GaSb quantum dot morphology for different growth temperatures and the dissolution effect of the GaAs capping layer

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Abstract

We compare the characteristics of GaSb quantum dots (QDs) grown by molecular beam epitaxy on GaAs at temperatures from 400°C to 490°C. The dot morphology, in terms of size, shape and density, as determined by atomic force microscopy on uncapped QDs, was found to be highly sensitive to the growth temperature. Photoluminescence spectra of capped QDs are also strongly dependent on growth temperature, but for samples with the highest dot density, where the QD luminescence would be expected to be the most intense, it is absent. We attribute this to dissolution of the dots by the capping layer. This explanation is confirmed by atomic force microscopy of a sample that is thinly capped at 490°C. Deposition of the capping layer at low temperature resolves this problem, resulting in strong QD photoluminescence from a sample with a high dot-density.

1. Introduction

Self-assembled quantum dots (QDs) [1] are an area of intense interest due to their unusual electrical and optical properties, especially their potential for applications [2]. The vast majority of this research has concentrated on type-I QDs such as InAs/GaAs, but there is increasing interest in type-II dots. In particular, GaSb/GaAs QDs [3,4,5], which can confine the holes but not the electrons [6], display novel physical phenomena [7,8] and are of interest for applications in memory devices [9,10], lasers [11,12], and solar cells [13]. For all these applications performance is highly dependent on QD density, size, uniformity and composition, so these should be studied as a prerequisite for the realisation of any device. In turn, several factors affect QD characteristics such as deposition time of GaSb [14], III-V beam flux ratio [15], growth rate and growth temperature [16]. For example, the capability of QDs to trap carriers is determined by the thermal activation barrier or localisation potential which determines the storage time of a QD memory. Very recent research has shown that GaSb/(Al)GaAs is a very promising material combination for such applications. Localization energies were calculated using eight-band k.p theory, leading to the prediction of an extraordinary 10^6 years hole storage time for GaSb/AlAs dots [10]. However, it was also shown that the storage time is highly sensitive to the compositional purity of the dots. $GaAs_{0.4}Sb_{0.6}$/GaAs QDs have a storage time of ~0.5μs [10], whereas pure GaSb/GaAs dots have an estimated storage time of 13 minutes [10], which is approximately 1 billion times greater. The dilution of the QD Sb content is very likely to occur in the capping process immediately after the QD formation. Hence, the capping layer material or growth procedure plays a crucial role in determining the quality of the device. Although we have not probed the purity of the dots, we do demonstrate that the capping layer growth temperature has a remarkably strong influence on QD properties.

Our investigation has two strands. Firstly we have studied the effect of growth temperature on GaSb QDs formed using the Stranski-Krastanow (SK) mode, covering a wider and lower temperature range compared with the only previous report [16]. We find that the QD morphology is a strong function of the growth temperature. We note that a different method based on droplet epitaxy has recently been used to grow GaSb QDs, where a Ga droplet was exposed to Sb to produce GaSb QDs. Much lower growth temperatures were used in this alternative technique to control the QDs' characteristics [17]. Secondly, by growing the capping layer at the same temperature as the dots, we can also see the effect of capping the QDs via the photoluminescence (PL) spectra. Although the issue of capping and its influence on the QDs' properties has been studied extensively, this has been largely restricted to the InAs/GaAs system [18-20]. There are only a few papers on the growth of GaSb QDs [14-17,21-25,32], and the specific issue of capping has not been addressed, even though effects may be more pronounced than in InAs/GaAs. Indeed, our data show that Sb-As exchange during capping is prevalent, and that it leads to dissolution of the dots unless a low capping temperature is used.

2. Experimental details

A series of undoped samples comprising GaSb QDs embedded in a GaAs matrix were grown on semi-insulating (001) GaAs substrates using a solid-source VG V80H molecular beam epitaxy system. The growth temperature was determined by using a calibrated pyrometer. After oxide desorption of the substrate, a ~500 nm thick GaAs buffer layer was deposited. Then the temperature of the GaAs substrate was reduced under As_2 flux for GaSb deposition. Four samples A, B, C, and D were grown at temperatures *T*, of 400°C, 430°C, 460°C and 490°C, respectively. The growth rate was fixed at 0.3 monolayer (ML) s^{-1} with 7 s deposition time, producing nominally 2.1 ML of GaSb with a III/V ratio of 2 to produce GaSb QDs using the SK mode. The

GaAs growth rate was determined based on reflection high-energy electron diffraction (RHEED) oscillations during deposition of the GaAs buffer layer. The calibration procedure was repeated for each sample prior to the growth of the GaSb layer.

Growth of an undoped GaAs capping layer was preceded by exposing the GaSb surface to Sb for 8 s. The dots were then covered by a 5±2 nm GaAs layer at the same growth temperature and growth rate as for the GaSb deposition, before the substrate temperature was raised to 580^{o}C within 180 s under As flux. A further GaAs layer with a thickness of 100 nm was deposited for 360 s with the growth rate at 1 ML s^{-1}. Finally, a second layer of GaSb was deposited under the same growth conditions as the first GaSb layer for morphological studies of surface QDs.

Atomic force microscopy (AFM) was used to study the surface topography. A Digital Instruments Multimode Scanning Probe Microscope was used to study the surface of QD samples to obtain information about their size and density. AFM was run in tapping mode in air with the resonance frequency ~300 kHz and a setpoint which varied for each sample. We used a high quality silicon tip to better follow the contours of sample's surface. The optical properties were studied with PL measurements at 4.2 K by immersing the samples in liquid helium. The 532 nm line of a Nd:YVO_4 laser was used to excite the samples optically and a spectrometer fitted with an InGaAs detector was used to wavelength-resolve the collected emission. The laser light was transmitted to the sample with an optical fibre giving a spot size of diameter ~2 mm with a power density of ~7 W/cm^2.

3. Results and discussion

The AFM profiles of the samples surfaces reveal the density, size and height of the uncapped QDs. In Figure 1 (a)-(d) we show a series of 1-μm^2-sized AFM images that illustrate the different surface morphologies of GaSb QDs at growth temperature of (a) 400°C, (b) 430°C, (c)

460°C and (d) 490°C. The size and density of QDs on the samples surfaces are summarized in Table 1. At 400°C, the majority of the QDs are rather flat, elongated and rectangular in shape, and non-uniform in size. The average width and height of the dots are 42 nm and 3.4 nm respectively. It is possible that these flat rectangular dots are interface misfit (IMF) [4] dots and not SK dots, which are typically more rounded. However, some smaller and more rounded QDs with 1.7 nm height and 27 nm diameter remain after the growth temperature was increased and can be found on each of the samples. By increasing the growth temperature further, the elongated dots are transformed to nearly round dots. For the sample grown at 430°C (sample B), the average QD size is significantly reduced to 2.4 nm in height and 25 nm in diameter. There is a corresponding rise in the density from 2.7×10^{9} cm^{-2} to 4.2×10^{10} cm^{-2}, the QD shape is more semi-spherical, indicating the onset of SK growth mode, but the size distribution remains broad. At a growth temperature of 460°C, the QD size distribution is narrower and the shapes are improved, resulting in more rounded QDs. The average diameter of the dots is 35 nm and the height is 3.1 nm with a decrease in density to 3.4×10^{10} cm^{-2} which is due to coalescence of the dots [15]. Finally, at the highest growth temperature of 490°C, the coalescence of the dots has decreased the density to 1.8×10^{9} cm^{-2}. The diameter is essentially unchanged at 36 nm, whilst the height has increased to 6.4 nm implying that a large volume of Sb has been lost. Previous investigations of Sb condensation on GaSb using in situ surface X-ray diffraction found that the process involves diffusion and coarsening which changes the size of the dots [24]. However, it should be noted that there is strong evidence that the surface dots may be quite different from the buried dots, as is now generally believed. Firstly, gradually cooling the sample after the end of the growth effectively results in the inclusion of a growth interruption which is absent for the capped dots in the sample, and may change their morphology [25], perhaps even the dot density. In addition the capping itself can change the dot substantially [26-31], as we now go on to discuss.

Several papers have reported the effect of As_2 and As_4 on the growth of As/Sb heterostructures. They found that anion exchange was more active using As_2 [32-34] compared with As_4 [34]. Nosho *et al*. [34] concluded that by using As_4 the anion exchange can be reduced in GaSb layer, leading to better film stability. Hence, it is likely that the probability of producing high quality and pure GaSb QDs in a GaAs matrix are lower in our case, since As_2 is expected to increase the exchange of group V elements at the interface. An unsuitably high capping-layer temperature is another factor that may destroy the buried dots. Several groups have investigated the effect of the capping layer using different materials [29,30]. Segregation [27,31] and/or anion exchange may change the size, shape and the density of the buried dots during the growth.

Figure 2 shows the PL spectra from the four samples A to D. Sample A, which was grown at the lowest temperature, exhibits bright PL with the QD peak well separated from the wetting layer (WL) PL. The observation of QD PL from this sample does not exclude the possibility that the rectangular dots in this sample are IMF QDs [4]. In contrast, for samples B and C which have the highest surface dot areal density, it can be seen that the QD peak is hardly discernable or even absent, merging with the WL PL. In sample D, the QD peak is very weak compared to the intense WL peak, but nevertheless it can be clearly distinguished.

AFM shows that the surface morphologies of dots for samples A (T=400˚C) and D (T=490˚C) have a low density with a large lateral size, and are higher for sample D, whereas samples B and C have a large density of smaller dots. The PL data indicates that the large QDs with low density seem to survive the capping better than the high-density/small QDs. This is illustrated by the schematics in Figure 2.

At growth temperatures <490˚C, the heights of the dots are small and therefore they are prone to dissolution during capping. Samples B (T =430˚C) and C (T =460˚C) have a high density of small dots with a height that is similar to sample A, but due to the low capping temperature the dots in sample A can survive, as demonstrated by the bright PL emission. The intermixing/segregation of Sb from the GaSb layer during deposition of the capping layer has been demonstrated by Timm *et al.* [28] and Ulloa *et al.* [30] using cross-sectional scanning tunneling microscopy. The stronger bonding energy between As-Ga compared to Sb-Ga increases the anion exchange resulting in the shrinkage of the QDs [10]. The capping layer may change the QD volume, as was recently demonstrated by transmission electron microscopy of a GaSb/GaAs structure [31]. Segregation of Sb from the dot layer causes a reduction of volume of capped dots and the Sb becomes incorporated into GaAs layer resulting in a GaAsSb alloy, the Sb may even be evaporated from the sample surface at high growth temperature. For sample D, a weak but well-resolved QD PL peak is observed, implying that the largest dots can endure the high temperature capping.

In order to confirm this explanation, two further samples were grown. For the first of these (sample E), the dots were thinly capped with ~10 nm of GaAs at a nominal temperature of 490°C while for the second (sample F), which had the same sample structure as samples A to D, the dots were initially capped at 430°C. AFM data for sample E is shown in Figure 1 (e) with a scan area of 10×10 μm^2, i.e. 100 times larger than the other images in the figure. The surface shows micron-size elongated features, strongly indicative of melting of the QDs by the thin GaAs capping layer due to its high growth temperature. This is consistent with the rapid disappearance of the spotty RHEED pattern during the capping of this sample. However, the exact composition of the capping layer is unknown.

In contrast, the spotty RHEED pattern survived substantially longer during the capping of sample F. Figure 1 (f) shows the AFM image of this sample. The uncapped surface dots have a relatively high dot density of 2.8×10^{10} cm^{-2}, with 2.6 nm in height and 42 nm in diameter. Despite the difference in QD density on the sample surface for samples D and F, both PL spectra have approximately similar value of QD peak energy. The difference in surface QD density between samples D and F illustrates that the QD morphology is highly sensitive to the growth temperature. However, the point to be stressed here is the importance of the temperature of the capping layer. In marked contrast to samples B to D, sample F shows very strong QD PL (Figure 3) with high intensity, well separated from WL peak; clear proof that capping at low temperature is needed to preserve the dots.

4. Conclusions

We have conducted atomic force microscopy and photoluminescence experiments on six GaSb/GaAs QDs samples grown by molecular beam epitaxy. Atomic force microscopy data show that the quantum dot morphology is highly sensitive to growth temperature in the 400 to 490°C range. At 400°C the dots are large and rectangular with low density and poor uniformity. As the growth temperature is raised there is an increasing tendency towards a more uniform distribution of rounded dots, with improved diffusivity leading to increasing QD size (volume) and decreasing density. In contrast, photoluminescence measurements on the same samples show that strong exchange/intermixing effects during capping make the dots highly prone to dissolution unless the capping temperature is low. This was further demonstrated by atomic force microscopy images of a sample that was thinly capped at 490°C, and by the substantially improved photoluminescence spectrum of a sample for which the dots were grown at high temperature and capped at low temperature.

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Figure Captions

Figure 1: AFM images of uncapped QDs for growth temperatures of (a) 400°C (sample A), (b) 430°C (sample B), (c) 460°C (sample C), and (d) 490°C (sample D). (e) and (f) are AFM images of QDs thinly capped at 490°C (sample E) and uncapped QDs of sample F.

Figure 2: PL spectra of the samples normalized to the GaAs intensity. The energy ranges where we expect to find quantum dot (QD), wetting layer (WL), and GaAs photoluminescence are shown. The schematics on the right of the figure depict the dissolution of the QDs as a result of GaAs capping.

Figure 3: Comparison of PL spectra for samples D and F, for which the QDs were grown at 490°C and capped at 490°C and 430°C, respectively.

Table 1: Summary of the uncapped GaSb QD size and density as measured by AFM.

Sample	Growth temperature (°C)	Height (nm)	Diameter (nm)	Density (cm^{-2})
A	400	(a) 1.7 ± 0.3 (b) 3.4 ± 0.5	(a) 27 ± 2 (b) 42 ± 6	2.7×10^9
B	430	2.4 ± 0.2	25 ± 5	4.2×10^{10}
C	460	3.1 ± 0.9	35 ± 8	3.4×10^{10}
D	490	6.4 ± 2	36 ± 8	1.8×10^9

[†] There are two population of size distribution in sample A. Majority dots in sample A are rectangular and elongated. The value given here is the width.

FIGURE 1: (M. Ahmad Kamarudin)

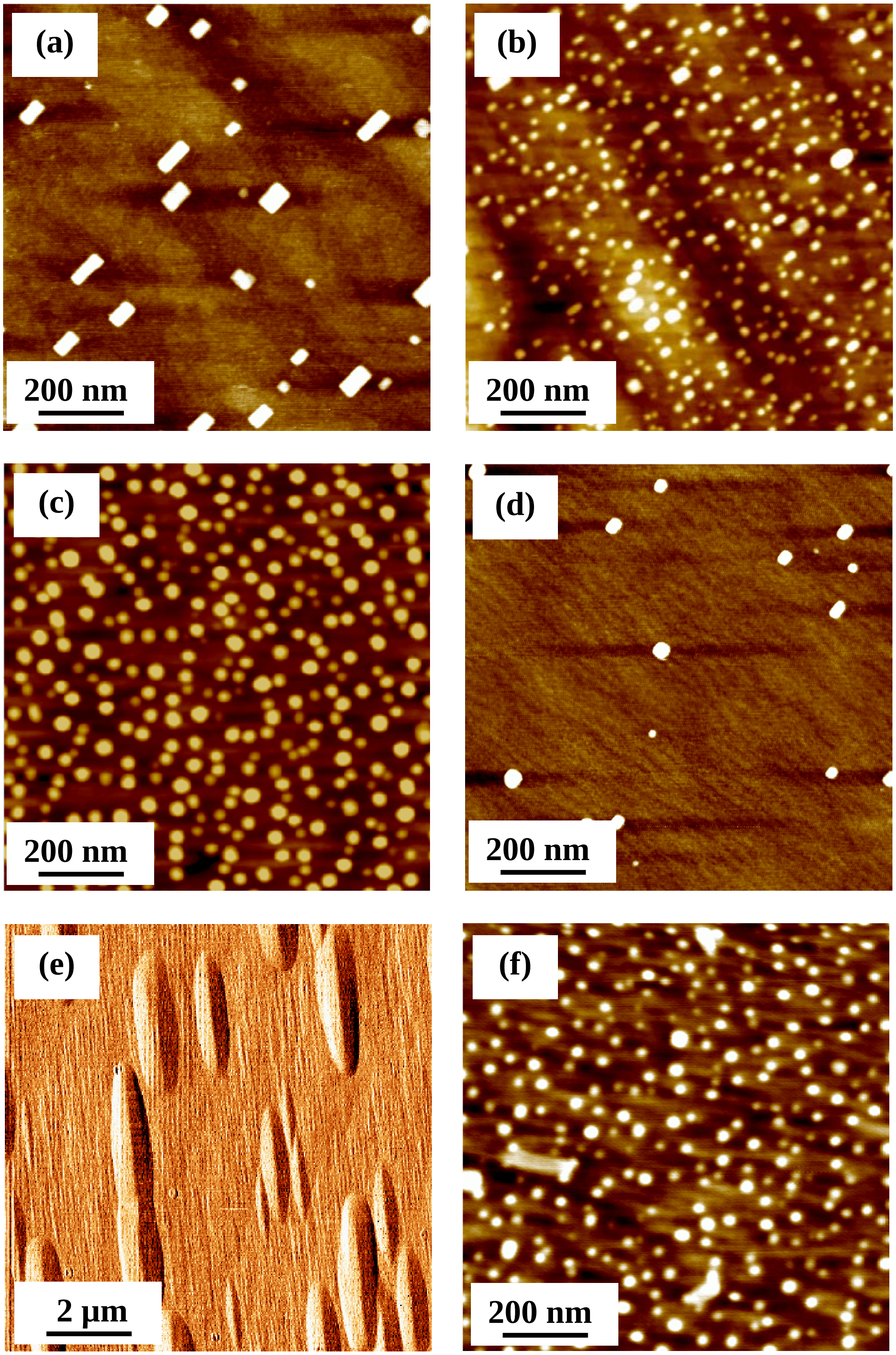

FIGURE 2: (M. Ahmad Kamarudin)

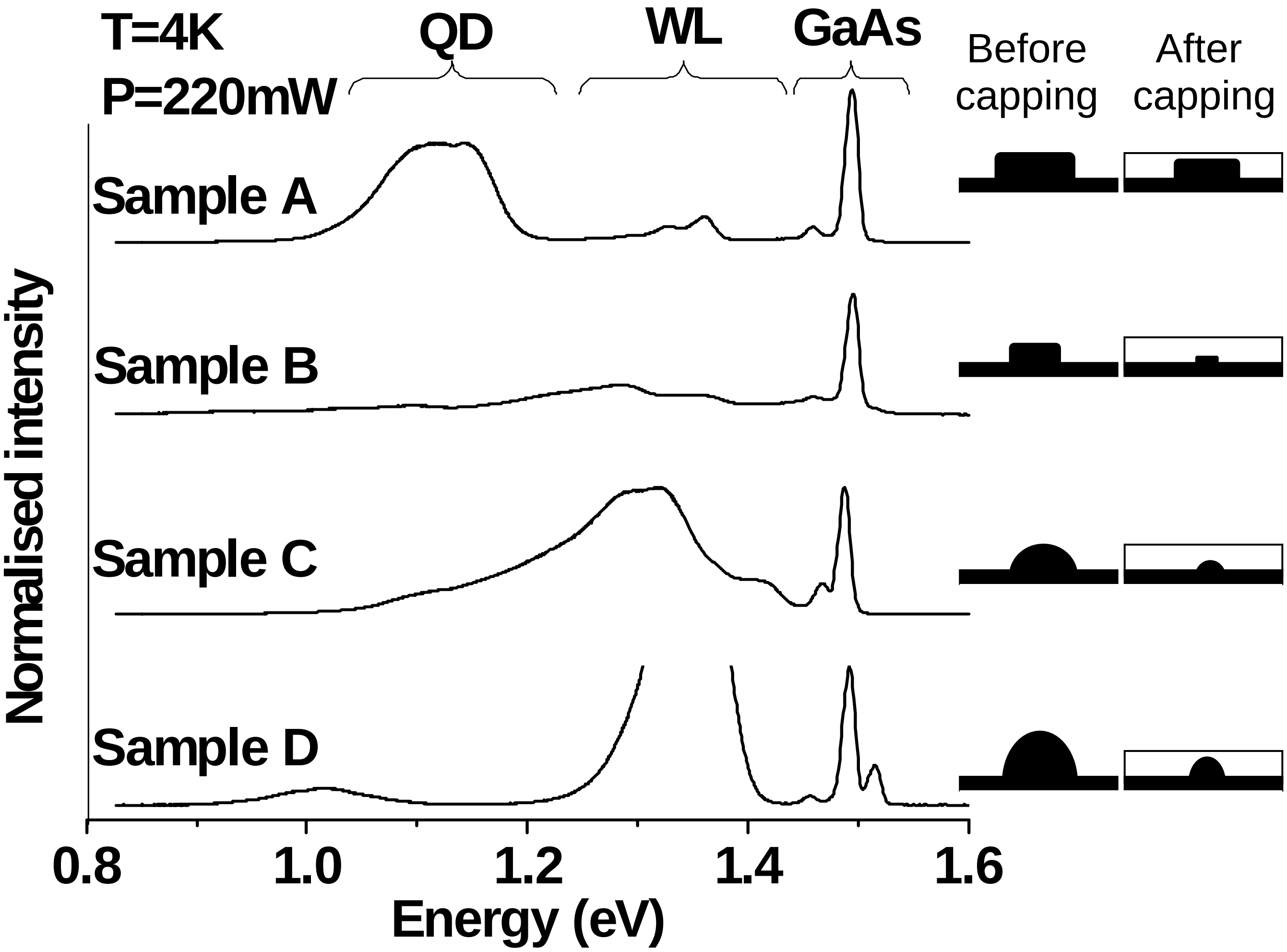

FIGURE 3: (M. Ahmad Kamarudin)