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Quadra-Quantum Dots and Related Patterns of Quantum Dot Molecules: Basic Nanostructures for Quantum Dot Cellular Automata Application

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1. Introduction

The concept of quantum dot cellular automata (QCA) was proposed by C. S. Lent et al., in 1993 based on current switch with a cell having a bi-stable charge configuration representing either "1" or "0" in a binary system. This function is operated without current flow into or out of the cell. The basic mechanism is "Coulomb repulsion force" among

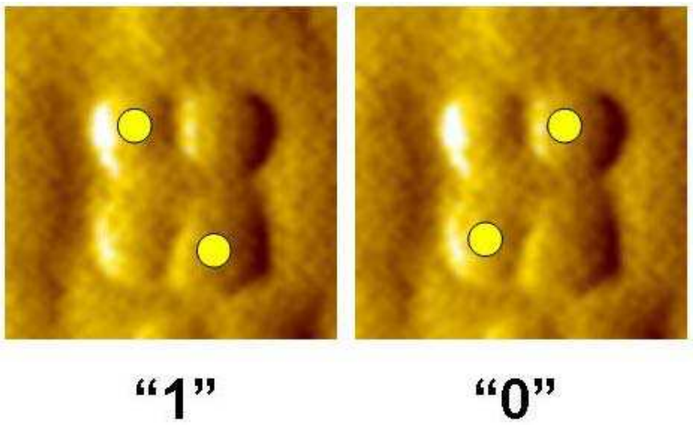


Fig. 1. Basic QAC cells represent digital data "1" and "0"

distributed charges in the individual QCA cells. Each charge configuration provides the field from one QCA cell to adjacent cells. The transfer of charge configuration, therefore, consumes very small power and occurs at a very high speed. QCA approach would break the limitation in shrinking of CMOS technology, which is normally based on "top-down technology". Quantum dots (QDs) in QCA cells are created by self assembly approach, which is "bottom-up technology". Each quantum dot in a QCA cell localizes charge because of its zero dimensional nanostructure. However, the barrier between dots has to be narrow enough so that a charge can quantum mechanically tunnels from one dot to another.

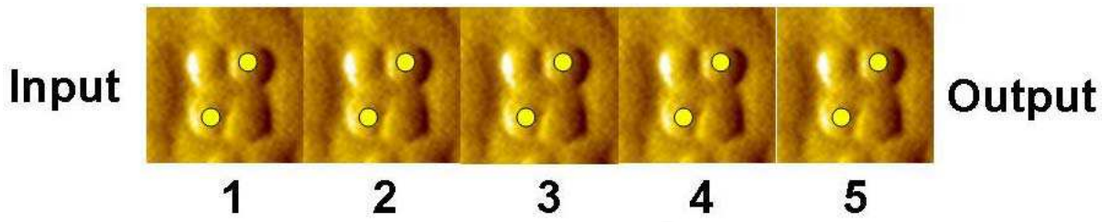


Fig. 2. Schematic display of QCA wire transferring data at high speed and low power consumption

Quantum dots are technically created from several semiconductor materials having lattice mismatch like InAs/GaAs (7% lattice mismatching). When InAs is grown on GaAs at monolayer (ML) scale by molecular beam epitaxy (MBE), strain is created at the interface. At a critical thickness of InAs called wetting layer, strain relaxation occurs and leads to quantum dot formation (J. Timler et al., 2003). The semiconductor quantum dots grown by self-assembly are defect free and provide good electrical and optical properties. They are potential nanostructures for several electronic and photonic device applications. Self-assembly of QDs is natural process. Therefore, the QD creation sites are random. QD size is not uniform. However, when the MBE growth parameters are precisely controlled, QD uniformity is improved and is applicable for many devices. The dot sizes and dot separations are normally in the order of tens of nanometers.

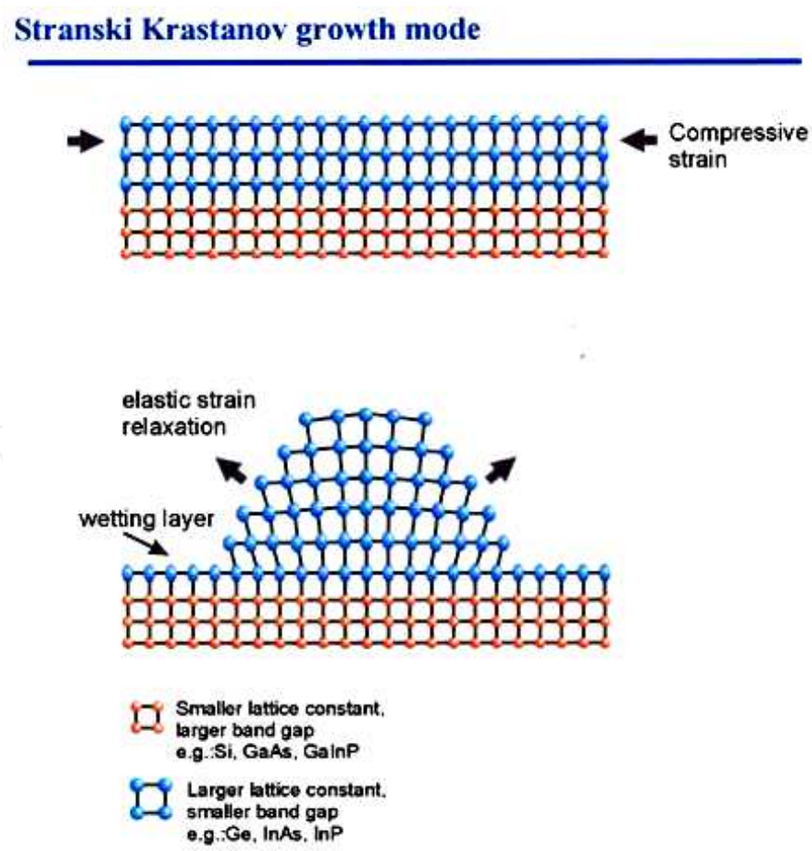


Fig. 3. Quantum dots grown by strained semiconductors

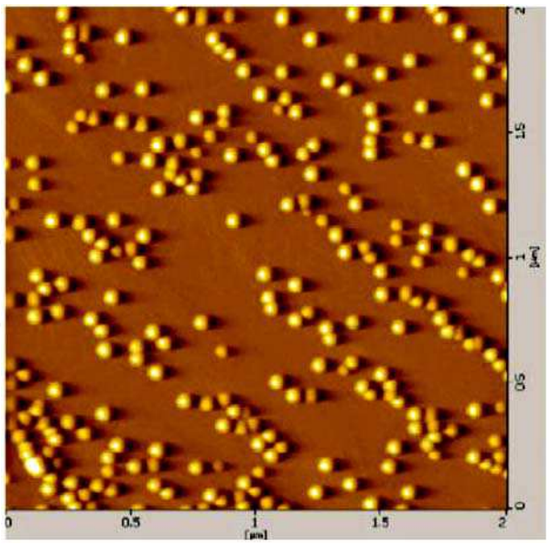


Fig. 4. Self-assembled quantum dots are naturally random.

The design feature of QCA cell is quadra-quantum dots (QQDs), which are composed of two stable orientations. Electrons in these two orientations are used to represent the two binary digits "1" and "0". The four dots in a QCA cell are at the corners of a square having two mobile charges. These two mobile charges or electrons can quantum mechanically tunnel between dots but not between cells.

QCA concept is performed on different patterns of QCA cells. Linear array of QCA cells works as binary wire where Coulomb interaction induces all QCA cells in the wire into the same polarization. Therefore, QCA data can be transferred from one end of the wire to the other. Corner interaction of QCA cells having anti-voting can perform as an inverter, where polarization changes from one diagonal direction to another or alters QCA data from "1" to "0" or from "0" to "1". Logic gates having three inputs and one output are realized by using a cross-over of QCA cells. They can work like a majority gate. Combinations of different QCA patterns are used in the design of QCA memory bits (Y. Lu et al., 2007). QCA is a promising tool for future computation using semiconductor quantum nanostructures.

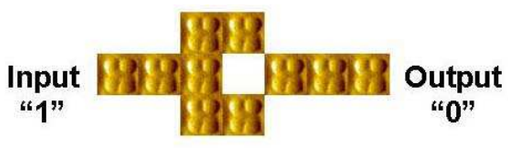


Fig. 5. Schematic display of QCA inverter changing data from "1" to "0" or from "0" to "1"

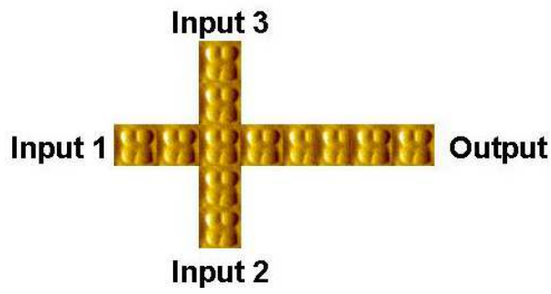


Fig. 6. Schematic display of a QCA logic working as a majority gate

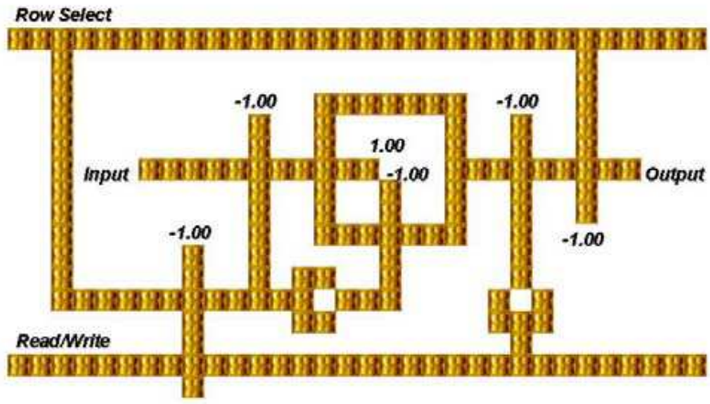


Fig. 7. Schematic design of a QCA circuit having one bit memory

2. Semiconductor quantum nanostructures

2.1 Quantum dot molecules

Self-assembled quantum dots are widely studied due to their unique properties such as zero dimensionality and the resultant delta-function energy states (O. Suekano et al., 2002, L.G. Wang et al., 2000, K. Jacobi et al., 2003). These properties lead to an improved device performance, such as quantum dot lasers with an ultra-low threshold current density (M. Asada et al., 1986, Y. Arakawa et al., 1982) and single electron devices with a high speed response and a low power consumption (Y. Ono et al., 2005). Self-assembly of quantum dots using various semiconductor systems including strained InAs/GaAs, always results in random dot formation with a variation of dot size (D. Leonard et al., 1993, T. v. Lippen et al., 2005). In many applications, it is desirable to have uniform quantum dots, and, thus, there is a need to develop some growth techniques which can provide some degree of dot alignment and uniformity. In order to obtain aligned dots, many approaches have been proposed and demonstrated (Z. M. Wang et al., 2004, T. Mano et al., 2004, Z. M. Wang et al., 2004), but those based on self-assembly are preferred over other techniques due mainly to the simplicity of the processes involved.

It has been demonstrated that thin capping of quantum dots provides anisotropic strain fields which lead to the elongation of capped nanostructures after annealing (S. Suraprapapich et al., 2005). The regrowth of quantum dots on elongated nanostructures results in aligned quantum dots and aligned quantum dot molecules (QDMs).

Lateral quantum dot molecules are closely packed quantum dots in the growth plane. Quantum dot molecules have several unique features, i.e., uniform dot sets with specific patterns, high dot density, and anisotropic nanostructure (T. v. Lippen et al., 2004, T. v. Lippen et al., 2005, W. Sheng et al., 2002). Therefore, they are potential candidates for nanoelectronic applications, such as that in quantum dot cellular automata because of their potential in obtaining four quantum dots arranged in a rectangular pattern (C. S. Lent et al., 1993). Lateral quantum dot molecules can be realized by several growth techniques (R. Songmuang et al., 2003, T. Mano et al., 2004). The thin-capping-and-regrowth molecular beam epitaxial process, which can be used to obtain quantum dot molecules in one continuous growth, has been demonstrated (S. Suraprapapich et al., 2005). Multiple cycles of the thin-capping-and-regrowth process of the quantum dots give improved dot alignment

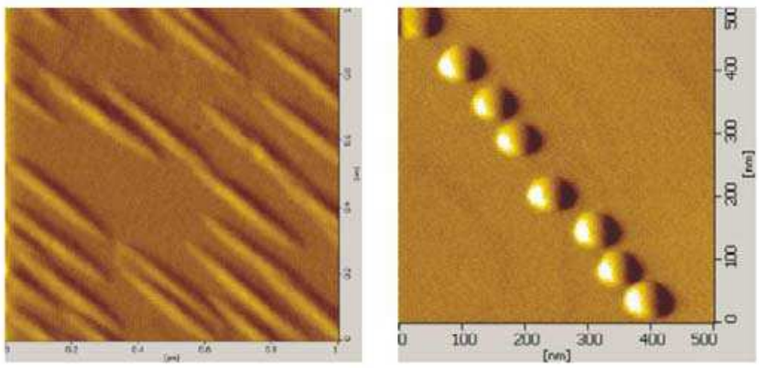


Fig. 8. Aligned quantum dots grown on elongated nanostructures after thin capping of as-grown quantum dots

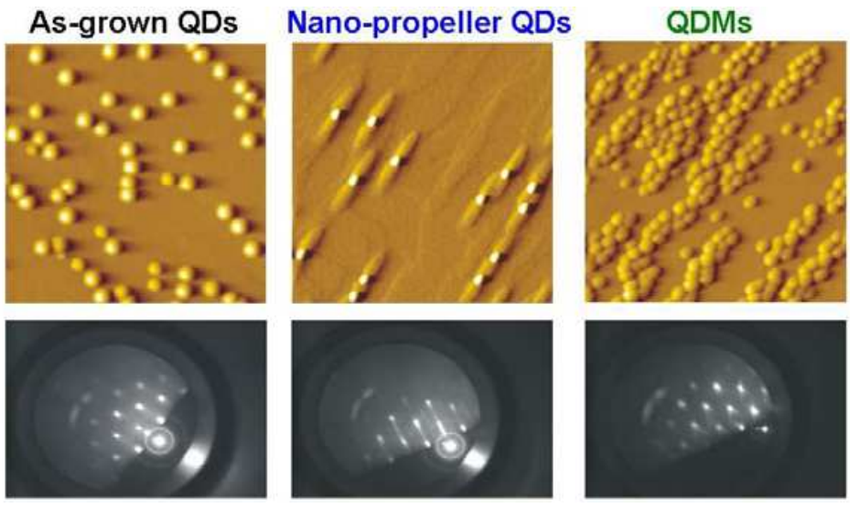


Fig. 9. As-grown QDs are transformed to nano-propellers and QDMs respectively after thin-capping-and-regrowth process as confirmed by RHEED patterns

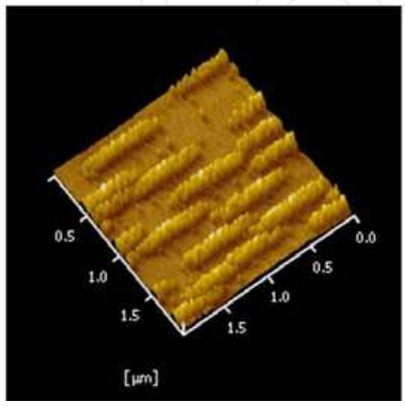


Fig. 10. Laterally close packed quantum dot molecules grown by thin-capping-and-regrowth MBE process

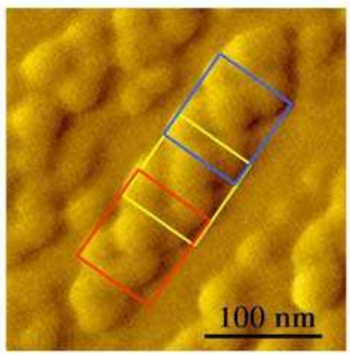


Fig. 11. Chain of overlapping quantum dot molecules grown by multiple thin-capping-and regrowth process

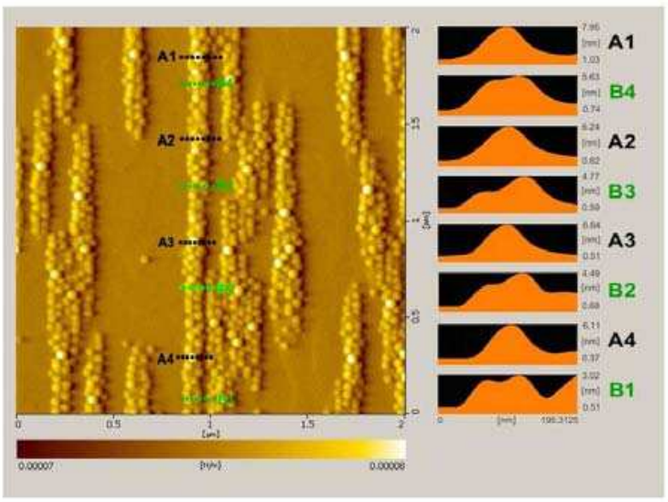


Fig. 12. Long chain of QDMs

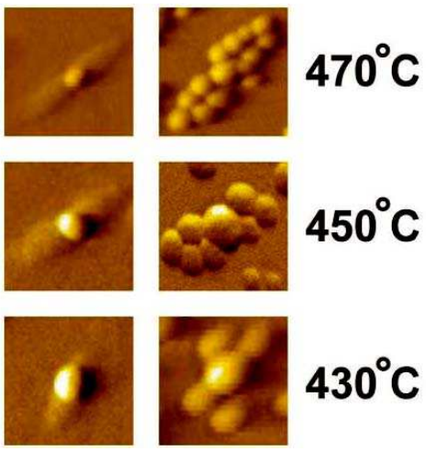


Fig. 13. Dot number per quantum dot molecule is controlled by different capping temperatures of as-grown quantum dots

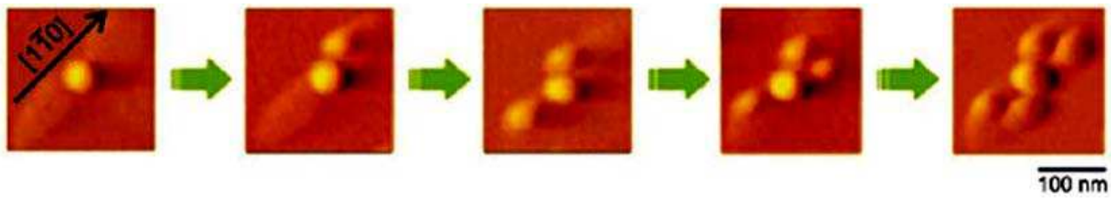


Fig. 14. Formation of 4 satellite dots on nano-propeller template along [1$\bar{1}$0] crystallographic direction

up to a certain number of growth cycles. In addition, with aligned quantum dots as templates, aligned quantum dot molecules can be obtained by this growth technique.
The dot number in quantum dot molecules grown by thin-capping-and-regrowth MBE process can be controlled by varying the capping temperature and the capping thickness (N. Siripitakchai et al., 2007). Quantum dot molecules with a small number of quantum dots per quantum dot molecule ensemble can be grown and used as a basic building block for quantum computation in accordance with the quantum dot cellular automata principle (M. Macucci et al., 2006). A quantum dot molecule with four to five dots per molecule can be grown with GaAs capping thickness and InAs regrowth thickness of 25 ML and 1.5 ML respectively (N. Siripitakchai et al., 2008). However, nonlinear strain distribution originated from underlying templates results in the center dot and satellite dots of each molecule having different dot size. By increasing the regrowth temperature, an optimized condition is found to reduce the size difference between the satellite dots and the center dot. By repeating the quantum dot molecule growth cycles several times, certain degree of dot alignment is obtained. An alignment of quantum dots is used as a template in creating aligned quantum dot molecules at the topmost surface albeit with increased size difference between the center dot and the satellite dots.

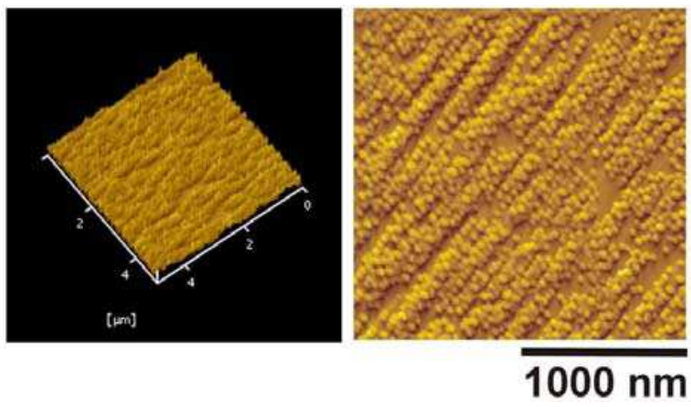


Fig. 15. High density quantum dot molecules is preferable for quantum dot solar cell application

2.2 Quantum dot alignment and cross-hatch

The statistical nature of quantum dot formation by conventional Stranski-Krastranow mode results in random dot position. In some applications, such as in quantum dot solar cells, the random dot positions and uniformity are not of major concerns, as long as dot density is high (S. Suraprapapich et al., 2006). Yet in certain applications, such as super luminescent

light emitting diodes, quantum dots are preferred to be large and non-uniform for efficient light emission (C. Y. Ngo et al., 2007). But in quantum dot based quantum cellular automata (C. S. Lent et al., 1997) and bit-patterned media (B. D. Terris et al., 2005), the positions of active elements or quantum dots must be deterministic.

In order to develop a growth technique for aligned quantum dots and cross-over pattern of aligned quantum dots, which are useful in the design of various functions of quantum dot cellular automata, self-assembled InAs quantum dots are grown on cross-hatch GaAs/InGaAs templates via molecular beam epitaxy with controlled parameters such as degree of excess growth, growth rate and capping of the quantum dot layer (S. Kanjanachuchai et al., 2009). The InAs quantum dots are grown on an InGaAs cross-hatch layer without any excess growth, the dot alignments are formed both along the $[110]$ and $[1\bar{1}0]$ directions as a result of chemical potential gradient and anisotropic strain fields. When the underlying InGaAs cross-hatch layer is covered by a thick GaAs spacer layer, subsequent growth of InAs quantum dots results in preferential alignment of quantum dots along either the $[110]$ or $[1\bar{1}0]$ direction, respectively.

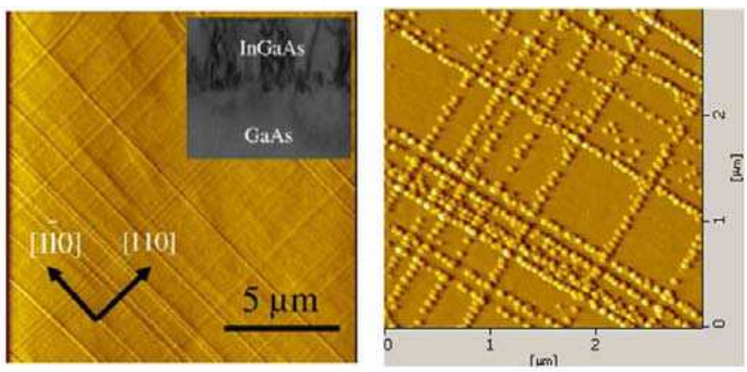


Fig. 16. Cross hatched substrate leads to quantum dot alignment and cross hatch

The MBE growth technique using cross-hatch substrates is a method for the self-assembly of dot alignment either in $[110]$ or $[1\bar{1}0]$ direction and the cross-over of quantum dot lines in perpendicular configuration, i.e., in the $[110]$ and $[1\bar{1}0]$ directions. This bottom-up approach is useful for the design of different QCA building blocks for various QCA applications.

2.3 Quantum Rings and Bi-Quantum dot molecules

Quantum dot molecules, in the form of a pair of vertically coupled quantum dots, are proposed and demonstrated to be a quantum gate controlling qubits in quantum computation (M. Bayer et al., 2001). In vertically coupled quantum dots, the upper self-assembled quantum dots are formed on top of the lower self-assembled quantum dots due to the strain field around the lower quantum dots. The vertical separation between the upper and the lower quantum dots controls the degree of coupling. In principle, however, laterally coupled quantum dots are preferred because they allow a large number of quantum gates in a two dimensional array on the surface (G. J. Beirne et al., 2006). Such lateral quantum dot molecules are more applicable-wise. Lateral quantum dot molecules have been achieved by several growth techniques, such as a combination of in situ etching and self-assembly (S. Kiravittaya et al., 2003), self-assembly by anisotropic strain

engineering on an InGaAs/GaAs (311 B) super-lattice templates and droplet epitaxy (J. H. Lee et al., 2006).

An uninterrupted molecular beam epitaxy process comprising partial capping and regrowth to obtain lateral quantum dot molecules within a single growth run has been developed. The key processing step is the partial capping of InAs quantum dots by thin GaAs layer. The surface morphology after the capping process is affected by several parameters such as as-grown quantum dot size and capping temperature. The same growth process is used in both solid source and gas source molecular beam epitaxies. Dramatically different lateral quantum dot molecules are obtained. As-grown quantum dots are transformed to camel-like nanostructures when As4 overpressure from a conventional Arsenic solid source is used in the molecular beam epitaxy machine. But when As2 overpressure from a gas source is used, quantum rings (QRs) are formed. The Arsenic species is a crucial parameter to in forming different shapes of nanostructures (S. Suraprapapich et al., 2007).

Under As2 overpressure, the migration length of Indium adatoms is shorter than under As4. Therefore, the shape of transformed nanostructures is less anisotropic, resulting in InGaAs quantum ring formation after the partial capping of as-grown InAs quantum dots. The surface migration of Indium adatoms, however, is still anisotropic, leading to higher Indium concentrations on the two regions of the InGaAs quantum ring in the $[1\bar{1}0]$ direction. At these particular parts of InGaAs quantum ring, strain becomes higher. When the amount of deposited InAs is increased during the regrowth process over InGaAs quantum ring template, the strain at these two regions relaxes, leading to the formation of InAs quantum dots which become a bi-quantum dot molecule (Bi-QDM). Bi-quantum dot molecules are formed on strained InGaAs quantum ring nanostructures; therefore, the amount of InAs required in bi-quantum dot molecules is less than that required on the flat GaAs surface. The formation of bi-quantum dot molecules starts at an InAs thickness of 0.6 ML compared to 1.8 ML InAs quantum dots conventionally grown on flat GaAs buffer layer. Hence, the individual dots making up the bi-quantum dot molecule are smaller than those of as-grown quantum dots.

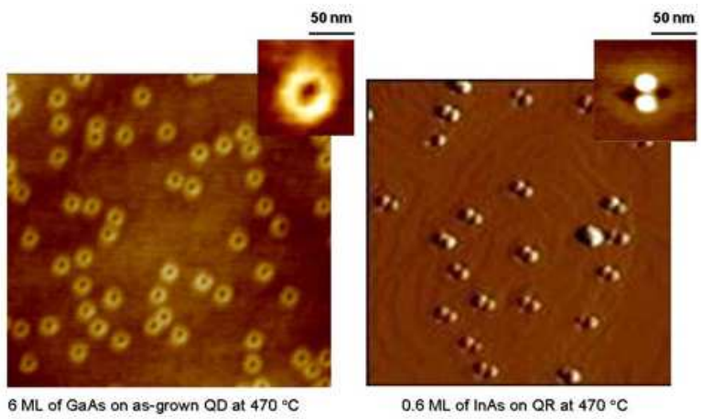


Fig. 17. Quantum rings and bi-quantum dot molecules grown by As_2 gas source MBE

Bi-quantum dot molecules are useful basic nanostructures for spintronic applications. The electron spins in coupled quantum dots of a bi-quantum dot molecule can work as a spin

qubit, spin-up and spin-down. The semiconductor material of bi-quantum dot molecules needs to be magnetic semiconductor like GaMnAs (K. S. Ryu et al., 2008, U. Wurstbauer et al., 2008, M. Bozkurt et al., 2010) for practical use in spintronics.

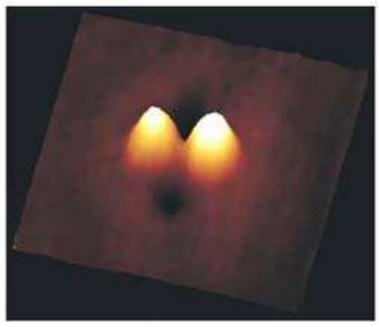


Fig 18. Bi-quantum dot molecule grown on quantum ring

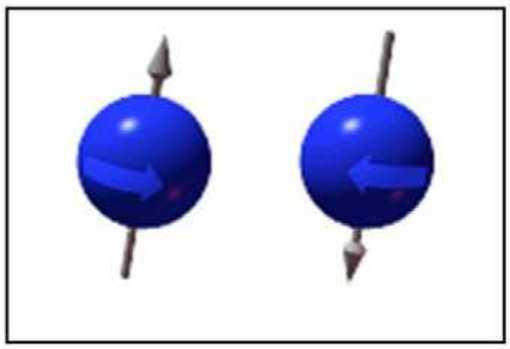


Fig. 19. Electron spins in bi-quantum dots work as a qubit

When InGaAs quantum rings are created after the partial capping of as-grown InAs quantum dots by As2 overpressure in gas source MBE, InAs bi-quantum dot molecules are achieved by the regrowth process at a low temperature of 470°C. But when the substrate temperature is increased to as high as 520°C, the number of quantum dots on a quantum ring becomes five to seven quantum dots per ring. This is how to form another InAs quantum nanostructure called quantum dot rings (QDRs).

Quantum dot rings

Quantum dot rings are created on the quantum ring templates with less anisotropy. InAs quantum rings always have unsymmetrical shapes due to high migration of Indium adatoms in Arsenic environment. When the quantum dot material is changed from Indium Arsenide (InAs) to Indium Phosphide (InP), Indium adatoms in Phosphorous environment change and become less migrating. This leads to InP ring shaped quantum dot molecules.

Instead of using Stranski-Krastranow growth mode, self-assembled InP ring shaped quantum dot molecules can be fabricated by solid source molecular beam epitaxy using the droplet epitaxy technique. This droplet epitaxy involves two processes, i.e., the deposition process of group III elemental droplets without the presence of group V element and the crystallization or incorporation process of group V element into droplets to form the III-V nanostructures (T. Mano et al., 2005, T. Mano et al., 2005). InP is chosen as a material for

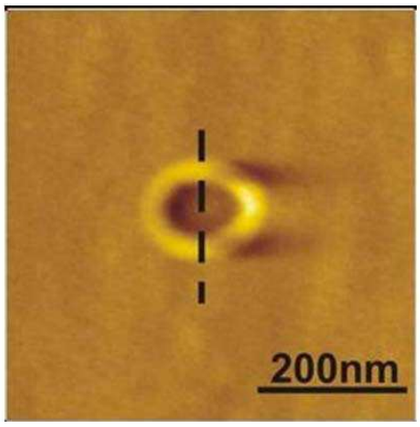


Fig. 20. InGaAs quantum ring grown by droplet epitaxy

quantum dot structure because of its lattice mismatch with In0.49Ga0.51P layer. In addition, droplet epitaxy technique can provide the circular ring shaped structure due to isotropic migration property of Indium adatoms under P2 pressure on the In0.49Ga0.51P layer during crystallization process (W. Jevasuwan et al., 2007). Therefore, the droplet epitaxy technique is favorable for the growth of ring shaped quantum dot molecule structure. In droplet epitaxy growth, the initial dimension of the Indium droplets, the Phosphorous atom migration and diffusion process determine the size and shape of the Indium Phosphide nanostructure. It is found that the crystallization temperature can affect the quantum dot size and density on the ring as well as the ring size and density (W. Jevasuwan et al., 2010). At a high crystallization temperature, the quantum dots on the ring and the ring itself become bigger; meanwhile the dot density as well as ring density is decreased. The explanation for this is that, with high crystallization temperature, initial Indium droplets can efficiently incorporate with each other and Indium adatoms can migrate farther from the center of droplets to minimize the energy of the system; thus, bigger InP quantum dots and bigger InP ring shaped quantum dots molecules are obtained.

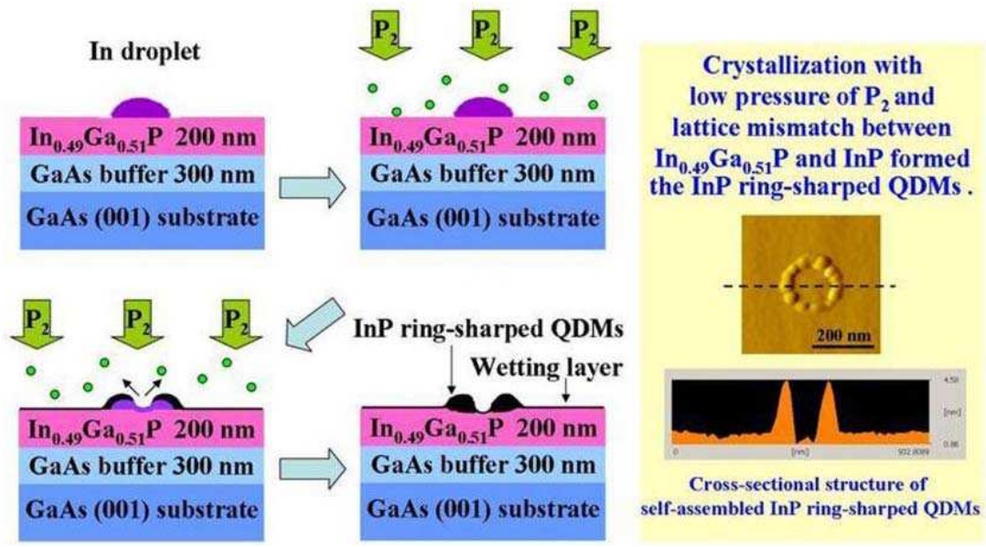


Fig. 21. Formation mechanism of InP quantum dot rings by droplet epitaxy

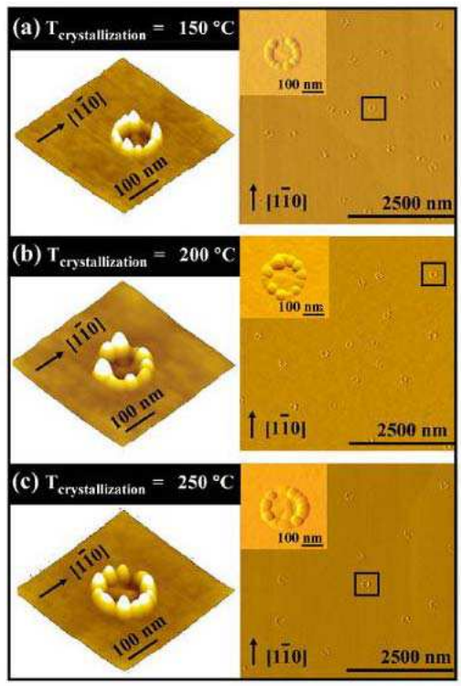


Fig. 22. InP quantum dot rings grown at different crystallization temperatures

This ring shaped quantum dot molecules called quantum dot rings (QDRs) have an interesting feature when the number of quantum dots on each ring is controlled. Quantum dot rings with eight dots are useful in extended quantum dot cellular automata (I. L. Bajec et al., 2006) where three encode values of "1", "0" and "1/2" are possible. By adding

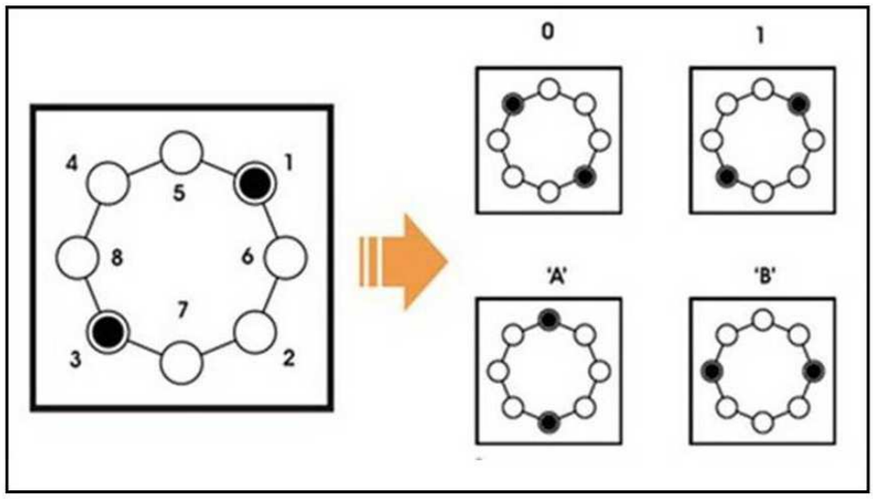


Fig. 23. Octa quantum dot molecule with ring-shape gives 3 logic values of "1", "0" and "1/2" in extended quantum cellular automata

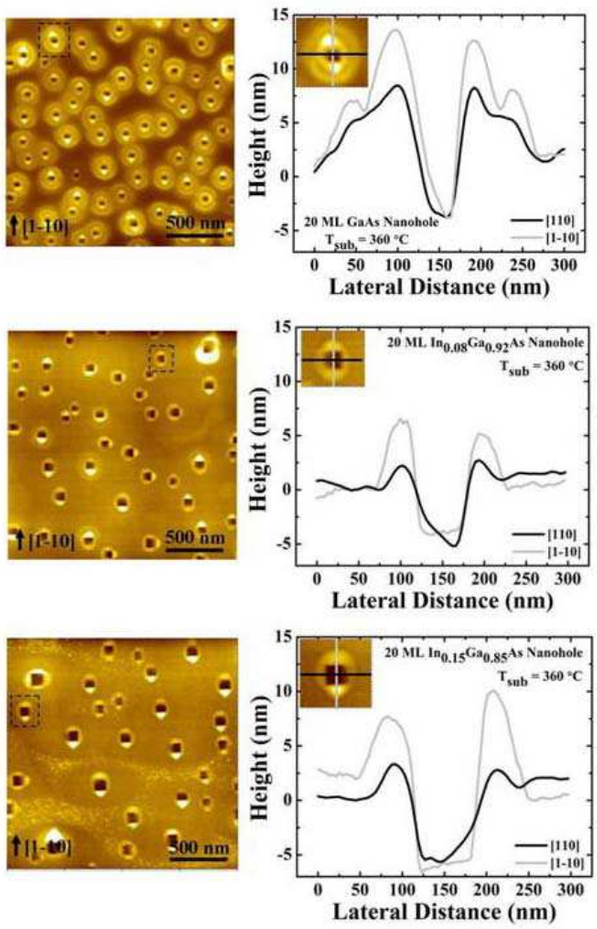
four more quantum dots to classical four quantum dot sets, quantum dot cellular automata can be extended to enlarged range of states. This leads to wider aspects of future quantum computation based on quantum dot nanostructures.

3. Quadra quantum dots

Quantum rings can be created by droplet epitaxy both in InAs and InP material systems. Nanoholes at the center of the quantum rings are used as templates for several patterns of quantum dot molecules. Several approaches using artificial substrate process methods, such as atomic force microscope (AFM) tip and scanning tunneling probe-assisted nanolithography have been developed to obtain nanohole templates for growing quantum dot molecules (J. S. Kim et al., 2006, S. Kohmoto et al., 1999, E. S. Moskalenko et al., 2005). These methods, however, require complicated and expensive substrate processing equipment and are also prone to defects and contamination. A combination of two different techniques is developed to create quantum dot molecules from quantum ring templates. The first technique is droplet epitaxy, which is used to grow InGaAs quantum ring structures having non-uniform ring stripes and deep square shaped nanoholes. The second is conventional Stranski-Krastanow growth mode, which creates four InAs quantum dots on the InGaAs nanoholes. Combining these two techniques has helped overcoming a few of limitations of individual technique, thus leading to a novel fabrication of four quantum dots in one quantum dot molecule.

When Indium-Gallium (In-Ga) droplets are used in the droplet epitaxy and become crystallized by arsenic pressure, quantum rings with square shaped nanoholes can be achieved at a specific growth condition (P. Boonpeng et al., 2010). The square shape of nanoholes is oriented along both the $[110]$ and $[1\bar{1}0]$ crystallographic directions. Most of the square-like nanoholes exhibit a V-shape profile along $[110]$ and a U-shape profile along $[1\bar{1}0]$. The difference in shape profiles is due to anisotropic behavior of the atomic diffusion from the center of the In-Ga droplet under As_4 flux during crystallization of the ring structure. The substrate temperature (330-390°C) during droplet deposition process is a key parameter for controlling the size of nanoholes. At a high substrate temperature of 390°C, larger In-Ga droplets are deposited and transformed to bigger quantum rings with larger nanoholes. Consequently, anisotropic behavior is pronounced leading to a difference in the height of ring lobes between those along $[110]$ and $[1\bar{1}0]$. The bigger nanoholes at 390°C give higher ring lobes along $[1\bar{1}0]$, nearly double compared with the shorter ring lobes along $[110]$. It is also clear that nanohole density is reduced at a higher temperature during droplet deposition in which small droplets are merged into large droplets having a small number of transformed nanostructures per unit area. Despite the larger nanoholes at high substrate temperature, the hole-depth is slightly shallower at 4nm. It is found that nanoholes prepared at 360°C are the most uniform with smallest deviation in their dimension. The formation mechanism of square-like nanoholes is based on the As4 diffusion in InGa droplets during the supply of As4 flux (P. Boonpeng et al., 2009).

Square-like nanoholes have a high strain at each corner of the nanostructure. Therefore, when quantum dots are grown on this square-like nanohole template, four quantum dots called quadra quantum dots (QQDs) are created. In the fabrication process of quadra quantum dots, the substrate temperature is increased to 450°C. InGaAs square-like nanoholes are transformed to the InGaAs nano-mounds. Then, InAs quadra quantum dots are grown on InGaAs nano-mound at the substrate temperature of 450°C. It is noticeable that quadra quantum dots in each group are not uniform in dot size depending on crystallographic directions in which respective lobes around nanohole templates prior to quantum dot growth are not the same in volume and shape.



(a) Double quantum rings grown by using GaAs droplet epitaxy

(b) In0.08 Ga0.92As quantum rings with square-shaped nanoholes

(c) $In_{0.15}Ga_{0.85}As$ quantum rings with deep square-shaped nanoholes

Fig. 24.

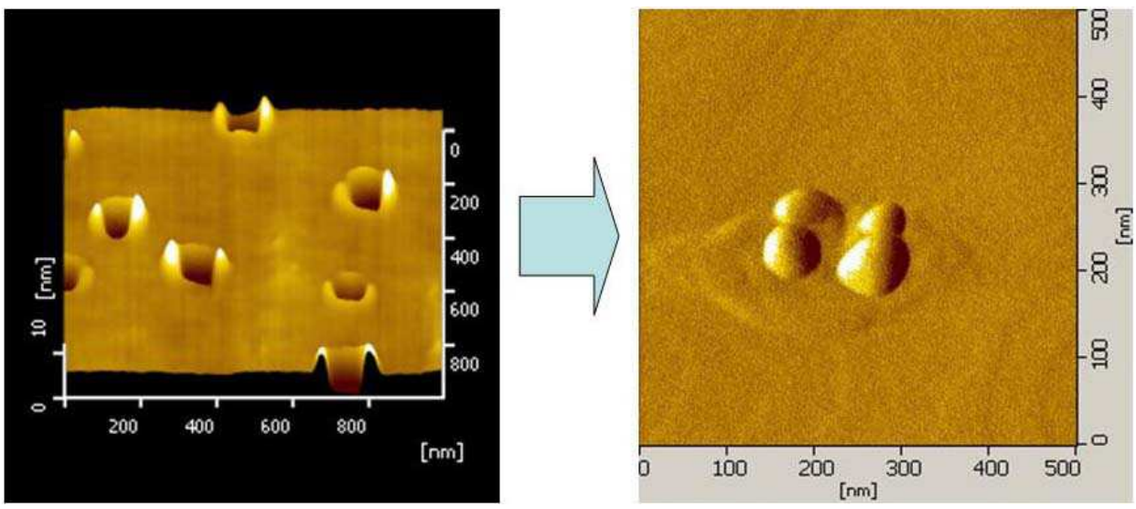


Fig. 25. Square-shaped nanoholes are used as templates for the formation of quadra quantum dots

These square shape quadra quantum dots can be used as a basic cell of quantum dot cellular automata in which two electrons are localized at two quantum dots along either diagonal direction representing either "1" or "0" in a qubit system. However, application-wise, uniform quadra quantum dots are required. Therefore, the square-like nanohole templates of which the holes exhibit the U-shape profile along both directions will be key nanostructures providing uniform quantum dot molecules. In addition, uniform square-like nanoholes with good alignment and cross-hatched are also required for practical design of QCA cells. Overlapping of nano-mounds along the $[1\bar{1}0]$ crystallographic direction is a demonstration of aligned quadra quantum dots by self-assembly approach. In engineering point of view, though, the self-assembly of nanostructures is a simple approach and needs few sophisticated fabrication instruments. The precise control of quantum dot molecules at designated sites is very important and makes quantum computation workable. Therefore, the final approach would be the combination of "bottom-up" and "top-down" at optimal requirement.

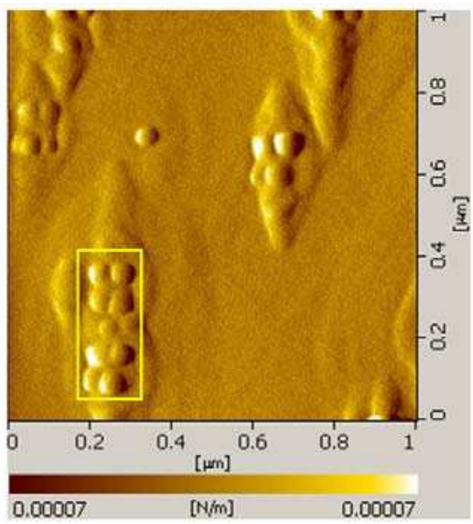


Fig. 26. Two sets of quadra quantum dots are aligned along $[1\bar{1}0]$ by oriented templates

4. Summary

Self-assembly of InAs quantum dot molecules with different features fabricated by the combination of conventional Stranski-Krastanow growth mode and modified MBE technique using thin or partial capping as well as droplet epitaxy has been reviewed. Quantum dot alignment and cross-hatch are demonstrated by using modified InGaAs/GaAs substrates. Partial capping of as-grown quantum dots leads to an elongation of the nanostructure having nanoholes at the original sites of the dots. Under some specific conditions, i.e. dot materials, growth parameters, these techniques can provide quantum ring nano-templates for InAs bi-quantum dot molecules and InP quantum dot rings. InGaAs quantum rings with square shaped nanoholes are realized by droplet epitaxy. They are utilized as nano-templates for quadra quantum dot molecules where four InAs quantum dots are situated at the four corners of a square. This quadra quantum dot set is a basic QCA

cell for future quantum computation. For practical use, precise control of quantum dot molecules both in their dot uniformity and dot sites needs further research and development.

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Modelling and simulation are disciplines of major importance for science and engineering. There is no science without models, and simulation has nowadays become a very useful tool, sometimes unavoidable, for development of both science and engineering. The main attractive feature of cellular automata is that, in spite of their conceptual simplicity which allows an easiness of implementation for computer simulation, as a detailed and complete mathematical analysis in principle, they are able to exhibit a wide variety of amazingly complex behaviour. This feature of cellular automata has attracted the researchers' attention from a wide variety of divergent fields of the exact disciplines of science and engineering, but also of the social sciences, and sometimes beyond. The collective complex behaviour of numerous systems, which emerge from the interaction of a multitude of simple individuals, is being conveniently modelled and simulated with cellular automata for very different purposes. In this book, a number of innovative applications of cellular automata models in the fields of Quantum Computing, Materials Science, Cryptography and Coding, and Robotics and Image Processing are presented.

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