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Self-assembled quantum dot formation during the growth of $In_{0.4}Ga_{0.6}As$ on GaAs(0 0 1) by metal-organic vapor phase epitaxy: The role of In segregation

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Abstract

The stabilization of planar $In_{0.4}Ga_{0.6}As$ on GaAs, above thickness typically reported to undergo spontaneous island formation, has been observed. The effect of growth temperature, AsH_3 partial pressure, and $In_{0.4}Ga_{0.6}As$ thickness on island size and density was evaluated. Planar films are stabilized by low growth temperatures and high AsH_3 partial pressures. Spontaneous island formation is enhanced by high growth temperatures and low AsH_3 partial pressures. The results presented are interpreted within the phenomenology of In segregation to the surface, which has been identified previously for the InGaAs/GaAs system. Based on equilibrium models for the critical thickness for island formation, the critical segregation layer thickness for island formation is predicted to be 0.6 nm.
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1. Introduction

The two-dimensional to three-dimensional transformation of compressively strained epitaxial layers has been identified as a route to self-assembled nanostructures. This transformation occurs for a large number of semiconductor systems and has been investigated extensively in attempts to optimize this process [1,2]. InGaAs on GaAs(0 0 1) is one such system that has optoelectronic applications [3,4]. The growth of $In_{0.4}Ga_{0.6}As$ on GaAs by metal-organic vapor phase epitaxy (MOVPE) to form self-assembled quantum dots (SAQD) with a ground state resonance at 1.24 eV (1.0 μm) has been investigated. There is an ongoing debate about the role of thermodynamics relative to kinetics during the islanding process. Thermodynamics in the form of surface segregation plays a role in SAQD formation. If surface segregation can be controlled, three-dimensional transformation can be suppressed to extend planar growth of highly strained layers. Enhanced three-dimensional growth could produce uniform, dense ensembles of SAQD.

Surface segregation occurs during the growth of alloys when one of the components is rejected from the bulk resulting in a surface phase that is enriched relative to the bulk composition. The surface phase is an alloy with a composition different from that of the bulk. The surface segregation of In during InGaAs growth on GaAs growth is well documented [5–7]. Surface segregation limits the growth of abrupt quantum wells and introduces interface scattering into InGaAs/AlGaAs high electron mobility devices [8,9]. For In(Ga)As deposition on GaAs, segregation of In to the surface has been observed using *in situ* strain sensitive techniques, surface science tools, and electron microscopy. The amount of strain-induced curvature of thin substrates during InAs growth suggests an amorphous or adsorbed layer exists on the surface [10].

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These results are consistent with thermodynamic analysis of highly strained heteroepitaxy, which also predicts an amorphous layer [11]. Angle resolved X-ray photoemission spectroscopy and Auger electron spectroscopy have been used to estimate the thickness of the segregated layer to be several monolayers thick [5] and reflection high-energy electron diffraction has been used to estimate the surface composition from the two-dimensional to three-dimensional transition [12]. Extensive transmission electron microscopy (TEM) and complementary analytical techniques have confirmed directly that In segregation does occur during the growth of In(Ga)As on GaAs [13–15].

The work presented confirms and extends previous results which suggest that process conditions can suppress or enhance three-dimensional growth. This investigation examines the growth of $In_{0.4}Ga_{0.6}As$ under different AsH_3 partial pressures and different growth temperatures. Lower growth temperatures have been identified as an effective route to reducing surface segregation [8,9], but the role of AsH_3 on SAQD formation has only been speculated [16–18]. The present study identifies conditions where surface segregation is suppressed and enhanced. It interprets previous observations and the current results in terms of surface segregation, predicting conditions that will limit or enhance SAQD formation. The results are explained in terms of changes to the thickness of the segregated layer on the surface.

2. Experimental details

$In_{0.4}Ga_{0.6}As$ was deposited by MOVPE in a vertical growth chamber at 70 Torr with the wafer holder rotating at 900 rpm. Triethyl gallium (TEGa) and trimethyl indium (TMIn), both held at 637 Torr and 20 °C, were the group III metal-organics used and arsine (AsH_3) was the group V hydride utilized. The general structure considered in this report is shown in Fig. 1. GaAs (0 0 1) $\pm 0.1^\circ$ substrates were heated to 650 °C and annealed under 0.28 Torr AsH_3 partial pressure for 5 min to desorb the surface oxide. After cooling to 600 °C, a 200-nm GaAs buffer was grown. The sample was then cooled under 0.28 Torr AsH_3 to the temperature desired for $In_{0.4}Ga_{0.6}As$ growth. The conditions for $In_{0.4}Ga_{0.6}As$ growth varied and are discussed in detail below. After deposition of the $In_{0.4}Ga_{0.6}As$, the chamber was purged for 10 s under hydrogen to remove AsH_3 and metal organics. After the hydrogen purge, the $In_{0.4}Ga_{0.6}As$ layer was capped with 3 nm of GaAs grown at the $In_{0.4}Ga_{0.6}As$ growth temperature. The capped sample was heated to 600 °C for growth of 300 nm of GaAs. The sample was subsequently cooled for growth of uncapped $In_{0.4}Ga_{0.6}As$ under the same conditions that were used for the buried layer. Three different sets of experiments were considered. In the first set, the growth temperature was set at 450, 500, or 550 °C with an AsH_3 partial pressure of 0.28 Torr and an InGaAs thickness of 4.5 nm. For the second set, the AsH_3 partial pressure was 0.28, 0.14, 0.06, and 0.02 Torr at a growth temperature of 500 °C and a thickness of 4.5 nm. The third set used 500 °C with an AsH_3 partial pressure of 0.02 Torr and the thickness of the InGaAs layer was set to 3.0, 2.0, or 1.5 nm. The growth rate of $In_{0.4}Ga_{0.6}As$ was held constant in all studies at $0.2\,nm\,s^{-1}$. Photoluminescence (PL) and atomic force microscopy (AFM) were performed on each sample. The InGaAs growth rate and composition were calibrated using high-resolution X-ray diffraction from InGaAs/GaAs superlattices around the symmetric (0 0 4) reflection and glancing incidence (2 2 4) and ($2\bar{2}4$) reflections, analyzing the results to account for strain relaxation [19]. Photoluminescence was performed at room temperature using an Accent RPM2000 photoluminescence mapping system excited with a 785-nm laser at a power density of $4.5\,W\,cm^{-2}$ and detected through a 530 nm high-pass filter with an InGaAs detector. AFM was performed in air at room temperature using a Digital Instruments Nanoscope III in tapping mode using silicon tips with a 5-nm radius.

3. Results

The morphologies of InGaAs films varies significantly as the growth temperature is varied at constant AsH_3 partial pressure and InGaAs coverage, as depicted in Fig. 2. At low growth temperatures (Fig. 2a, 450 °C) the morphology is dominated by a low density ($46\,\mu m^{-2}$) of small elliptical islands. The islands range from 30×20 nm to 45×30 nm in lateral size, with heights of 4 ± 2 nm. As the temperature is increased to 500 °C (Fig. 2b), the density of three-dimensional islands drops to near-zero. Only small two-dimensional, monolayer islands are observed with a density of approximately $100\,\mu m^{-2}$. As the temperature is increased further to 550 °C (Fig. 2c) the density of larger

Layer description	Thickness [nm]	Growth temperature [°C]	AsH_3 partial pressure [Torr]
Purge	10 seconds	Varied	None
Surface InGaAs SAQD	Varied	Varied	Varied
High temperature GaAs	3200	600	0.28
Low temperature GaAs	3	Varied	0.28
Purge	10 seconds	Varied	None
Buried InGaAs SAQD	Varied	Varied	Varied
GaAs buffer	200	600	0.28
GaAs substrate		Annealed at 650	0.28

Fig. 1. Diagram of structure considered and the growth conditions used for each layer.