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Journal of Crystal Growth 278 (2005) 57–60
JOURNAL OF CRYSTAL GROWTH
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Indium segregation during multilayer InAs/GaAs(0 0 1) quantum dot formation

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Available online 26 January 2005

Abstract

Indium segregation during the growth of multilayer InAs/GaAs(0 0 1) quantum dot (QD) structures has been studied using reflection high-energy electron diffraction (RHEED) measurements of the critical coverage (θ_{crit}) for second layer QD formation. A model bilayer structure was used in order to separate the effects of segregation and strain. The structure comprises an upper QD layer formed on top of a buried two-dimensional InAs layer. Growth temperature and the GaAs spacer layer thickness (S) are both found to have a significant effect on θ_{crit}. Indium segregation during growth of the capping layer leads to the presence of a surface In adatom population prior to deposition of the second InAs layer. Segregation occurs for S up to 8 nm at 510 °C, this value being reduced by ~50% at 450 °C.

PACS: 61.14.Hg; 66.30.Pa; 68.65.Hb; 81.15.Hi

Keywords: A1. Growth models; A1. Reflection high energy electron diffraction; A1. Segregation; A2. Molecular beam epitaxy; B2. Semiconducting III–V materials

1. Introduction

Low-dimensional semiconductor structures such as quantum dots (QDs) are attracting considerable interest due to their potential application in a wide range of optoelectronic devices. With lasers and other devices being produced at 1300 nm [1], the InAs/GaAs(0 0 1) QD system has been the most intensively studied, and longer wavelength QD emitters are now being demonstrated at ~1500 nm [2]. Multilayer QD structures are required for these applications with the QD layers often separated by relatively small spacer layers (S~10 nm). Several reports have shown that the growth of multilayer InAs/GaAs QDs can be complex with In/Ga intermixing, strain, segregation and spacer layer morphology all having a significant influence on the resulting structural and optical properties [3–8].

The role of In segregation during multilayer InAs/GaAs QD growth has received surprisingly

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0022-0248/$ - see front matter
doi:10.1016/j.jcrysgro.2004.12.053

little attention, although segregation effects are known to be an important issue in the formation of InGaAs quantum wells [9]. One key problem is the difficulty in deconvoluting segregation and strain effects in multilayer QD growth. The strain fields from a buried QD layer extend through relatively large spacer layers ($S<40$ nm), the actual magnitude depending on factors such as the size of the initial QD layer [3–5]. By contrast, In segregation is likely to be limited to much lower values of S, typically <10 nm [10].

In this study we use a model bilayer structure to quantify In segregation during the growth of second layer InAs/GaAs QDs. The inhomogeneous effects of strain are eliminated by using a buried two-dimensional (2D) InAs layer (1.75 ML) instead of a QD layer. Reflection high-energy electron diffraction (RHEED) is used to monitor the 2D→3D growth mode transition (the critical thickness, θ_{crit}) as a function of temperature (T_S) and S. The results show that segregation during growth at 510 °C is significant for $S<8$ nm.

2. Experimental details

All samples were grown on epi-ready GaAs(0 0 1) substrates (n^+, Si doped) in an MBE system (DCA Instruments) equipped with RHEED. The InAs growth rate (0.016 ML s^{-1}) was carefully controlled by growing a single QD layer at the temperature of the (2×4)–$c(4\times4)$ reconstruction change (510 °C). The growth mode transition (monitored by the abrupt appearance of chevrons in the $[\bar{1}10]$ azimuth of the RHEED pattern) always occurred at 130 s ± 2 s ($\theta_{crit}=2.08$ ML) under these conditions. More details of the growth process can be found elsewhere [4].

3. Results and discussion

Fig. 1 shows the influence of substrate temperature on θ_{crit} for a single InAs/GaAs QD layer, and also for the bilayer structure in which θ_{crit} was recorded for a QD layer formed on top of a buried 2D InAs layer of thickness 1.75 ML (~0.5 nm).

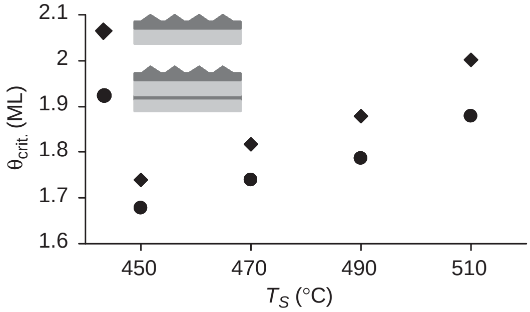

Fig. 1. Plot of critical coverage (θ_{crit}) as a function of substrate temperature (T_S) for InAs/GaAs QD formation in a single QD layer and a bilayer sample in which the initial layer comprises a 2D InAs layer of thickness 1.75 ML.

The GaAs spacer layer thickness was 4 nm. Temperature clearly has a pronounced effect in both cases and θ_{crit} is reduced from 2.1 ML in the single layer and 1.9 ML in the bilayer structure to 1.8 and 1.75 ML, respectively, when T_S is reduced from 510 to 450 °C. Temperature-dependent changes in θ_{crit} for single InAs/GaAs QD layers have been reported previously [11,12] and rationalised in terms of alloying in the 2D wetting layer, which is more pronounced at higher temperatures [13]. The strain which builds up during InAs deposition is partly relieved as a result of this intermixing, and when T_S is increased, the amount of InAs that has to be deposited in order to achieve the degree of strain necessary to cause the 2D→3D growth mode transition increases.

The same basic trend is seen in the double layer structure—the only difference compared with the single QD layer structure is that θ_{crit} is reduced by between 0.06 (450 °C) and 0.12 ML (510 °C). To rationalise these small differences it is necessary to consider the segregation of In during growth of the 4 nm GaAs capping layer which leads to the presence of a surface In adatom population before deposition of the second InAs layer. The amount of InAs required to reach θ_{crit} is therefore reduced when compared to the single QD layer structure. The observed reduction in θ_{crit} cannot be a result of strain since the underlying InAs layer is a pseudomorphic 2D layer.

Verification of the existence of this surface In adatom population can be obtained from a more detailed analysis of the RHEED patterns observed during growth. Directly after growth of the GaAs spacer layer, the RHEED pattern shows a mixed c(4 × 4) and (1 × 3) surface reconstruction in the [$\bar{1}$10] azimuth. The (1 × 3) structure is direct evidence for the presence of an $In_xGa_{1-x}As$ surface alloy [13], whilst the c(4 × 4) phase is the stable surface reconstruction for GaAs at this temperature. However, if the capped surface is annealed at a temperature of 580 °C (for 10 min) prior to deposition of the second InAs layer, the reconstruction changes to a (2 × 4) phase, indicative of pure GaAs and loss of In due to desorption from the surface. In this case, the θ_{crit} value for the dots in the upper layer corresponds exactly to the value obtained for the isolated single QD layer grown at the corresponding temperature.

The In segregation length will depend significantly on the GaAs overgrowth temperatures. This is confirmed by comparing the θ_{crit} values for the two types of QD structure grown at different substrate temperatures. At 450 °C, this difference—which is a measure of the amount of InAs on the surface and therefore the segregation efficiency—is less pronounced (0.06 ML) than at 510 °C (0.12 ML). We estimate therefore that In surface segregation is already reduced by ~50% if the 2D InAs layer is overgrown with GaAs at 450 °C. For even lower deposition temperatures the two values for θ_{crit} should eventually converge, because of the low rate of In segregation.

Fig. 2 shows how θ_{crit} varies as a function of spacer layer thickness for a series of QD samples in which the basic structure comprised an InAs 2D layer with a thickness of 1.75 ML (i.e. $<\theta_{crit}$) grown at 510 °C, followed by a GaAs spacer layer of varying thickness S. The second InAs layer was then deposited without any growth interruption. There is a significant drop in θ_{crit} as S is reduced below 6 nm (~21 ML). This reduction is again due to the existence of surface In adatoms as a consequence of segregation through the GaAs spacer layer. The thinner the GaAs spacer layer, the higher the concentration of surface In adatoms and the more pronounced the reduction in θ_{crit}. In contrast to the InAs-on-GaAs interface, the GaAs-on-InAs interface is not atomically sharp.

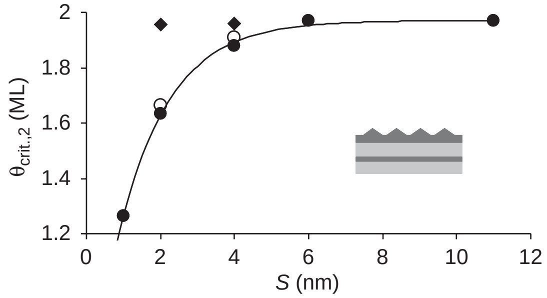

Fig. 2. Plot of θ_{crit} as a function of spacer layer thickness (S) for the bilayer sample with the sequence 2D InAs layer/GaAs spacer layer/QD layer (full circles). The diamonds (clear circles) show samples that were annealed for 10 min at 580 °C (510 °C) after growth of the spacer layer. The solid line is based on an exponential fit for the In segregation efficiency.

This behaviour is again confirmed by close observation of the RHEED patterns. After deposition of 2 nm GaAs, the RHEED pattern along the [$\bar{1}$10] azimuth shows a (1 × 3) reconstruction, indicative of an $In_xGa_{1-x}As$ alloy. For $S = 4$ nm, the RHEED pattern reveals a mixed phase, with the (1 × 3) reconstruction dominating, although the existence of the c(4 × 4) reconstruction indicates some domains of pure GaAs. For $S = 6$ nm, the situation changes and the c(4 × 4) pattern dominates, although the (1 × 3) reconstruction can still be observed. The RHEED pattern is essentially pure c(4 × 4) for $S = 8$ nm, indicating that the surface In concentration is negligible and it is reasonable to conclude that the segregation of In atoms through the growing GaAs layer occurs up to a maximum thickness of 8 nm for these growth conditions.

The effects on θ_{crit} of the introduction of a growth interrupt and annealing step can also be seen in Fig. 2. For the samples with $S = 2$ and 4 nm, a growth interruption of 10 min was performed before depositing the second InAs layer. The clear circles represent an interrupt with unchanged temperature (i.e. 510 °C) whereas the diamonds correspond to samples which were annealed at 580 °C before second layer deposition. Annealing at the growth temperature before second layer InAs deposition does not change the RHEED pattern, regardless of the value of S. For this reason θ_{crit} in the second layer remains unchanged and the rate of In desorption is clearly very low at this temperature. During annealing at higher temperatures (580 °C), however, the RHEED pattern changes in all cases within 1–2 min into a (2×4) pattern, the stable surface reconstruction for GaAs(0 0 1) at this temperature. This change is due to the thermal desorption of the segregated In atoms and explains why the growth of the upper QD layer after annealing at higher temperatures is similar to the growth of an isolated QD layer.

An estimate of the segregation efficiency can be obtained using a simple model originally developed by Toyoshima et al. [10] for InGaAs growth on GaAs(0 0 1). If the fraction of In which segregates through each ML of GaAs during deposition is defined by R, then the amount of segregated InAs in the top layer (X_n) is given by $X_n = X_0 R^n$, where X_0 is the amount of free InAs (in ML) on the starting surface when GaAs growth is initiated, and n is the number of deposited GaAs monolayers. The best fit to the experimental data is shown as the solid line in Fig. 2, with $X_0 = 1.46$ ML and $R = 0.814$. It should be noted that X_0 is significantly different from the total amount of InAs deposited (1.75 ML). This discrepancy can be rationalised by assuming that only a fraction of the deposited In is incorporated as InAs (0.29 ML), the rest remaining afloat on the surface. It should be emphasized that this simple model is based on a step-by-step and not a continuous flow mechanism. Strictly speaking the model is only valid for $n \geqslant 2$, otherwise the amount of surface In in the top layer would be >1 ML (for $n < 2$).

It is interesting to note that the In segregation ratio ($R = 0.814$) is close to the one determined by Toyoshima et al. (0.81–0.85), who performed growth at the slightly higher temperature of 520 °C [10]. This suggests that the segregation of In is qualitatively and quantitatively similar for overgrowth of InAs with GaAs and the deposition of InGaAs on GaAs. Considering the higher In flux used in the work of Toyoshima et al. ($0.09\,\mathrm{ML\,s^{-1}}$ compared with $0.016\,\mathrm{ML\,s^{-1}}$) this means that the segregation process is controlled thermodynamically under these conditions.

4. Conclusions

Accurate measurement of the critical thickness during second layer InAs/GaAs QD formation has been used to provide quantitative information regarding the segregation of In through the GaAs spacer layer in a multilayer QD structure. Segregation during growth means that a surface population of In exists before the deposition of the second InAs layer, and the amount of InAs required for the 2D→3D growth mode change is therefore reduced. This effect only operates for GaAs spacer layers <8 nm for a growth temperature of 510 °C and is reduced by more than 50% if growth is carried out at 450 °C. The segregation effect is eliminated by applying a high temperature (~580 °C) annealing step before growth of the upper QD layer.

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