InAs and GaAs quantum dots grown by hyperthermal source beams

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Abstract

A growth method which uses group III (triisopropylindium and triisobutylgallium) and V (trisdimethylarsenic) metalorganic molecules with hyperthermal energies has been developed for the growth of GaAs and InAs quantum dots. It grows uniform quantum dots with an extremely high density of 1 x10^{14} cm^{-2}, which cannot be attained using Stranski-Krastanov (S-K) growth mode. GaAs and InAs quantum dots grown by this technique show a sharp and strong emission peak in the photoluminescence spectra, suggesting that the dots are very uniform in size and have good quality. It is demonstrated that the growth mechanism is quite different from that of the S-K growth mode.

Key Words:
Growth method, Chemical beam epitaxy, Semiconducting III-V materials

1. Introduction

Quantum dots (QDs) are of interest for development of new electronic devices like single electron transistors, quantum dot lasers, and high density memories [1-3]. Strained layer epitaxy is one possible method to fabricate such quantum dots. For example, the InAs/GaAs system has 7% lattice mismatch between the epitaxial layer and the substrate. This induces the Stranski-Krastanow (S-K) growth mode where two-dimensional growth changes into three-dimensional growth after a few monolayers are deposited. 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 the InAs/GaAs system [4, 5].

If the initial nuclei are controlled by 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. This would allow the substrate material to be freely selected without any consideration of the lattice constant.

The initial nucleation on the substrate surface can be made by energetic ions with a translational energy above 100 eV which break the surface atomic bonds. However this also produces a lot of lattice defects on the surface [6]. Coulomb repulsion makes it difficult to decrease the translational energy below 10 eV where less damage is created on the surface. The supersonic molecular beam is another method capable of producing source molecules with hyperthermal energy [7]. Since this beam consists of neutral molecules without coulomb repulsion, a high flux beam can be attained with energies below 10 eV. In the energy range from several eV to 10 eV, direct reactions between incident molecule and substrate atom are still

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enhanced and may produce the initial nucleation of QD growth [8-10].

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 quite a different nucleation mechanism from the S-K growth mode typically used in the initial stage of QD growth, resulting in an extremely high density and highly 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 the 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 detection of desorbed molecules 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 group III source beam was formed out of a 1.1 mm hole in a source chamber evacuated by two cryo-cooled oil-diffusion pumps (2000 ls^{-1}). The chamber used to generate the arsenic source was pumped by a turbomolecular pump (5000 ls^{-1}). 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 maintain the low pressure in each chamber, a three-step differential pumping system was used. The target chamber is linked by a transfer module to a molecular-beam epitaxial chamber which prepares a GaAs buffer layer and a GaAs cap layer for the double heterostructure. Triisopropylindium (TIPIn) and triisobutylgallium (TIBGa) were used as indium and gallium sources and trisdimethylaminoarsenic (TDMAAs) as the arsenic source. A group III molecular beam with hyperthermal energy was produced by a seeding technique [7]. The beam energy was varied by changing the species of seeding gas and the density of source gas (organic molecules). Hydrogen was employed as a seeding gas. TIPIn and TIBGa source molecules could gain extremely high translational energies in this seeding technique due to their large masses. The arsenic source, TDMAAs, was not seeded 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 alternately injected onto the substrate surface as two separate beam pulses. The pulse duration of the group III source (TIPIn or TIBGa) was 0.5 sec and that of TDMAAs was 3 sec. A non-injecting time of about 1 msec was inserted between the pulses. The dot density and size were measured by scanning electron microscopy (SEM) and atomic force microscopy (AFM).

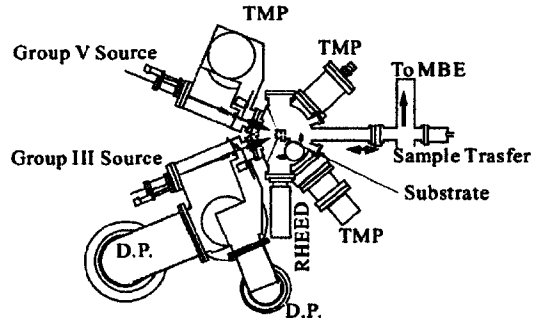

Fig.1 Growth system for InAs and GaAs QDs. The system has two supersonic beams for group III and V sources. A sample was located on the temperature controlled manipulator with 5 axes.

2. Results and discussion

It was found that the surface density of the InAs QDs largely depended on the incident translational energy of the TIPIn molecule. Fig.2(a) shows the variation of the InAs dot density as a function of the incident TIPIn in the translational energy range below 1.6 eV and at a substrate temperature of 400°C. 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

InAs dot size decreased with the incident energy from $3.6x10^2$ nm at 0.06 eV to zero nm at 0.95 eV.

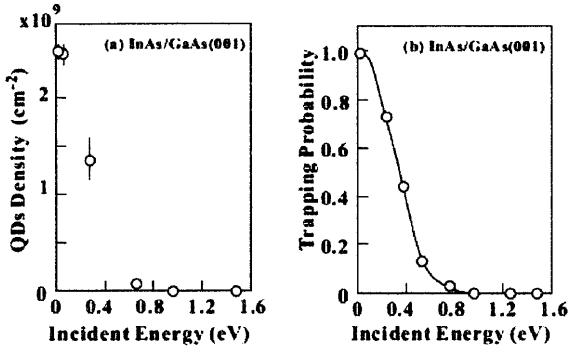

Fig.2 Variations of InAs QDs density (a) and surface trapping probability of TIPIn (b) as a function of incident energy of TIPIn source at low incident energies

The growth in this incident energy range (Fig.2 (a)) may be understood as an S-K mode resulting from the difference between the InAs and GaAs lattice-constants. The decrease in QD density with increasing 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 [11]. At low incident energy, the incident molecule may be easily trapped in the potential well, such as a physisorption well, existing on the GaAs surface and may behave as an effective precursor for 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 translational energy. This model may be understood by comparison with the energy dependence of the surface trapping probability of the TIPIn molecule on GaAs(001).

The incident TIPIn molecule was assumed to have two reaction channels; one is direct scattering from the GaAs surface and the other the surface trapping which may contribute to the GaAs growth. The surface trapping probability was estimated from the measurement of the direct-scattering intensity of TIPIn from the surface[12]. At the low incident energy of 0.04 eV, no TIPIn molecules scattered from the GaAs surface were detected by QMS. When the incident energy of TIPIn was increased beyond 0.1 eV, the TIPIn molecules began to be observed and the scattering intensity increased with the incident energy and leveled off at the incident energy above ~1.0 eV. From these measurements, the surface trapping probability of TIPIn was estimated as shown in Fig.2(b). Here the surface trapping probability was assumed to be zero at the incident energy of 3.0 eV. The surface trapping probability of the TIPIn molecule exhibited similar energy dependence to that of the density of the QDs. From the energy dependence of the trapping probability (Fig.2(b)), we can estimate the depth of the surface potential well to be ~ 0.35 eV.

In the higher translational energies from 1 to 4 eV, no changes were observed on the substrate surface at substrate temperature of 400℃. However, InAs dots suddenly appeared at the incident energy of 4.1 eV and the dot density drastically increased with increasing incident energy as shown in Fig.3. The highest dot density achieved was about 1.2×10^{14} cm^{-2} at 4.6 eV. The dot size varied from 6.5×10^1 nm to 3.8×10^2 nm depending on the flux density, the growth temperature, and the TIPIn and TDMAAs pulse durations. 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 substrate temperature was changed from 300℃ to 500℃ for the incident energy of 4.6 eV. Above 350℃, the QD density was about 1.2×10^{14} cm^{-2} and nearly independent on the substrate temperature. But the QDs density was drastically decreased with decreasing the substrate temperature below 350℃ because of the decrease of the decomposition rate of TDMAAs molecule.

The growth of GaAs QDs was studied on GaAs(001) substrate using TIBGa and TDMAAs sources at 500℃. When the incident energy of TIBGa was lower than 5.6 eV, the 2-dimensional GaAs layer grew on GaAs(001) instead of the QDs. Fig.5(a) shows the GaAs layer thickness as a function of the TIBGa incident energy. The layer thickness decreased with the incident energy and became to be nearly zero at the incident energy ~0.6 eV. This variation can be

explained by the energy dependence of TIBGa trapping probability on the GaAs(001). Fig.5 shows the incident-energy dependence of the TIBGa trapping probability on GaAs(001) at 500°C. The similar energy dependence to the growth rate of GaAs layer can be seen in the trapping probability of TIBGa molecule as in the case of TIBIn on GaAs (Fig.2(b)).

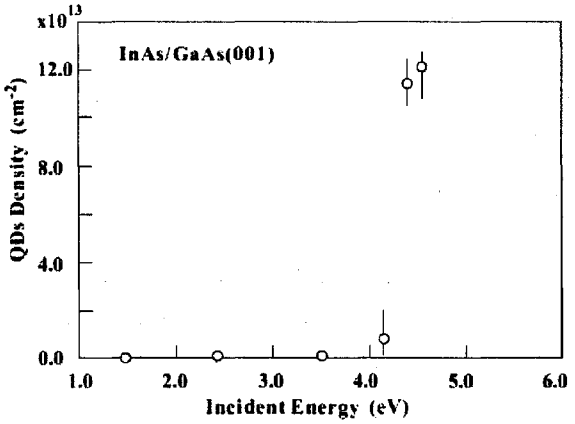

Fig.3 Energy dependence of InAs QDs density in a higher energy region. InAs QDs appear at 4.1 eV and the density increases with further increases in the incident energy.

When the incident energy of TIBGa beam was increased above 5.6 eV, a high density of GaAs dots appeared on the GaAs surface. The dot density increased with increasing incident energy as shown in Fig.3. The highest dot density achieved was about 1.0 x 10^{14} cm^{-2} at 6.2 eV. The dot size varied from 8.5 x 10^{1} nm to 2.1 x 10^{2} nm depending on the growth condition. These values were the same order with the InAs QDs on GaAs(001).

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

The appearance of InAs and GaAs dots is somewhat surprising because the majority of incident molecules is 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 of the InAs and GaAs dots. Also, the high flux density of 10^{-3} Torr (BEP) in the supersonic beam may cause a high density of nucleation clusters on the GaAs surface.

The other hyperthermal molecular beams of TMGa, TEGa, and TMIn were investigated for the precursors, but no QDs could be observed on the GaAs surface in any growth conditions. This result may be explained by the fact that these molecules have relatively small masses so that they cannot gain sufficient translational energies to react with the surface atom (for example, the maximum translational energy of TMGa was about 1.9 eV in the seeding by hydrogen gas).

There is a possibility that gallium clusters are being formed in the gas phase prior to arrival at the substrate surface. But this may be removed for the following reasons:

(1) The interaction between source molecules may be neglected in the supersonic molecular beam. Therefore the gas reactions in the supersonic nozzle must be considered.

(2) The QDs appeared on the surface, when the incident beam energy increased at the hyperthermal energy (> 4.1 eV for TIPIn and > 5.6 eV for TIBGa). The incident energy was controlled by diluting TIPIn or TIBGa with hydrogen gas; the higher energy source beam was produced by the more diluted gas, resulting in the weaker reactions between source molecules.

(3) The incident source beam was directly analyzed by QMS up to the mass-number of 2000. The fragmentation pattern from the TIBIn or TIBGa molecule was observed in the QMS spectra, but no fragments coming from gallium atom, arsenic atom, and their clusters could be detected in the source beam.

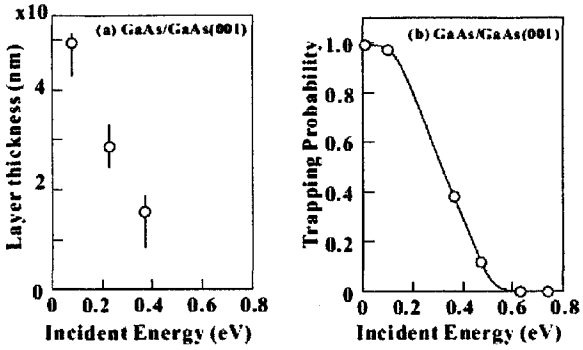

Fig.4 Variations of GaAs layer thickness (a) and surface trapping probability of TIBGa (b) as a function of incident energy of TIBGa molecule at low incident energies

In the nucleation of quantum dots, 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[13]. This also affects the formation of such a high density of QDs.

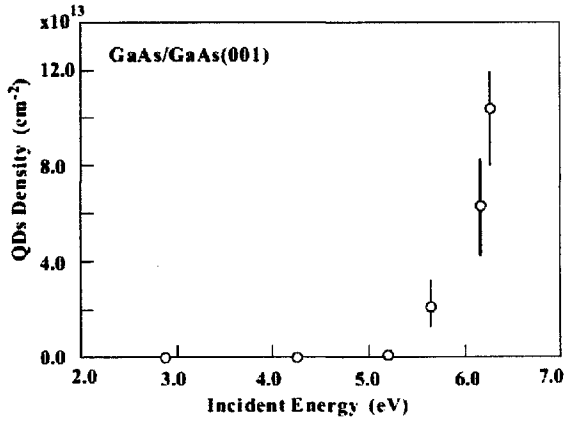

Fig.5 Energy dependence of GaAs QDs density in a higher energy region. GaAs QDs appear at 5.6 eV and the density increases with further increases in the incident energy.

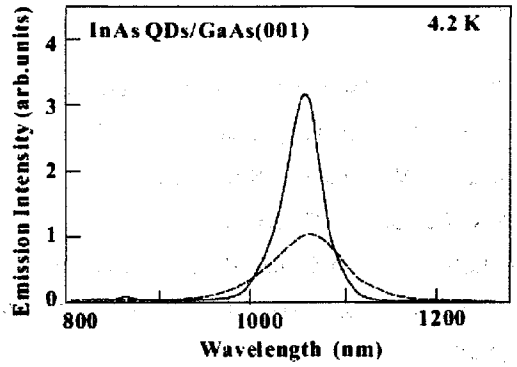

Fig.6 Photoluminescence spectra of InAs QDs at 4.2 K. A strong emission is observed at 1070 nm with a halfwidth of 43 meV (solid line) for the incident beam energy of 4.6 eV. A weaker spectrum with a halfwidth of 91 meV (dotted line) is from the QDs grown by S-K mode (the incident beam energy: 0.04 eV).

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 spectra taken for the InAs QDs grown by this new technique (hyperthermal molecular beam: HTMB) and the conventional S-K mode. A strong PL emission appeared at the wavelength of 1070 nm with a narrow halfwidth of 43 meV for the HTMB sample, which suggested that the size of the InAs dots had a very uniform distribution. On the other hand, a PL emission with a broader halfwidth of 84 meV was observed for the S-K mode sample. The strong and sharp emission observed in the HTMB sample 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 a density of 10^{10} cm^{-2} to an extremely high density of 10^{14} cm^{-2} for InAs and GaAs. Such high levels cannot be attained using traditional Stranski-Krastanow (S-K) growth mode. InAs quantum dots grown by this technique showed a very sharp and strong peak in the photoluminescence spectra, indicating that the dots are very uniform in size and have good quality.

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