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Surface Morphology of $In_{0.5}Ga_{0.5}$ Quantum Dots Grown using Stranski-Krastanov Growth Mode

(Morfologi Permukaan Bintik Kuantum $In_{0.5}Ga_{0.5}As$ yang ditumbuhkan Menggunakan Mod Pertumbuhan Stranski-Krastanov)

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ABSTRACT

In this research an atomic force microscopy (AFM) study on self-assembled $In_{0.5}Ga_{0.5}As/GaAs$ quantum dots (QDs) was performed. Surface morphology of self-assembled $In_{0.5}Ga_{0.5}As$ QDs changes with different growth time. Increasing growth time increased the dots size and decreased the dots density. In addiditon, self-assembled $In_{0.5}Ga_{0.5}As$ QDs was grown on $In_{0.1}Ga_{0.9}As$ underlying layer with different after-growth AsH_3 flow time during cooling-down. The underlying layer caused lattice strain relaxation in the QDs on the surface. Increasing the period of AsH_3 flow during cooling-down reduced the diameter of the dots and increased the density. The migration of groups III species in the growth of $In_{0.5}Ga_{0.5}As/GaAs$ system was influenced by AsH_3 flow during cooling-down period. This was due to the increase in surface population of active arsenic species. Underlying layer and the period of AsH_3 flow during cooling-down are the two key factors in the fabrication of small and dense $In_{0.5}Ga_{0.5}As$ QDs.

Keywords: Quantum dots; Stranski-Krastanov

ABSTRAK

Dalam makalah ini, penyelidikan mikroskop daya atom kepada bintik kuantum $In_{0.5}Ga_{0.5}As/GaAs$ yang terhimpun sendiri telah dilaksanakan. Morfologi permukaan bintik kuantum $In_{0.5}Ga_{0.5}As/GaAs$ yang terhimpun sendiri berubah dengan masa penumbuhan yang berbeza. Peningkatan masa penumbuhan meningkatkan saiz tetapi merendahkan ketumpatan bintik. Disamping itu, bintik kuantum $In_{0.5}Ga_{0.5}As/GaAs$ yang terkumpul sendiri telah ditumbuhkan di atas lapisan bawahan $In_{0.1}Ga_{0.9}As$ dengan pengaliran AsH_3 selepas penumbuhan yang berbeza semasa proses penyejukan. Lapisan bawahan telah merehatkan terikan kekisi di dalam bintik kuantum di atas permukaan. Penambahan tempoh pengaliran AsH_3 semasa proses penyejukkan mengurangkan diameter bintik dan menambahkan ketumpatannya. Migrasi spesis kumpulan III dalam penumbuhan sistem $In_{0.5}Ga_{0.5}As/GaAs$ adalah dipengaruhi oleh pengaliran AsH_3 semasa dalam tempoh proses penyejukkan. Ini adalah disebabkan peningkatan populasi permukaan spesies arsenik aktif. Lapisan bawahan dan tempoh pengaliran AsH_3 semasa proses penyejukkan merupakan dua faktor penting dalam fabrikasi bintik kuantum yang kecil dan padat.

Kata kunci: Bintik kuantum; Stranski-Krastanov

INTRODUCTION

Over the past decade there have been many experimental and theoretical studies of the development of size, uniformity, and optical properties of nanostructure such as quantum wires and quantum dots (QDs). The unique physical properties of QDs are expected improved the performance for a multitude of optoelectronic devices (Wang et al. 2006). Several methods have been reported in the fabrication of QDs structures. One of the promising options is the use of Stranski-Krastanov growth mode, of which QDs structures can easily be self-assembled. In this method, the growth initially starts in two-dimension and then beyond the transition thickness, islands are formed spontaneously leaving a thin wetting layer under the 3-D islands (Srinivasan et al. 2005). The Stranski-Krastanov growth mode depends strongly on growth parameters and misorientation of substrate (Hsu et al. 2006). However, the formation process of the 3-D islands has not been clarified. It is important to study this process to obtain QDs of high quality using this method.

Indium gallium arsenide (InGaAs) and related III-V semiconductors have attracted much interest as the most prospective materials for optoelectronic devices, and high power–high temperature device applications. The self-assembled $In_xGa_{1-x}As$ QDs grown on GaAs substrate have attracted much attention due to their potential applications in future high-performance electronic and optoelectronic devices. Recently, many studies have been conducted in the development of devices based on $In_xGa_{1-x}As$ QDs using metalorganic chemical vapour deposition (MOCVD) and molecular beam epitaxy (MBE). Among them are quantum dots lasers (Bimberg 2005; Germann et al. 2007), infrared photo-detectors (Jiang et al. 2005; Xu et al. 1998), quantum dots solar cells (Dimroth et al. 2000) and single-electron transistors (Osborn et al. 2004). The performance of these QDs devices is influenced by their size, shape, homogeneity, composition and density of self-assembled QDs (Ishihara et al. 2002). Surface morphology of self-assembled QDs is dependent on the growth condition, thus the surface morphology and optical properties of self-assembled $In_xGa_{1-x}As$ QDs is influenced strongly by the temperature of the substrate, V/III ratio, indium content, misorientation of the substrate and growth rate (Kladko et al. 2007). It is very important to understand the relationship between the growth procedures to the evolution of surface morphology and optical properties of the $In_xGa_{1-x}As/GaAs$ QDs structures.

The structural properties of self-assembled QDs have been generally investigated using various methods, such as transmission electron microscopy (TEM) (Leon et al. 2000), double X-ray diffraction (Ng & Missous 1996), diffuse X-ray scattering (Hanke et al. 2004), AFM (Kim et al. 2005) and reflection of high-energy electron-diffraction (RHEED) measurement (Joyce et al. 2001). Among various kinds of structural investigation techniques, AFM is a powerful method for observation of the surface growth on atomic scale. AFMs probe the sample and make measurements in three-dimensions, *x, y,* and *z* (normal to the sample surface), thus enabling the presentation of three-dimensional images of a sample surface. In this paper, we present the surface morphology studies of self-assembled $In_{0.5}Ga_{0.5}As$ QDs grown on GaAs (100) substrate using MOCVD. The Stranski–Krastanov (SK) growth mode of three-dimensional island formation in self-assembled $In_{0.5}Ga_{0.5}As$ QDs has also been observed using AFM.

Experiment

The samples used in this study was grown on GaAs (100) substrate using Stranski-Krastanov in the vertical reactor Nanoepi Versatility 2×1 MOCVD systems. Precursor used for the growth of GaAs layer and $In_{0.5}Ga_{0.5}As$ QDs were trimethylgallium (TMGa), trimethylindium (TMIn) and arsine (AsH_3). Prior to growth, the substrate temperature was increased up to 700°C for 10 min under arsine flow to remove oxide on the substrate surface. The growth was initiated from the GaAs buffer layer with a thickness of 200 nm of 650°C. The temperature was then decreased to 550°C for the growth of self-assembled $In_{0.5}Ga_{0.5}As$ QDs with deposition time of 4.5, 5.0, and 6.0 s. Total reactor pressure of 76 torr was maintained during growth, and all samples were grown under the same growth conditions with additional 3 min after-growth AsH_3 flow during cooling-down. The growth rate was estimated to be 1.1 µm/h obtained from the field emission scanning electron microscopy (FE-SEM). For the other samples, 10 nm $In_{0.1}Ga_{0.9}As$ underlying layer were added before the growth of self-assembled $In_{0.5}Ga_{0.5}As$ QDs (with deposition time of 4.5 s and after growth AsH_3 flow time of 1 and 3 min). The structure of self-assembled $In_{0.5}Ga_{0.5}As$ QDs on GaAs (100) substrate were analysed using AFM. The AFM is ideally suited for both visualisation of nano-structured materials and for measuring the spatial dimensions of features at the surface of nano-materials. Surface morphology of self-assembled $In_{0.5}Ga_{0.5}As$ QDs was taken in contact to AFM mode with a scan rate of typically 1 Hz.

Results and Discussion

Self-assembled $In_{0.5}Ga_{0.5}As$ QDs samples were successfully grown on 200 nm GaAs buffer layer using MOCVD. The surface morphology of 200 nm GaAs buffer layer is show in Figure 1. These AFM images show that the multi-atomic steps were formed on the surface of the 200 nm GaAs buffer layer. The formation of these steps was along [110] direction as determined by Kitamura et al. (1997). GaAs multi-atomic steps were naturally formed on GaAs (100) substrate during epitaxial growth using MOCVD. The terrace were atomically flat with width between 100 and 250 nm. The larger terrace width indicates that the kinetics of the steps is widespread and the surface diffusion of adatoms along the surface is long enough due to the low deposition rate inhibiting from frequent nucleation of adatoms (Son et al. 2008). From AFM measurement, the heights of the step on the surface were between 0.3 to 0.5 nm and the average step width was 10 nm. In another study by Ishihara et al. (2002), the misoriented substrate caused the formation of the multi-atomic steps on the surface of the buffer layer. The formation of multi-atomic steps from GaAs buffer layer affects the formation of QDs on the surface.

The 2D and 3D AFM images of self-assembled $In_{0.5}Ga_{0.5}As$ QDs grown on GaAs (100) substrate with different growth time are shown in Figure 2 (a) - (c). These AFM images show the evolution of surface morphology of the QDs with increasing growth time. Figure 2(a) also shows the existence of $In_{0.5}Ga_{0.5}As$ steps on the surface of the sample, like those of GaAs, and almost all of the QDs are seen formed on these steps edges. The dots formed by the releasing the elastic strain energy which compressive stress induced of the lattice mismatch between $In_{0.5}Ga_{0.5}As$ QDs and GaAs layer (Son et al. 2008). The average height, diameter, and density from QDs were approximately 4 nm, 18 nm and 1.04×10^{10} cm^{-2}, respectively. This result is different compared to sample (b) and (c) where the existence of $In_{0.5}Ga_{0.5}As$ steps was not seen on the surface and larger dots appear on the surface. AFM measurement shows that the average height, diameter, and density for samples (b) and (c) were 9 nm, 24 nm and 1.59×10^{10} cm^{-2} and 13 nm, 35 nm and 1.14×10^{10} cm^{-2}, respectively. Increasing growth time of self-assembled $In_{0.5}Ga_{0.5}As$ QDs from 4.5 to 5 s increased the dot formation, with increasing dots size and density. Larger dots however, were formed on the surface, with average height 19 nm and average diameter 55 nm. In contrast to sample (c), with increasing

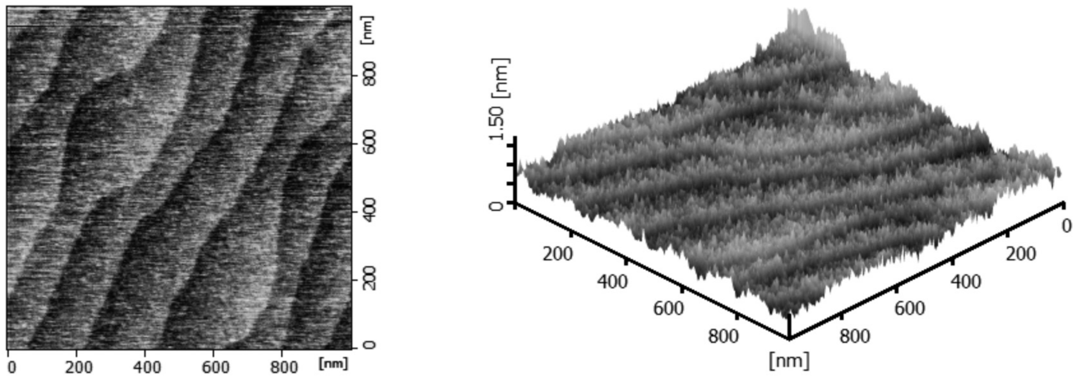


FIGURE 1. AFM images of surface morphology of the 200 nm GaAs buffer layer

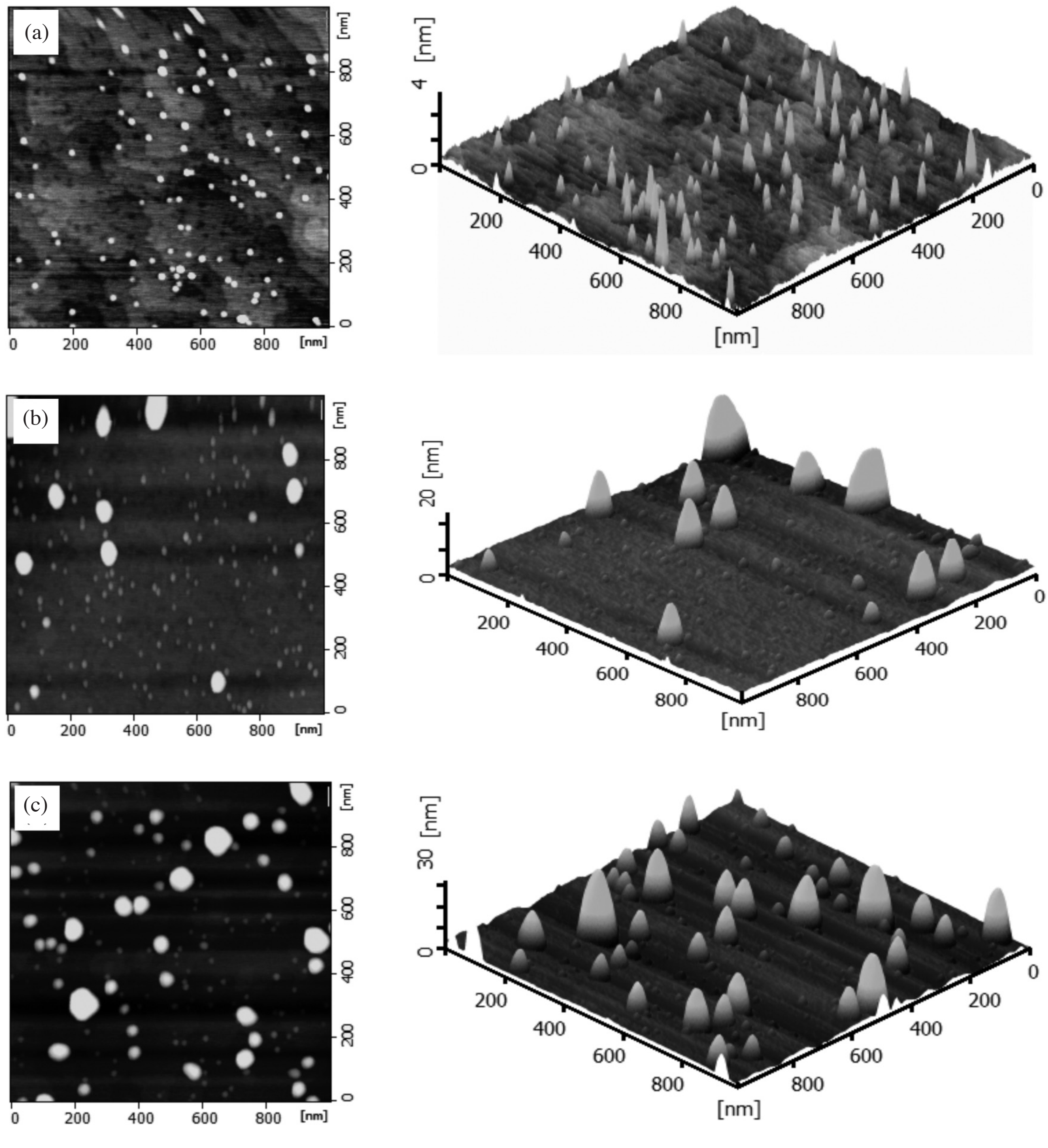


FIGURE 2. AFM images of $In_{0.5}Ga_{0.5}As$ QDs on GaAs (100) substrate with growth time
(a) 4.5 s, (b) 5 s, and (c) 6 s

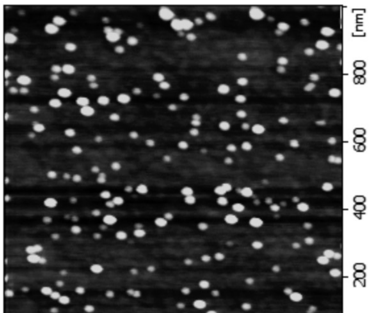
growth time caused the dots to be larger and decreased in the dot density. It was mainly due to the coalescence of $In_{0.5}Ga_{0.5}As$ QDs to much larger ones. One piece of evidence for the coalescence was the increase in the number of relatively larger QDs as shown in Figures 2 (b) and (c). This is similar to the study conducted by Kim and Kim (2006), where the size of the QDs gradually increased and the dots density decreased with increasing InAs QDs thickness.

Although the mechanism of large dot formation is not yet well understood, they probably formed due to the large migration distance of indium atom along the GaAs steps edges (Ishihara et al. 2002). The Stranski-Krastanov growth mode is damage-free formation of dot structures directly on the epilayer surface by self-assembled mechanisms. However, the QDs are not sufficiently uniform in size and distribution.

Figure 3 shows the self-assembled $In_{0.5}Ga_{0.5}As$ QDs grown on GaAs (100) substrate with a 10 nm $In_{0.1}Ga_{0.9}As$ under layer and 3 minutes AsH_3 flow during cool-down period. This sample was similar in growth conditions as sample (a) in Figure 2, but with additional 10 nm $In_{0.1}Ga_{0.9}As$ underlying layer grown after 200 nm GaAs buffer layer but before the final grown of self-assembled $In_{0.5}Ga_{0.5}As$ QDs. The AFM measurement shows that the average height, diameter and density were 5 nm, 20 nm and 1.62×10^{10} cm^{-2}, respectively. This density is higher compared to sample (a). It is well known that, due to the stress induced in the underlying layer, the strain relaxation on the surface will control the nucleation and growth of the dots above (Jiang et al. 1999). Xu et al. (2005) showed that the relief of strain in the wetting layer is believed to account for the transition from the 2D to 3D growth mode.

In this work, the $In_{0.5}Ga_{0.5}As$ QDs grown using $In_{0.1}Ga_{0.9}As$ underlying layer has increased in size compared to the one without on underlying layer. This shows that the growth of on underlying layer before self-assembled QDs reduced the critical layer thickness. It occurs due to the indium segregation above the $In_{0.1}Ga_{0.9}As$ template. A significant contribution to QDs formation is made when a floating layer of indium forms on the top of the growing $In_{0.1}Ga_{0.9}As$ layer and its thickness reaches a few monolayers. The general rule is that the thicker the $In_{0.1}Ga_{0.9}As$ template or the higher the indium composition, the smaller the critical thickness. It is also known that the indium accumulation in QDs is determined by strain minimisation during growth (Offermans et al. 2005). Chang et al. (2001) found that in the growth of $In_xGa_{1-x}As$ QDs using InAlAs underlying layer, the fraction of $In_xGa_{1-x}As$ for dot formation becomes greater, and increased the dots size and density. In another study by Jiang et al. (2005) using InGaAlAs under layer before the growth of $In_xGa_{1-x}As$ QDs, showed that the InGaAlAs underlying layer was the key factor in the fabrication of small and dense $In_xGa_{1-x}As$ QDs. The addition of aluminium to $In_xGa_{1-x}As$ resulted in the formation of smaller InGaAlAs QDs with higher density. The absence of underlying layer is evidence based on the self organized effect of the homo-species in the lattice mismatched hetero-interface. The surface morphology of the self-assembled $In_{0.5}Ga_{0.5}As$ QDs drastically changed using $In_{0.1}Ga_{0.9}As$ underlying layer.

Surface morphology of $In_{0.5}Ga_{0.5}As$ QDs grown on thin $In_{0.1}Ga_{0.9}As$ underlying layer with the 1 minute AsH_3 flow during cooling-down period is shown in Figure 4. Compared to Figure 3, the AFM images show an increase in the dots density with decreasing AsH_3 flow time during cooling-down period due to formation of smaller dots. The lateral diameter, average height and density of $In_{0.5}Ga_{0.5}As/In_{0.1}Ga_{0.9}As/GaAs$ QDs with 1 min AsH_3 flow during cooling-down are 16 nm, 6 nm, and 3.69×10^{10} cm^{-2}, respectively. AsH_3 flows during cooling-down process have important effect in the dots nucleation. Sears et al. (2008) showed that the presence of AsH_3 during cooling-down period resulted in strong island ripening. Longer period of AsH_3 flow produces a faster nucleation process of $In_{0.5}Ga_{0.5}As$ QDs, which then produces larger dots and low dots density. The different period of time for AsH_3 flow during cooling-down period affect the migration of gallium and indium. This is the effect of increasing surface population of active AsH_3 species. As presented by Riel et al. (2002), the migration of group III species is attributed to the AsH_3 pressure. The exact mechanism by



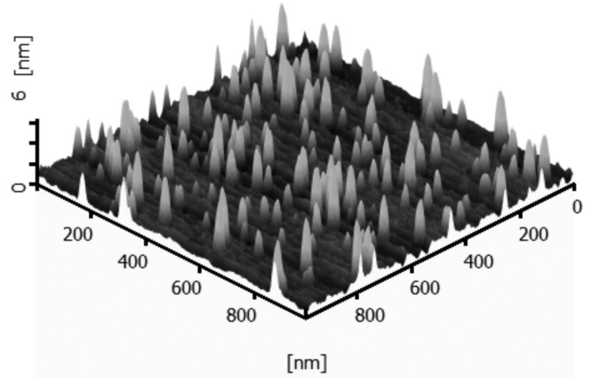


FIGURE 3. AFM images of $In_{0.5}Ga_{0.5}As/GaAs$ QDs grown using 10 nm $In_{0.1}Ga_{0.9}As$ wetting layer with 3 min AsH_3 flow during cooling-down period

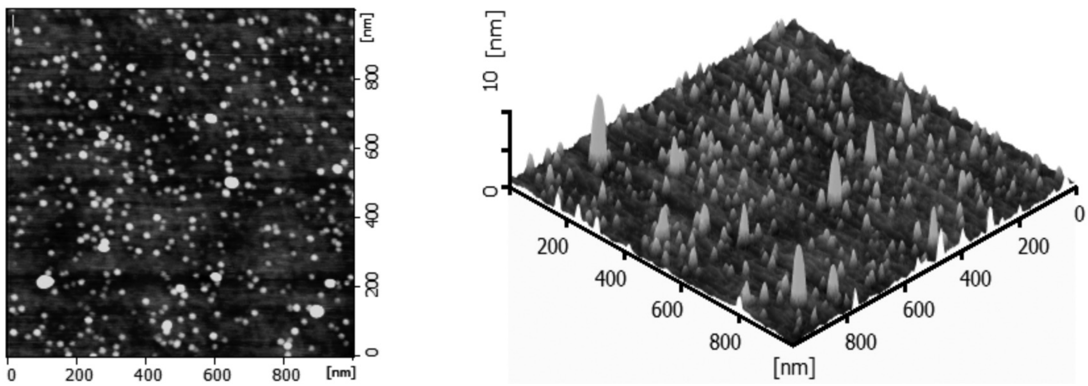


FIGURE 4. AFM images of $In_{0.5}Ga_{0.5}As$/GaAs QDs grown using 10 nm $In_{0.1}Ga_{0.9}As$ wetting layer with 1 min AsH_3 flow during cooling-down period

which AsH_3 encourages indium and gallium redistribution is still unclear. The AsH_3 flow during cooling-down period affects the kinetic mechanism of the dots. Binding energies might be changed due to changes in AsH_3 flow time which then lead to the change in the surface energy and in the state of thermodynamics equilibrium of the QDs ensemble (Riel et al. 2002). In principle, both kinetic limitations and thermodynamics can influence size, shape, uniformity, density and composition of the dots.

The $In_{0.5}Ga_{0.5}As$ QDs formed on GaAs under Stranski-Krastanov mode. This growth mode begins with initial two-dimensional layer deposition on the substrate. After a critical layer thickness is achieved, the surface transforms into three-dimensional highly strained dot structures that grow coherently on the surface (Kladko et al. 2007). In the Stranski-Krastanov mode, when the thickness reached over a critical value, two or more dots generally merge into one larger dot and the strain energy relaxes as dislocations are incorporated in the dots. This result can be seen in Figures 2 (b) and (c).

Thermodynamically, the balance between surface energy and strain energy of the system is thought to dictate the QDs formation, evolution, and defect introduction (Xie et al. 1999). QDs having different shapes have been observed in a number of material systems as a route for strain relaxation. Kita et al. (2000) using RHEED showed that the wetting layer does not change even after the start of the QDs formation. The present underlying layer before wetting layer causes the relaxation of lattice strain in QDs on the surface. In the Stranski-Krastanov growth modes, the structure and important role of the underlying layer before wetting layer affect the QDs formation process. The other study also shows the period of AsH_3 gas flow during cool-down influences the kinetic and thermodynamic process, this affect on the formation of the dots.

CONCLUSION

Self-assembled $In_{0.5}Ga_{0.5}As$/GaAs QDs samples have been grown with varying growth time and using $In_{0.1}Ga_{0.9}As$ underlying layer before self-assembled QDs by MOCVD technique. Different surface morphology of $In_{0.5}Ga_{0.5}As$ QDs analysed using AFM has been observed. Increasing the growth time had caused the formation of several large dots on the surface and increased the average size of QDs due to the dots coalescence. In the Stranski-Krastanov growth mode, two or more dot generally merges into one large dot and the strain energy relaxes as dislocations are incorporated in the dots, when the thickness increases over a critical value. The absence of $In_{0.1}Ga_{0.9}As$ underlying layer before the growth of $In_{0.5}Ga_{0.5}As$ QDs is one piece of evidence based on the self organized effect of the homo-species in the lattice mismatched hetero-interface. Underlying layer before wetting layer causes the lattice strain relaxation in QDs, so underlying layer is an important factor in the growth of self-assembled QDs. The AsH_3 flow during cool-down period affect the nucleation of the dots on the surface. This result is quite obvious to most of the researchers especially working on the growth of QDs. The growth parameters, growth conditions and growth methods are important factors in the fabrication of QDs using MOCVD or MBE system.

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