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 addition, 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

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-Kranstanov 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-Kranstanov 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$_{0.5}$Ga$_{0.5}$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$_{x}$Ga$_{1-x}$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 InGaAs 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 InGaAs/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 InGaAs QDs grown on GaAs (100) substrate using MOCVD. The Stranski–Krastanov (5K) growth mode of three-dimensional island formation in self-assembled InGaAs 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 InGaAs QDs were trimethylgallium (TMGa), trimethylindium (TMin) and arsenic (AsH3). Prior to growth, the substrate temperature was increased up to 700°C for 10 min under arsenic 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 InGaAs 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 AsH3 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 InGaAs underlaying layer were added before the growth of self-assembled InGaAs QDs (with deposition time of 4.5 s and after growth AsH3 flow time of 1 and 3 min). The structure of self-assembled InGaAs 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 InGaAs as QDs was taken in contact to AFM mode with a scan rate of typically 1 Hz.

**RESULTS AND DISCUSSION**

Self-assembled InGaAs 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 InGaAs 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 InGaAs QDs 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 InGaAs/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 × 1010 cm−2, respectively. This result is different compared to sample (b) and (c) where the existence of InGaAs 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 × 1010 cm−2 and 13 nm, 35 nm and 1.14 × 1010 cm−2, respectively. Increasing growth time of self-assembled InGaAs 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$_{0.5}$Ga$_{0.5}$As underlayer 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$_{0.5}$Ga$_{0.5}$As underlayer 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 $1.62 \times 10^{10}$ 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$_{0.5}$Ga$_{0.5}$As underlayer 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 reduces the critical layer thickness. It occurs due to the indium segregation above the In$_{0.5}$Ga$_{0.5}$As template. A significant contribution to QDs formation is made when a floating layer of indium forms on the top of the growing In$_{0.5}$Ga$_{0.5}$As layer and its thickness reaches a few monolayers. The general rule is that the thicker the In$_{0.5}$Ga$_{0.5}$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$_{0.5}$Ga$_{0.5}$As QDs using InAlAs underlying layer, the fraction of In$_{0.5}$Ga$_{0.5}$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$_{0.5}$Ga$_{0.5}$As QDs, showed that the InGaAlAs underlying layer was the key factor in the fabrication of small and dense In$_{0.5}$Ga$_{0.5}$As QDs.

The addition of aluminium to In$_{0.5}$Ga$_{0.5}$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$_{0.5}$Ga$_{0.5}$As underlayer.

Surface morphology of In$_{0.5}$Ga$_{0.5}$As QDs grown on thin In$_{0.5}$Ga$_{0.5}$As underlaying 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$_{0.5}$Ga$_{0.5}$As/GaAs QDs with 1 min AsH$_3$ flow during cooling-down are 16 nm, 6 nm, and $3.69 \times 10^{10}$ 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$_{0.5}$Ga$_{0.5}$As wetting layer with 3 min AsH$_3$ flow during cooling-down period](image-url)
which AsH\textsubscript{3} encourages indium and gallium redistribution is still unclear. The AsH\textsubscript{3} flow during cooling-down period affects the kinetic mechanism of the dots. Binding energies might be changed due to changes in AsH\textsubscript{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\textsubscript{0.5}Ga\textsubscript{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\textsubscript{3} gas flow during cool-down influences the kinetic and thermodynamic process, this affect on the formation of the dots.

**CONCLUSION**

Self-assembled In\textsubscript{0.1}Ga\textsubscript{0.9}As/GaAs QDs samples have been grown with varying growth time and using In\textsubscript{0.5}Ga\textsubscript{0.5}As underlying layer before self-assembled QDs by MOCVD technique. Different surface morphology of In\textsubscript{0.5}Ga\textsubscript{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\textsubscript{0.1}Ga\textsubscript{0.9}As underlying layer before the growth of In\textsubscript{0.5}Ga\textsubscript{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\textsubscript{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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