Growth of GaAs and InAs Quantum Dots by Metalorganic Sources of Hyperthermal Energies

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1. Introduction

Quantum dot (QD) is an attractive material system in the development of new electronic devices like single electron transistors, quantum dot lasers, and high density memories.[1,2] Strained layer epitaxy is one possible method to fabricate such quantum dots. For example, InAs/GaAs system has 7% lattice mismatch between the epitaxial layer and the substrate. This induces the Stranski−Krastanow (S−K) growth mode where a two−dimensional growth changes into a three dimensional one after few monolayers growth. The density of InAs QDs on GaAs drastically increases at a critical InAs coverage (about 1.5 monolayer) on top of the thin InAs wetting layer. The attained highest density, which is determined by the lattice strain between the epitaxial layer and substrate material, is about 10^{11} cm^{-2} in InAs/GaAs system.[3]

If initial nuclei is controlled by the another method which does not use the lattice strain of epitaxial layer and followed by the formation of a quasi stable island, a unique growth method of QDs is expected, where the dot density is controlled independently of lattice strain and therefore the substrate material is freely selected without any consideration of the lattice constant.

The initial nucleation on the substrate surface can be made by the energetic ion with a translational energy above 100 eV which breaks the surface atomic bonds but a lot of lattice defects are produced on the surface.6 A coulomb repulsion makes it difficult to decrease the translational energy below 10 eV at which less damages are formed on the surface. The supersonic molecular beam is another method to produce a source molecule with a hyperthermal energy. Since this beam is consisted of neutral molecule without coulomb repulsion, a high flux beam can be attained below 10 eV. In the energy range from several eV to 10 eV direct reactions between incident molecule and substrate atom is still enhanced and may produce the initial nucleation of QD growth.[4]

Another effect of supersonic molecular beam on the nucleation of QDs comes from a high density of source flux beam. The beam equivalent pressure (BEP) of the supersonic beam can be increased to two or three order higher than that of the conventional MBE cell. It may give a possibility of qualitative change in the nucleation of QDs through multiple coalescences between precursors.

In this paper, we present a novel growth method using supersonic molecular beams for the fabrication of InAs and GaAs QDs on GaAs substrate. The method presents a quite different nucleation mechanism from the S−K mode growth in the initial stage of QDs growth. It grows an extremely high density and uniform QDs, which cannot be attained in the S−K growth mode.

2. Experimental

For the growth of QDs of GaAs and InAs, we have developed a new state−of art growth system, which has two double supersonic molecular beams for group III and group V sources as shown in Fig.1. The system consists of two source chambers for producing supersonic molecular beams for group III and V organometallics, two chopper chambers which produce pulsed beams, a growth chamber (a target chamber), and a rotatable quadrupole−mass−spectrometer (QMS) for angular distribution of desorved molecule from the surface. Two supersonic beams for group III and V sources were focused onto the same spot on the GaAs substrate. The spot size was about 5 mm in diameter.

The beam of group III source was formed out of a 1.1 mm hole in a source chamber evacuated by two cryo−cooled oil−diffusion pumps of 2000 ls^{-1} and that of arsenic source by a turbomolecular pump of 5000 ls^{-1}. The surface of target GaAs (001) was prepared by molecular beam epitaxy. A typical vacuum pressure in the source chamber is the order of 10^{-3} to 10^{-4} Torr, while the pressure in the growth chamber (the target chamber) is below 10^{-10} Torr. To keep the each chamber pressure, a three−step differential pumping system was used to keep the pressure of each chamber. The target chamber is linked by a transfer module with a molecular−beam epitaxial chamber which prepares a GaAs buffer layer and a GaAs cap layer for a double heterostructure.

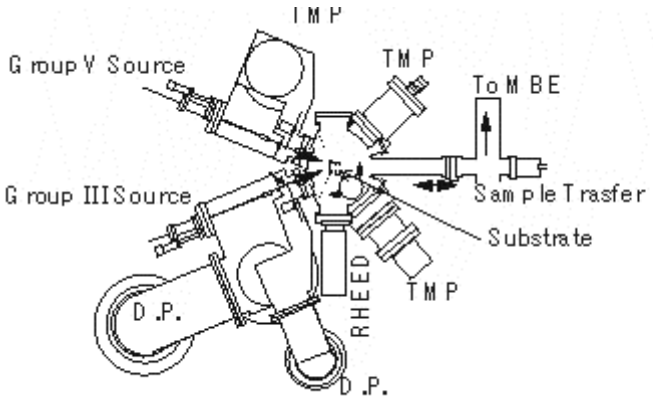

Fig.1 Growth system for InAs and GaAs QDs. The system has two supersonic beams for group III and V sources. A sample was located on the temperature controlled manipulator with 5 axes.

Triisopropylindium (TIPIn) and triisobutylgallium (TIBGa) was used as indium and gallium sources and trisdimethylaminoarsenic (TDMAAs) as an arsenic source. A group III molecular beam with a hyperthermal energy was produced by a seeding technique. The beam energy was varied by changing the species of seeding gas and the density of source gas (organic molecules). Hydrogen molecule was employed as a seeding gas. On the other hand, the arsenic source TDMAAs was not seeded (100%) in this experiment. The beam energy was determined from the measurement of the flight−time between a high−speed chopper and the mass spectrometer.

The group III and V source beams were alternatively injected onto the substrate surface as two beam pulses. The pulse duration of the group III source (TIPIn or TIBGa) was 0.5 sec and that of TDMAAs was 3 sec. The non−injecting time of about 1 msec was inserted between the both pulses. The dot density and size were measured by scannng electron microscope (SEM) and atomic force microscope (AFM).

3. Result and Discussions

The dot density drastically decreased with the incident energy of TIPIn from 2.5x 10^9 cm^{-2} at the thermal energy to nearly 0 cm^{-2} at 1.0 eV. The dot size of InAs decreased with the incident energy from 3.6x10^2 nm at 0.06 eV to zero nm at 0.95 eV.

The growth mode in this incident energy range (Fig.1 (a)) may be understood as S−K mode resulting from the difference between InAs and GaAs lattice−constants. The decrease in QDs density with the incident energy was caused by the decrease of the surface trapping probability of the incident molecule due to the increase of the inelastic direct scattering from the substrate surface. [5] At a low incident energy in so−called thermal energy, the incident molecule may be easily trapped in the potential well such as a physisorption well existing on the GaAs surface and may form an effective precursor in the growth of QDs. As the incident energy of the source molecule is increased, the incident molecule tends to be directly scattered from the substrate surface without any trapping in the potential well due to the large traslational energy. This model may be understood by the comparison with energy dependence of surface trapping probability of TIPIn molecule on GaAs(001) as shown in Fig.2(b) which was carried out by the independent measurement. [6] The figure exhibited that the surface trapping probability of TIPIn molecule changed in the similar energy dependence to that of the density of QDs. From the energy dependence of the trapping probability (Fig2(b)), we can estimate about 0.35 eV as the depth of the surface potential well.

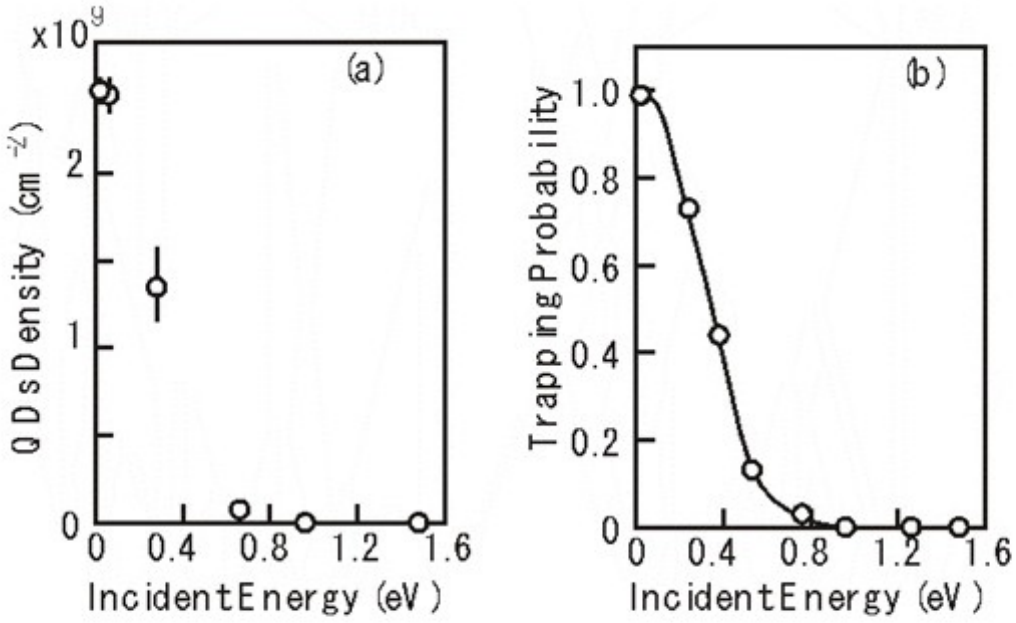

Fig.2 Variation of InAs QDs density (a) and surface trapping probability of TIPIn (b) as a function of incident energy of TIPIn source. The both energy dependences show similar energy dependence.

In the higher translational energies from 1 to 4 eV, no changes were observed on the substrate surface (the substrate temperature: 400). However, InAs dots suddenly appeared at the incident energy of 4.1 eV and the dot density drastically increased with the incident energy as shown in Fig.3. The highest density of InAs dot reached about 1.2 x 1014 cm−2 at 4.6 eV. The dot size was changed from 6.5 x 10^1 nm to 3.8 x 10^2 nm which depended on the growth condition such as the beam energy, the flux density, the growth temperature, and the TIPIn and TDMAAs pulse duration. The uniformity of the dot size was within about 5% which was the same order with the experimental error of size

measurement by SEM and AFM.

The essenntially same result was obtained in the homoepitaxial growth of GaAs QDs on GaAs substrate. When the incident energy of TIBGa beam increased above 5.6 eV, a high density of dot was appeared on the GaAs surface.

The QDs growth observed here is quite different from that of S−K growth mode. It does not depend on lattice constant difference between epitaxial layer and the substrate but largely on the incident energy of source molecules. It enables us to grow an extremely high density QDs of InAs and GaAs on the GaAs substrate compared with the QDs growth by S−K mode. The new growth method can grow a high density of QDs at a relatively high growth temperature above 400 , which is important for the growth of high quality QDs.

The appearance of InAs dot is somewhat surprising because the majority of incident molecules directly scattered from the surface at these high energies. We have no definite model on the nucleation in this case, but probably it certainly comes from the high energy incident source beam. When a high energy molecule violently collides with the surface atom, the large translational energy may change to the reaction energy, which induces a direct reaction between the incident molecule and the surface atom followed by the nucleation in InAs dot growth. Also, the high flux density of 10^{-3} Torr (BEP) in the supersonic beam may cause a high density of nucleation on the GaAs surface.

In the nucleation of quantum dot, the surface diffusion or migration of the precursor plays an important role since the coalescence between precursors largely affects the nucleation in the crystal growth. The trajectory simulation of the supersonic molecule on GaAs(001) showed that the surface molecule has a very high migration speed and may experience multiple collisions with the other molecule. This also affects the formation of such a high density of QDs.

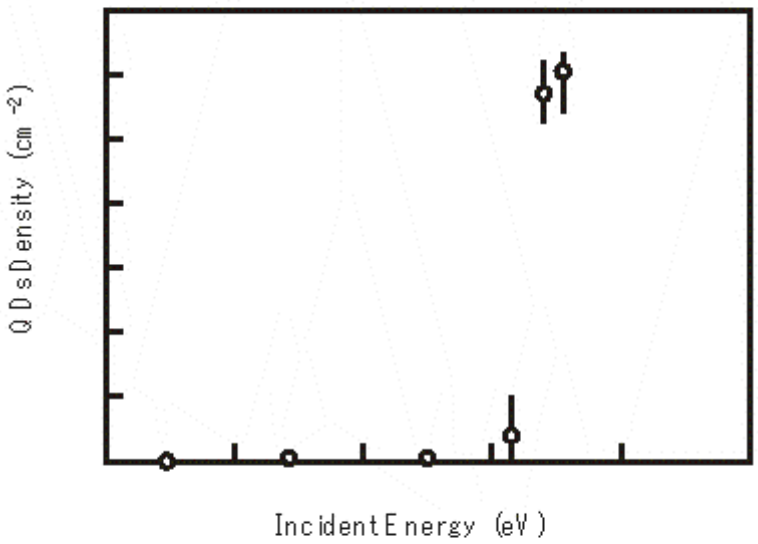

Fig.3 Energy dependence of QDs density at a higher energy region. The QDs appeares at 4.1 eV and the density abruptly increases with the incident energy.

The double−heterostructure, GaAs cap layer/InAs QDs/ GaAs buffer layer/GaAs substrate, was fabricated by this epitaxy and MBE. Fig.4 shows the photoluminescence spectrum from the InAs QDs. A strong PL emission appeared at the wavelength of 1070 nm with a narrow halfwidth of 43 meV, which suggested that the size of InAs dots had a very uniform distribution. The strong and sharp emission observed here means that the epitaxy is promising to the application to the nanostructure devices such as a quantum−dot laser.

In summary, a newly developed epitaxy for QDs grows uniform quantum dots from the density of 3.2×10^{10} cm^{-2} to an extremely high density of 1.2×10^{14} cm^{-2} , which cannot be attained in the growth of Stranski−Krastanow (S−K) mode. InAs quantum dots grown by this technique showed a very sharp and strong peak in the photoluminescence spectra, indicating that the dot is very uniform in size and has good quality. The detailed growth mechanism is now investigating which suggests that time lag of arsenic pulse after the indium pulse is an important factor in the QDs nucleation.

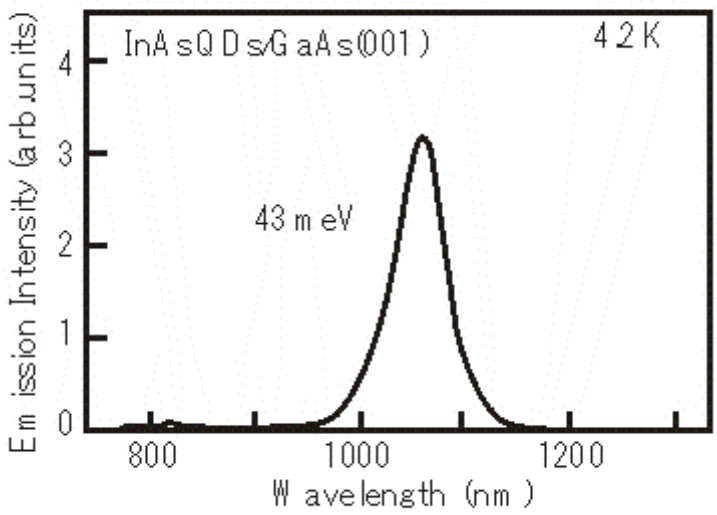

Fig.4 Photoluminescence spectrum of InAs QDs at 4.2K. A strong emission is observed at 1070 nm with a halfwidth of 43 meV.

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