Chapter from the book *Nonmagnetic and Magnetic Quantum Dots*

Chapter 3

Droplet Epitaxy as a Tool for the QD-Based Circuit Realization

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Additional information is available at the end of the chapter

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Abstract

The chapter describes a novel technology, called droplet epitaxy, in the view point of quantum-circuit realization. This technology is useful when quantum dots are to be produced, of different shape and size in various densities. There are self-assembling methods to achieve spatial ordering or spatial positioning. Out of some of the possible applications as an example, the register and cellular automata circuit will be described.

Keywords: droplet epitaxy, quantum dot, self-assembling, lateral alignment, vertical stacking

1. Introduction

The most frequently quoted integration tendency in microelectronics is covered by the so-called Moor's law, which predicts the growth of the component concentration on microchips, doubling in every 2 years and forecasting further miniaturization. The CMOS technology itself is approaching its theoretical limits. Further limitations are also caused by quantum effects and certain anomalies in the technology in materials science. The spread in size, when CMOS technology is approaching the nano-region, represents further problems in microchip design. These difficulties make us wander about the next step in microchip technology, which would follow the present CMOS technology. The answer is hidden either in the promising state of spinotronics, an electronics based on graphene, or in circuits based on Josephson junction [1–4]. Quantum dots (QDs) or groups of QDs are also possible candidates of a new technology for the creation of electronic circuitry (**Figure 1A**). Nanotechnology based on GaAs and related compounds are also the most likely candidates for the development of new technology.

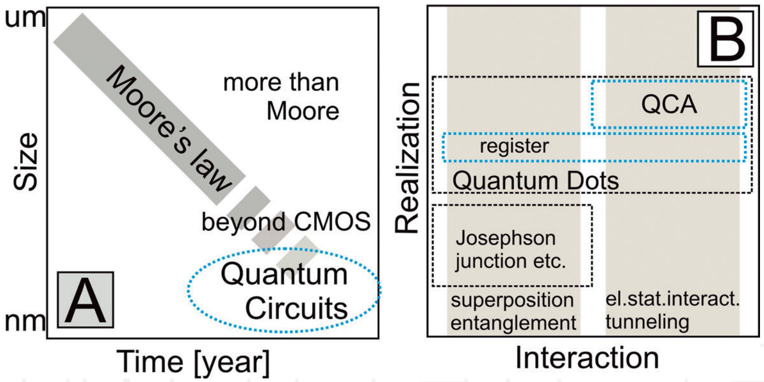

Figure 1. (A) Quantum circuit is a possible solution beyond CMOS technology; (B) realization of quantum circuits such as register or quantum dot cellular automata (QCA). The quantum dot-based realization can bridge the bit- and qubit-based circuits.

The work of quantum circuits (like quantum computing) is based on quantum mechanical phenomena, so the realization must be in a nanometer scale. In this field, one of the promising candidates is the QD-based technology. The QD-based computing technology fundamentally differs from earlier systems. Conventional digital computing technology uses voltage values to represent binary states. By contrast, QD-based computing system uses the position of electrons in QDs to represent binary states. Here, we can distinguish two main types according to the interactions. One of them utilizes superposition and entanglement, and another one utilizes electrostatic interaction and tunneling. For the computation, the first one uses the so-called qbits.

The quantum computer uses the quantum states to encode and process information. The unit of quantum information is the qubit, which can be shown as a two-stage system such as a QD. Opposite to some classical object, a quantum system can exist not only in the ground state |0 > or the excited state |1>, but in some linear superposition of these two stages. The possibility of the handling of these stages provides the main advantage of quantum computing [5]. One type uses the charge of an electron to form a qubit. The qubit realization is possible by single or two electron QDs. In close neighbor, two semiconducting QDs can be coupled with each other. They spatially confine an individual charge carrier in a discrete energy level, interact quantum mechanically with each other. The ordered QD pair ensemble system offers the potential of implementing tunable qubit arrays. The utilization of the ordering of charge-coupled QDs enables to realize also quantum circuits with utilization of classical bit. One of them is called as computational register and the other one is memory register, respectively [6, 7]. One of the main tasks of the quantum computer is the encoding of the qubit. The QD-based circuits can bridge the qubit- and bit-based circuits.

In this chapter, a very novel technology, called droplet epitaxy (DE), will be discussed in the applicational view point. What kind of possibilities can be served by DE for the technology of quantum circuitry? This method is useful when QDs are to be produced, of different shape and size in various densities. This technology is used already, for boosting efficiency, in device technology, such as lasers, LEDs, and solar cells. Their accurate positioning in complex structures like nano-sized circuits is very important. The lithographic direct processing used in microelectronics cannot be applied anymore; instead, the material's self-assembling properties is to be used, which is an inherent feature of every substance used.

There are various self-assembling methods to achieve spatial arranging or positioning. The nucleation of the nano-structure, which can be induced by local stress field, is to be used for locating laterally or vertically some nano-objects. This method, called controlled self-assembling, has three different forms. The first forms self-contained objects like QDs, QD pairs, or QD clovers (four-coupled QDs) by manipulating the technological parameters. The second uses the natural features (steps of monolayers (MLs), or dislocations, etc.) to induce the required ordering by self-assembling effect. The third induces the required order by applying artificial influence on the process, like for instance focused ion beam (FIB) or creation of nanoholes (NHs). The combination of these methods can provide possibility to create complex nano-structures. This is called hierarchical self-organization, which provides potentional creation of quantum circuits.

The chapter is organized in the following way. In the first parts, following "Introduction" section, we briefly describe the technique of DE. The following part describes the recent opportunities of the self-organizing-based creation of DE nanostructures. The last part describes two possibilities of applications: as an example, the QD register and circuit of quantum cellular automata will be discussed. The purpose of this paper is to speak to people engaged in circuit research with the aim of bringing together material scientists and circuit designers onto a common platform in order to overcome the problem of the present restrictions in further miniaturization.

2. Fundamentals of droplet-epitaxial technology

For the fabrication of QDs and other zero-dimensional nano-structures, various techniques have been developed. The molecular beam epitaxy (MBE) is the most advanced technology in this area for nano-structure preparation. For a long time, the only known method for the production of epitaxially grown zero-dimensional structures was the strain-induced method, based on lattice mismatch in Stranski-Krastanov growth mode [8–12]. InAs-based QDs on GaAs surface are the archetypal system. The driving force of the self-organized QD formation is the strain energy induced by the lattice mismatch, which in approximately 7% of the case the conditions restrict the material choice. Two groups of shape formations, like pyramids and domes, can be created with defect-free QD transformations.

The DE is a viable alternative technology to the production of strain-driven QDs [13–20]. Here, the material choice is not restricted by the lattice mismatch condition, which is a further advantage to a process, based on the strain-induced growth mode. DE also makes possible the fabrication of strain-free QDs and other nano-structures. This shape diversity of the produced nano-structures makes it advantageous in applications. The technology used for the growth governs the size, shape, and the elementary distribution of the developed structures. These physical parameters are very important in applications.

In DE applications, GaAs and related substances will be used as sample materials. That case, the clustering on the surface is carried out with the help of Volmer-Weber growth mode. This is a common idea, based on the splitting of the III- and V-column material supply, during the MBE growth (**Figure 2A**). The QD preparation consists of two main parts such as the formation of metallic nano-sized droplet on the surface and its crystallization. Here, the QD preparation consists of two main parts such as the formation of metallic nano-sized droplet on the surface and its crystallization with the help of the non-metallic component of the compound semiconductor [20]. In this way, not only conventional-shaped QDs but ring-like or double-ring-like zero-dimensional nano-structures can be created. Further possible nano-structures are the filled nano-hole and QD pairs or other ensembles, depending on growth parameters (**Figure 2B**). It must be noted that this DE technique is entirely compatible with the MBE technology. This attribute makes possible to combine the DE method with the other conventional MBE processes.

A typical QD preparation is illustrated in the following [21]: at first, on GaAs (001) wafer, an $Al_{0.3}Ga_{0.7}As$ layer is grown. After the layer preparation, the sample is cooled to 200°C. Following this, Ga ($\theta = 3.75$ ML) is deposited with the flux of 0.75 ML/s without any arsenic flux. After the Ga deposition, a 60-s waiting time comes. The annealing is carried out at a temperature of 350°C and at an As pressure of 5×10^{-5} Torr. The process of GaAs crystallization starts at the edge of the droplet, initialized by the three-phase line at this point, serving as discontinuity for the crystal seeding. Although, in principle, interaction can take place at any point of the droplet, due to the thermal movement, the atoms, arriving to the edge, will start the seeding of the crystallization process.

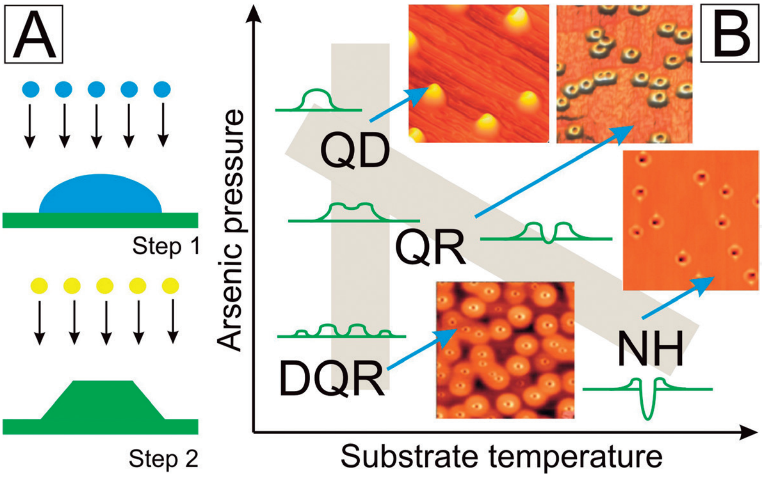

Figure 2. (A) The droplet epitaxial nano-structure production consists of two basic growth sequences; (B) versatile shaped nano-object can be created depending on the technological parameters (where QR is quantum ring, DQR is double quantum ring, and NH is nano-hole).

DE formation of ring-like QDs is similar as the previous description earlier, but the technological parameters are somewhat different; however, the AlGaAs layer preparation process is the same [22]. After that, the sample is cooled to 300°C. On the surface, Ga is deposited as described before. The same Ga is deposited with the flux of 0.19 ML/s without any arsenic flux. During the annealing, the temperature remained the same (300°C), but the arsenic pressure changed to 4×10^{-6} Torr. During the nano-structure formation, diffusion of the constituents has an important role.

A further recent method for the fabrication of strain-free QDs is the filling of nano-holes [23]. The nano-hole is created by localized thermal etching by liquid metallic droplet, and the created nano-hole is filled subsequently. A localized thermal etching takes place at conventional MBE growth temperatures, and we expect only very low level of crystal defects. The nano-holes are created in a self-organized fashion by local material removal. For inverted QD fabrication, nano-holes are generated by using Al droplets on AlAs surface. Following that, the holes are filled with GaAs to form QDs of controllable height. The nano-hole filling is carried out with GaAs in pulsed mode. The creation of QDs occurs with an inverted technology (**Figure 3**).

QD pairs can be prepared on AlGaAs surface by using the anisotropy of the (001)-oriented surface [25]. There are two known preparational processes. One of them is carried out under lower temperature, with a fewer amounts of deposited MLs. The other one is prepared under higher temperature at a higher amount of deposited Ga. In the first case, AlGaAs with an Al content of 0.27% is grown on GaAs (001) surface. Following that, Ga droplets are created at 330°C temperature on the substrate. The crystallization occurs at 200°C, under strict control of the arsenic flux. The resulting structure basically consists of two QDs aligned in the $[0\bar{1}1]$ crystallographic direction. In the other technology also, AlGaAs surface is being used. At 550°C substrate temperature, a large amount of Ga is deposited, to create droplets on the surface.

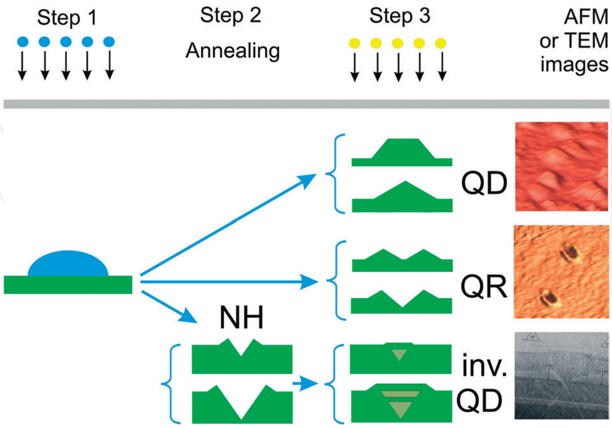

Figure 3. According to the technological parameters, the initial metallic droplet can lead to various zero-dimensional semiconductor nano-structures (where QR is quantum ring, NH is nano-hole, inv.QD is QD produced by nano-hole filling: inverted QD technology) (the AFM and TEM pictures originate from Refs. [22, 24], respectively).

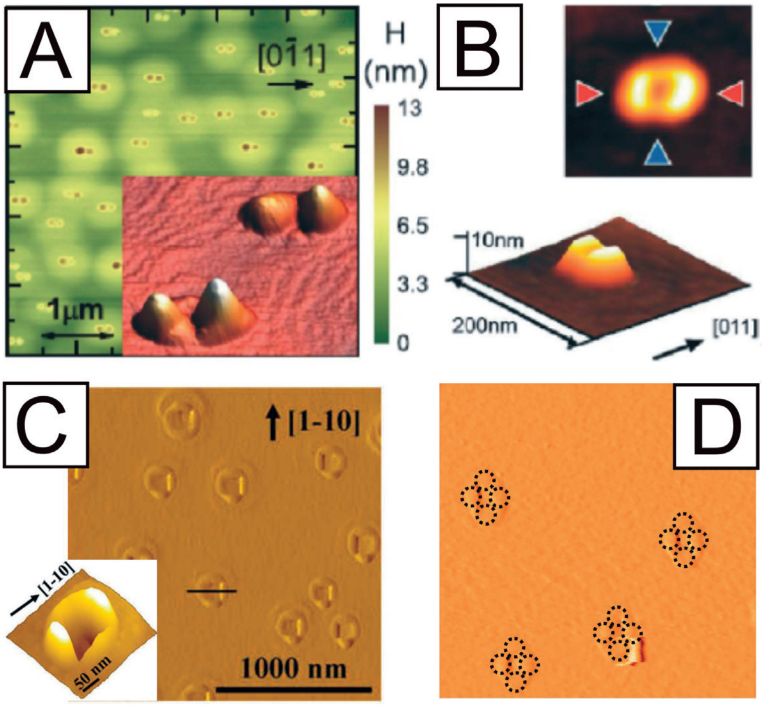

Figure 4. (A) and (B) AFM picture of QD pairs; (C) and (D) QD clovers (the AFM pictures originate from Refs. [24–26], respectively).

The structure is crystallized by an accurate control of the flux. The resulting dots are rather large. The individual pairs have an interdot distance of about 130 nm and are aligned along the $[0\bar{1}1]$ direction. QD pairs are shown in **Figure 4A** and **B**.

Nano-objects consisting of four parts can also be grown by DE. The structure is a split-ring formation. A typical technological process, when the samples are grown on GaAs (001) substrates, is as follows. First, $In_{0.15}Ga_{0.85}$ of 20 ML is deposited with a rate of 1 ML/s at 360°C. Then, the formed droplets are exposed to arsenic beam for 5 min at a temperature of 200°C to crystallize the nano-droplets. Following this, the substrate temperature is raised to 450°C for the regrowth process with a growth rate of 0.05 ML/s. The structures are shown in **Figure 4C** and **D** [26, 27].

3. Ordered nano-structures

The self-assembling ordered QDs can be linearly, circularly, and also vertically alignmented. The most promising method for achieving long-range laterally ordered self-assembled QDs is the combination of substrate pre-patterning and self-assembled growth. The pre-patterning can be carried out by using the naturally occurring anomalies on the crystalline surface or can be made artificially by external influences. There are three kinds of linear alignmentation methods (**Figure 5**). One of them is the surface cross-hatch-induced mode, and the other kind is the alignmentation created by ML step. These are utilization of naturally formed surface effects. The third one is a fully artificial method, where the alignmentation is induced by ion beam-created surface damage.

The dislocations, generated at the substrate/layer junction, show themselves on the surface as ridges and troughs. At a sufficiently high density of dislocations, the development of misfit dislocation network shows up at this junction. Such a network, consisting of two arrays of single dislocations with alternating glide planes, will result in a quadratic surface structure. The dislocation network shows itself at the surface, which is called as a cross-hatch pattern. This pattern coexists with the crystallites, giving the possibility of using the interplay between the two strain-relief mechanisms for self-ordering of QDs.

The cross-hatch pattern creation has been already demonstrated in different material systems such as $\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{As/GaAs}$ [28], $\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{As/InP}$ [29], and $\mathrm{Si}_{1-x}\mathrm{Ge}_x\mathrm{/Si}$ [30], attributed to misfit dislocations and glides. Its production is as follows. Self-assembled InAs QDs are grown on cross-hatched surface, consisting of 50 nm $\mathrm{In}_{0.15}\mathrm{Ga}_{0.85}\mathrm{As}$ layers on GaAs (0 0 1) substrate. The lattice-mismatched $\mathrm{In}_{0.15}\mathrm{Ga}_{0.85}\mathrm{As}$ layer is left growing well beyond the critical layer thickness for the formation of misfit dislocation in order to form long orthogonal cross-hatch pattern oriented along the [1 1 0] and $[0\bar{1}1]$ crystalline directions (**Figure 5A**). On top of the cross-hatched surface, InAs layer growth at a low growth rate of 0.01 ML/s and at a thickness of 0.8 ML originates spontaneous QD formation. It was found that the

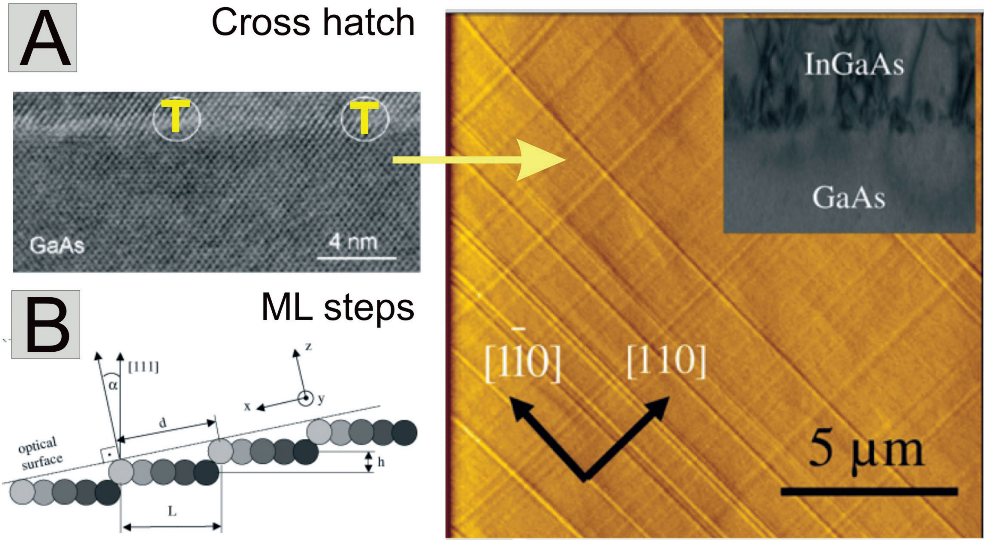

Figure 5. Linearly aligned QDs; (A) the QD alignment is induced by cross-hatch (B) and by monolayer (ML) steps, (C) and by ion-induced surface damage (the AFM and SEM pictures originate from Refs. [31, 39], respectively).

substrate temperature reduction immediately after the QD formation will result in a majority of QDs alignment on the cross-hatch pattern. With a short growth interruption, duration of 30 s, before reducing the substrate temperature, the QDs will form, almost exclusively on the cross-hatches and the surface formation is named QD hatches. Exceeding the optimum interruption time will result in inhomogeneous, sparsely connected QD hatches, possibly due to desorption of In atoms [31].

The second possibility to self-aligned QD ordering in a crystalline layer uses step bunching of preexisting ML steps on the miscut (0 0 1) substrate (**Figure 5B**). Crystallite ordering on vicinal-oriented (0 0 1) surface is guided by spontaneously formed step-bunched ripple patterns. The ripple distance and orientation can be engineered by varying the polar and azimuthal miscut directions of the substrate [32].

The focused ion beam bombardment is a widely used technique for surface preparation and nano-patterning for the fabrication of self-assembling nanostructures such as nano-ripples, nano-needles, nano-holes, and also QDs. FIB-induced self-assembly of ordered nano-structures has been reported on metals, semiconductors, and insulators as well [32–38].

Ordered Ga nano-droplets can be self-assembled under ion beam bombardment at off-normal incidence [39]. The homogeneity, size, and density of Ga nano-droplets can be controlled by the incident ion beam angle. The beam current also plays a crucial role in the self-ordering of Ga nano-droplets. It has been found that the droplets exhibit a similar droplet size but higher density and better homogeneity with an increased current of ion beam. Compared to the destructive formation of nano-droplets by direct ion beam bombardment, the controllable assembly of nano-droplets on intact surfaces can be used as templates for DE fabrication of arranged semiconductor nano-structures (**Figure 5C**).

The start of circularly aligned QD molecule can be initialized by a droplet edge (**Figure 6A**) or by a rim of nano-holes (**Figure 6B**). It is a simple method of preparing ring-shape InP nano-structures on $In_{0.49}Ga_{0.51}P$ by using DE. The surface morphology of the structure depends strongly on the ML of In. For instance, the ring-shaped nano-structure is formed at 1.6-ML In thickness. The ring-shaped QD molecule is formed when the deposited ML of In is less than 3.2 ML. It has been found that the density, height, and average number of QD per molecule are dependent on the In MLs and on its deposition rate [40].

A relatively simple way to fabricate vertical QD molecules is to grow stacks of QDs (**Figure 7A**). It is known that the surface strain field modulation from a buried island layer influences the island nucleation in the next layer and this leads to a spontaneous vertical alignment [41, 42]. The electronic coupling between vertically aligned QDs has been demonstrated earlier [40, 43–48]. Further vertically stacked QD ensembles can be created by sequentially filled nano-hole. First, Al or Ga droplets are created on the AlGaAs surface. After them an annealing appears, where the substrate temperature is ranged between 550 and 650°C, the arsenic pressure is under 10^{-7} Torr (**Figure 7B**). During this annealing, the initial droplet transforms into a nano-hole surrounded by a protrusion. The nano-hole is filled by pulsed mode. The filling consists of 0.5-s GaAs deposition followed by a 30-s pause. The stacked QDs are separated by an AlGaAs barrier layer deposition [49].

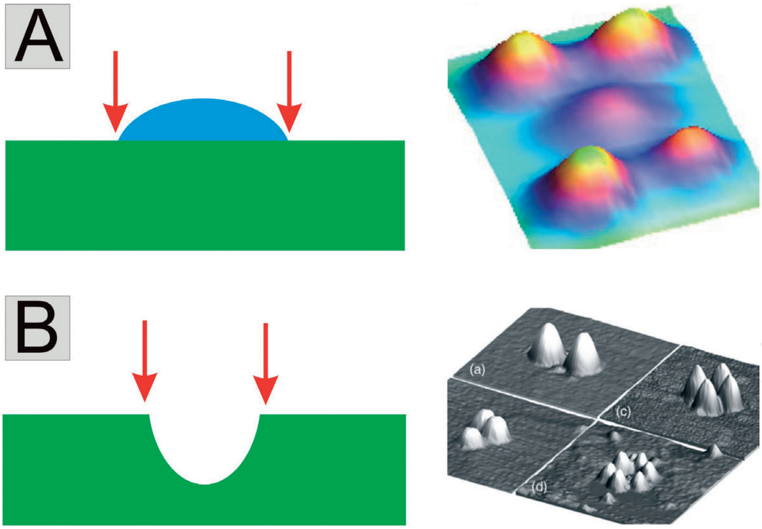

Figure 6. Circularly aligned QDs (QD molecule); (A) the QD nucleation is at droplet edge (B) and at the rim of hole opening. The arrows indicate the seeding places (the AFM pictures originate from Refs. [46, 47], respectively).

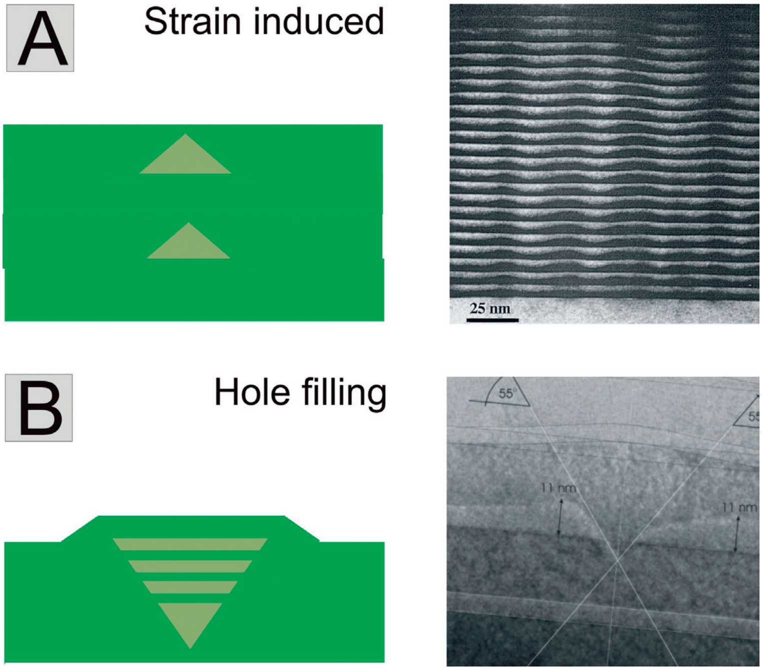

Figure 7. The vertical alignment; (A) the vertical stacking is induced by strain, (B) vertically coupled QDs by nano-hole (NH) filling (the TEM pictures originate from Refs. [24, 48], respectively).

4. Applications in quantum circuitry

In this chapter, we discuss two types of circuits composed from aligned QDs. One of them is the linearly aligned register. A QD register for quantum computing can be realized by uniformly aligned QDs or by QD pairs with the help of directed DE assembly [50]. A possible realization can be the following. The linearly aligned GaAs QDs is created on an AlGaAs surface. This structure is embedded by a barrier material of AlGaAs. When the cover layer few MLs only then the subsequently deposited metallic droplets are positioned most likely by the QD sites below (**Figure 8**).

The second discussed structure is the QD cellular automata, which was firstly proposed in the beginning of 1990s [51]. The QD-based cellular automaton is one of the most promising device structures in the future [52–55]. The circuit consists of coupled QD array to realize Boolean logic functions [9] and to perform useful computations. Two main advantages of QD cellular automata are the exceptionally high logic integration derived from the small QD size, and the low power consumption. QD cellular automata can be used to implement complex digital circuits by properly arranged QD clovers. Such circuits are, for example, full adder, multiplexer, programmable logic array, multivibrator or can be also designed memory circuits, such as quantum dot cellular automatic random access memory and serial memory. The basic building block of QD cellular automata device named cell is presented in **Figure 9A**. QD cellular automata unit cell consists of four QDs in a square array coupled by tunnel barriers, and two

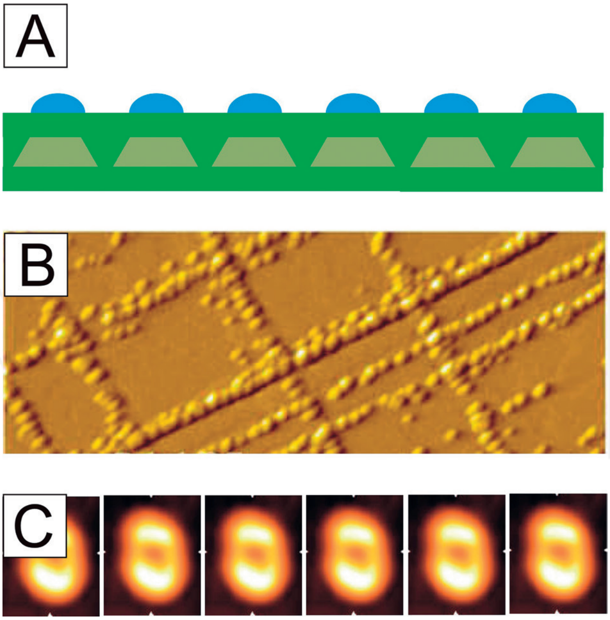

Figure 8. Realization of QD register: (A) cross section of literally aligned QD series with vertically positioned self-assembling metallic clusters; (B) along cross-hatch-aligned QDs; (C) the alignmentation of QD pairs is also possible.

electrons are injected into the cell (**Figure 9B**). Due to Coulombic repulsion, the two electrons reside in opposite corners representing two polarizations. Some basic elements for QD cellular automata logic implementation are wire, inverter, and majority voter [52–60].

The DE-grown QDs as building elements for quantum computing were first proposed in 2009 [61, 62]. The alignmentation of the QD clovers can be realized like a single dot, which can lead to the wire implementation. Here, the linear inhomogeneity of the surface can be utilized. For the gate realization, these inhomogeneities for directed assembly must be generated artificially. The QD cellular automata can be realized in more levels (**Figure 9C** and **D**). The couplings between the circuits on the adjacent levels can be carried out with vertical alignment.

For any required operation, it is very important to determine the optimal size of the QDs and their distances from each other. Not only the size but the working temperature is also important. At the realization, it is important to take into consideration that the switching fidelity increases with decreasing temperature [63]. It is predicted that the density of the QD-based circuits could exceed the device density of 10^{12} cm^{-2} and the operating speed could reach the frequency of THz region [64]. The clocking in THz region can also be realized with the help of DE. One of the effective ways to generate THz pulses is realized by near-infrared femtosecond laser irradiation on semiconductor/metal surfaces with the help of plasmon enhancement [65–67]. The DE is an appropriate technology to create such positioned semiconductor and metallic nano-particles. The structure can be realized by DE-grown self-alignment of QD molecules and metallic nano-particle [25, 68]. Promising perspective is provided with a recent result to the realization of the nano-positioned metallic nano-particle on QD molecule, which can be useful not only at the THz clocking but also at QD register, too [69].

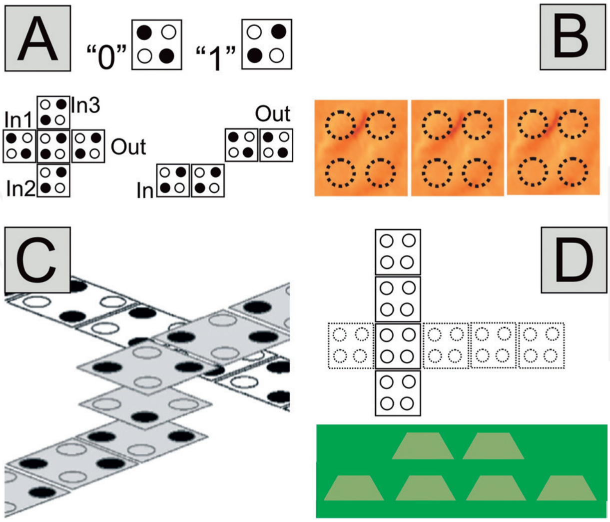

Figure 9. Realization of QD clover-based QD cellular automata; (A) the basic unit and the majority gate and inverter gate of the QD cellular automata; (B) the realization of QD cellular automata by QD.

5. Conclusion

There are still a number of open scientific problems awaiting a solution. For the perfect operation of the circuits, the optimal QDs and their distances from each other must be determined. It is a fact that the sizes and the shape of the QD are not independent from their elementary density, which finally determines the distances among the QDs. The task is rather complex. If we can understand the details of the evolution mechanism of the DE-grown nano-structures, we can approach the technological solution of the circuit formation. It is another possibility to take into account the technological capability at the circuit design, which increases the importance of the mutually common thinking among different professionals. Lately, the number of published papers in this area has increased drastically, which is an encouraging sign for the possible technological solution.

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