droplet epitaxy*

1 Introduction

The low-dimensional structures, grown by molecular beam epitaxy (MBE), has revolutioned the electronic devices both in their potentials and efficiency. Nowadays, the growth of self-assembled nanostructures has been intensively investigated for basic physics and device applications.

These applications can be demonstrated for instance by solar cells (Fig. 1). The efficiency of commercial solar cells ranges between 5 and 15%. A simple pn-junction GaAs solar cell - which material is very suitable for solar energy conversion - has an efficiency of less than 30%. We can fabricate an almost continuously varying band gap solar cell with the help of quantum mechanical effect (this is called band gap tailoring in the literature). Our aim is to prepare quantum wells (QWs) with various widths (2D). The efficiency of a multi quantum well GaAs-based solar cell can exceed 40%. By embedding quantum dots (QDs) in the solar cell material (0D), we produce a so-called intermediate band inside of the band gap. With the help of suitably chosen intermediate band, solar cells can achieve efficiency over 60% [1,2]. Metal induced nanowires (1D)

can also be grown by MBE, where the structure efficiency can improve by many orders of magnitude. The solar cells are typical samples for the application of low-dimensional structures [2]. These nano-structures can revolutionize electronics and computing technology (quantum computing) as well.

Here, we deal with growth kinetics and properties of the low-dimensional nanostructures. The technology used for the growth determines the size, shape and the elementary distribution of the developing structures. These physical parameters are very important in both theoretical and practical point of view. This work is dealing with III-V-based low-dimensional structures (especially with QDs and quantum rings (QRs)), grown by MBE (Fig. 2). MBE is a top technology, where different structures can be grown under controlled conditions and in-situ observation.

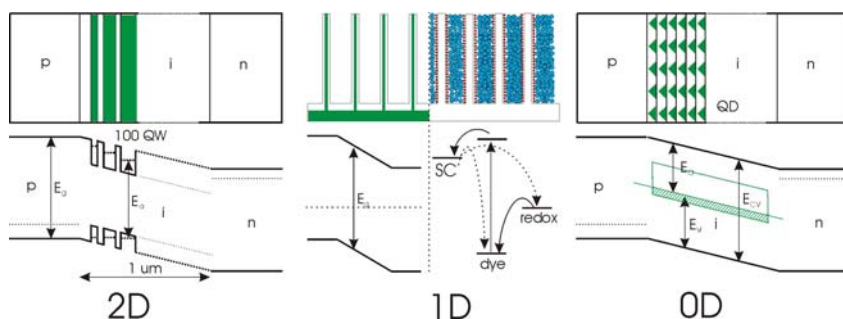


Fig. 1. Enhancement of solar cell efficiency with the help of different low-dimensional structures. (2D): Solar cells with super lattices are the so-called multi-quantum-well solar cells; (1D): left part: solar cells with coaxial nanowires; right part: dye sensitized electrochemical solar cell with bulk junction; (0D): solar cells with quantum dots are the so-called intermediate-band solar cells

It is very important to understand their growth kinetics and to know their shape. For a long time, for zero dimensional system production, the strain-induced method, based on the lattice mismatch, was the only known process. Based on Koguchi's discoveries, a droplet epitaxial technique evolved, giving greater opportunities for the development of the self-organising nanostructures [3,4]. In this method, the lattice-mismatch lost its significance and with the new procedure, it is possible to create quantum dots (QDs) [5-9], quantum rings (QRs) [10-12] (Fig. 2), double quantum ring (DQRs) [13, 14] as well as nano holes (NH) [15-17]. The electronic structure of these nanoobjects depend very much on their shape. Roughly, the droplet epitaxial process is the following: first, metal (e.g. Ga) droplets are generated on the surface in the Volmer-Weber-like growth mode. After, the crystallization of the droplets and their transformation into e.g. GaAs QDs under arsenic pressure. In order to control the process it is necessary to understand the kinetics of the growth process. Here, no theoretical description is available yet of the underlying growth mechanism. Here, we are dealing with the III-V based (e.g. GaAs) structures. The forms of the GaAs nanostructures, grown on AlGaAs (001) substrate, is strongly dependent on the applied technology.

2 Technological Preliminaries

Here, the technological parameters of five different nanostructures are given. Later, the formation kinetics of these nanostructure are qualitatively discussed. The solution process investigated at two cases. The NHs (sample i) was generated at 570 °C in AlGaAs surface applying 6.4 ML Ga [16]. In this case, The AFM measurement shows NHs and very large clusters [16]. The next nanostructure (sample ii) was prepared similarly, but the Ga coverage was different (3.2 ML) [15,16]. Here, the AFM picture shows deep NHs surrounded by ring like bulge formations and shallow NHs, with plane rims (without any bulge) [15,16]. The crystallization process is investigated at further three samples. The first structure is QD (sample iii). The crystallization of the Ga droplet occurs during the high arsenic supply (2.5×10^{-4} Torr) at 150 °C [8]. The elemental map of TEM shows, that the QD had Ga inclusions [8]. The next sample (iv) is crystallized at 2×10^{-6} Torr arsenic pressure at low temperature (200 °C) [13]. We received DQR [13]. The last sample (v) is crystallized at about the same arsenic background (1.1×10^{-6} Torr) but at high temperature (500 °C) [17]. The middle of the structure is surrounded by single ring like bulge formations [17].

3 Discussion

We attempt to give a consistent explanation for the growth of the nanostructures, a mentioned earlier. As widely known, the properties in nano region differ from the bulk properties. Generally, the nano-properties are unknown and only the tendency of the change is established, relative to the bulk properties. Usually the starting and the end states are known. The explanation of the final process comes from the coherent explanations of the component processes. These process explanations are coherent and consistent only, when they describe the growth of a large number of different kind of nano-structures. It becomes only then consistent.

After the deposition of Ga, part of the deposited Ga will combine with the surface arsenic atoms and the rest will form droplets. In order to form droplets at this temperature, the Ga atoms must migrate (process 1) on the GaAs and the Ga surfaces. It is well known, that with diminishing size the melting temperature is dropping and saturation concentration increases. This describes our present Ga droplets well (process 2) (this is the dominant process in cases of complete lack or in the presence small quantities of arsenic). It is also well known that, the crystallization starts at three-phase-line (Fig. 3/A), when the conditions became favourable. In our case, this starts in line of the rim of the droplet (process 3). It is noted, that the excess arsenic incorporates into the GaAs epitaxy when the temperature is low (about 300 °C or lower), creating stress in the lattice. (In presence of larger quantity of arsenic, these are the dominant processes).

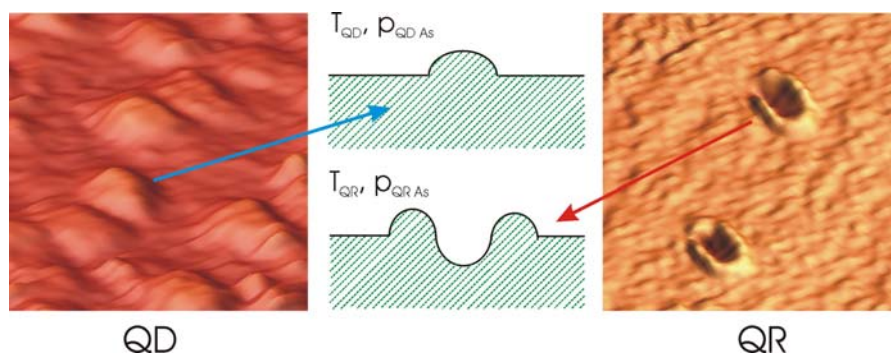


Fig. 2. For nanostructure production, droplet epitaxy is a novel alternative to the strain-induced method; left side: atomic force microscopy (AFM) image of QDs; right side: AFM image of QRs; middle: sketched cross sections of these nanostructures

Process 1 makes possible the phenomenon called Ostwald ripening. At the same time process 2 causes the differentiation between QDs, QRs and NHs. Due to this process at the same temperature and during the same time the hole originated under the smaller droplets is deeper than under the larger droplets (Fig 3./B.). (That is supposing the time duration is not too long to run out of the material from the smaller droplets.) These findings are justified by the two experiments, where the deposited Ga quantity was 6.4 ML (lower part of Fig. 3./B) and 3.2 ML (upper part of Fig. 3./B), respectively. At a given temperature there is a critical droplet size under which the solution begins. After the Ga deposition, droplets form, followed by the growth of the larger droplets in the expense of the smaller ones. When the critical size is reached, the substrate solution by the droplet begins. We start with investigating the case of Ga 6.4 ML [16]. During the experiment, the formation of small NHs and large QDs can be observed (sample i) [16]. We can follow the process on Fig. 3/B, where large quantity of Ga deposited [18]. The sizes of the droplets formed, are above the critical size. After the deposition starts the differentiation of the droplets. The smaller droplets reach the critical size and start solving the substrate. This state is frozen by the opening the arsenic cell.

The second case is when the deposited Ga is 3.2 ML [15,16]. Here, we can observe shallow NHs, with plane rims (without any bulge), and deep NHs surrounded by ring like bulge formations (sample ii) [15,16]. The explanation is given in Fig. 3/B [18]. In this case, the quantity of the deposited Ga is small. The formed droplets are under the critical size, therefore the solution starts under the droplets. Under the small droplets the solution is faster, but the material is used up in a short time. The reduction in material is due first to the solution and second to the material's migration towards the larger droplets. After a short time at the smaller droplets the solution stops, whilst it carries on further under the larger ones. The larger droplets will not be spent, therefore the surrounding ring will freeze after opening the arsenic cell.

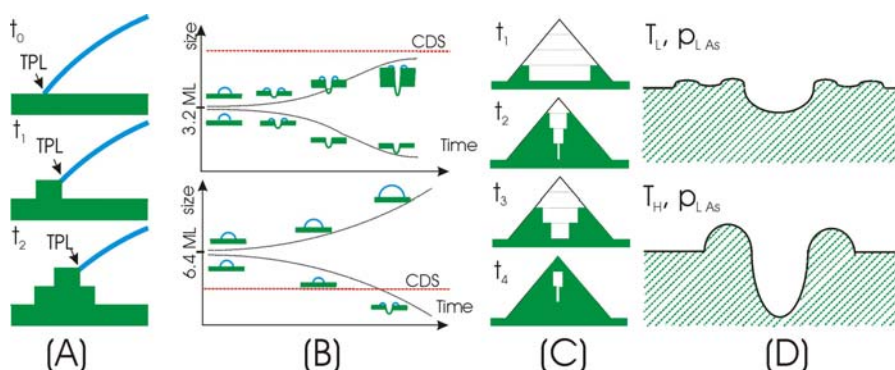


Fig. 3. (A) Explanation of the process 2, (crystal seed formation at the droplet rim) (see text); (B) upper part: Explanation of the temporal differentiation and solution process in case of large amount of deposited Ga (6.4 ML) [16] (see text); lower part: Explanation this process at small Ga amount (3.2 ML) [15,16] (see text); where CDS means critical droplet size; (C) Explanation of the formation of Ga inclusion inside of the QD ($T_{\text{sub}} = 150\text{ }^{\circ}\text{C}$, $p_{\text{As}} = 2.5 \times 10^{-4}\text{ Torr}$) [8] (see text); (D) upper part: cross section of a DQR ($T_{\text{sub}} = 200\text{ }^{\circ}\text{C}$, $p_{\text{As}} = 2.6 \times 10^{-6}\text{ Torr}$) [11]; lower part cross section of a NH ($T_{\text{sub}} = 500\text{ }^{\circ}\text{C}$, $p_{\text{As}} = 1.1 \times 10^{-6}\text{ Torr}$) [17]; where TPL means three-phase-line

Due to the 3rd process the crystallization originates at the rim of the droplets. At lower temperature the excess arsenic incorporates into the lattice. The interstitial arsenic migrates towards the inside of the structure, reducing the lattice stress. A new interface is created in the inner and upper regions of the crystal centres [18]. The growth of these centres are propagating in those directions (Fig. 3./C). The arsenic is fed from the outer surface of the structure. It was observed, that when the arsenic quantity was large the QD had Ga inclusions (sample iii) [8]. Its origin is explained above. The growth in height is similar to that of the basic circle; so the speed of growth in height is a linear function. The speed of growth towards the inner regions is accelerating, because assuming constant arsenic absorbing surface, the concentric area is diminishing with the reduced radius going towards the centre. The presence of the excess arsenic at lower temperature, is the explanation for the formation of the DQRs [18]. The migration and the crystallization of the Ga would result in the creation of a larger ring (upper part of Fig. 3./D, sample (iv)) [13]. Instead, in excess to the original ring we end up with a second ring and this limiting effect on the size of the area is the result of the elimination of the lattice stress. The creation of DQRs can only be observed at lower temperature. At higher temperature DQRs cannot be created because arsenic is not incorporated in the lattice, which causes the stress in the lattice (lower part of Fig.3./D, sample (v)) [17]. Electronic structures of droplet epitaxial QDs and QRs were published elsewhere [19].

Conclusions

The droplet epitaxial method allows us to prepare not only QDs but QRs and other special shaped nanostructures. The development of the MBE droplet epitaxial

technology, which will probably revolutionise the technology of semiconductor devices.

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References

- [1] Á. Nemcsics: Solar Cells and their Developing Perspective; Akadémiai Kiadó, Budapest (2001)
- [2] Á. Nemcsics: New trends in the solar cell technology; Elektronikai Technológia, Mikrotechnika 48 (2009) pp 39-45
- [3] N. Koguchi, S. Takahashi, T. J. Chikyow: New MBE growth method for InSb quantum well boxes; J. Cryst. Growth 111 (1991) pp 688-692
- [4] N. Koguchi, K. Ishige: Growth of GaAs epitaxial microcrystals on S-terminated GaAs substrate by successive irradiation of Ga and As molecular beams; Jpn. J. Appl. Phys. 32 (1993) pp 2052-2058
- [5] T. Mano, K. Watanabe, S. Tsukamoto, H. Fujikoa, M. Oshima, N. Koguchi: New self-organized growth method for InGaAs quantum dots on GaAs(001) using droplet epitaxy; Jpn. J. Appl. Phys. 38 (1999) pp L1009-L1011
- [6] J. M. Lee, D. H. Kim, H. Hong, J. C. Woo, S. J. Park: Growth of InAs nanocrystals on GaAs (100) by droplet epitaxy; J. Cryst. Growth 212 (2000) pp 67-73
- [7] Ch. Heyn, A. Stemmann, A. Schramm, H. Welsch, W. Hansen, Á. Nemcsics: Faceting during GaAs quantum dot self-assembly by droplet epitaxy; Appl. Phys. Lett. 90 (2007) pp 203105 (1-4).
- [8] T. Mano, K. Mitsuishi, Y. Nakayama, T. Noda, K. Sakoda: Structural properties of GaAs nanostructures formed by a supply of intense As₄ flux in droplet; Appl. Surf. Sci. 254 (2008) pp 77707773
- [9] Á. Nemcsics, L. Tóth, L. Dobos, Ch Heyn, A. Stemmann, A. Schramm, H. Welsch, W. Hansen: Composition of the „GaAs” quantum dot, grown by droplet epitaxy; Superlatt. Microstr. 48 (2010) pp 351-357
- [10] T. Mano, N. Koguchi: Nanometer-scale GaAs ring structure grown by droplet epitaxy; J. Cryst. Growth 278 (2005) pp 108-112
- [11] T. Kuroda, T. Mano, T. Ochiai, S. Sanguinetti, K. Sakoda, G. Kido, N. Koguchi: Optical transitions in quantum ring complexes; Phys. Rev. B 72 (2005) pp 205301 (1-8)
- [12] Á. Nemcsics, Ch. Heyn, A. Stemmann, A. Schramm, H. Welsch, W. Hansen: The RHEED tracking of the droplet epitaxial growth quantum dot and ring structures; Mat. Sci. Eng. B 165 (2009) pp 118-121.
- [13] S. Sanguinetti, M. Abbarchi, A. Vinattieri, M. Zamfirescu, M. Gurioli, T. Mano, T. Kuroda, N. Koguchi: Carrier dynamics in individual concentric

- quantum rings: photoluminescence measurement; Phys. Rev. B 77 (2008) pp 125404 (1-4)
- [14] T. Kuroda, T. Mano, T. Ochiai, S. Sanguinetti, T. Noda, K. Kuroda, K. Sakoda, G. Kido, N. Koguchi: Excitonic transitions in semiconductor concentric quantum double rings; Physica E 32 (2006) pp 46-48
- [15] Ch. Heyn, A. Stemmann, W. Hansen: Nano hole formation on AlGaAs surfaces by local droplet etching with gallium; J. Cryst. Growth 311 (2009) pp 1839-1842
- [16] Ch. Heyn: Kinetic model of local droplet etching; Phys. Rev. B 83 (2011) pp 165302 (1-5)
- [17] Zh. M. Wang, B. L. Liang, K. A. Sablon, G. J. Salamo: Nano holes fabricated by self-assembled gallium nanodrill on GaAs (100); Appl. Phys. Lett. 90 (2007) pp 113120-3.
- [18] Á. Nemesics: To the understanding of the formation of the droplet epitaxial III-V based nanostructures; Proc. of the 7th Int. Conf. on Low-dimensional Structures and Devices; Techac, Nuevo Yucatan, Mexico 22-27 May 2011 (under publication)
- [19] Á. Nemesics, J. Balázs, B. Pődör, J. Makai, A. Stemmann: Photoluminescence studies of GaAs quantum dots and quantum rings; Phys. Stat. Sol. C, 8 (2011) pp 2826-2829