Critical film thickness dependence on As flux in $In_{0.27}Ga_{0.73}As/GaAs(001)$ films

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The transition between planar and nonplanar growth is examined for compressively strained $In_{0.27}Ga_{0.73}As/GaAs(001)$ films using reflection high energy electron diffraction, atomic force microscopy, and scanning tunneling microscopy (STM). For a narrow range of temperature and composition, the critical thickness (t_{SK}) is strongly dependent on As flux. For high values of As flux, t_{SK} increases by more than a factor of 2. The morphology of three-dimensional islands formed during the initial stages of nonplanar growth is also characterized by high resolution STM. © *2007 American Institute of Physics*. [DOI: 10.1063/1.2476259]

At moderate and high misfit strains, the growth of heteroepitaxial films on a crystalline substrate often occurs in the Stranski-Krastanov (SK) growth mode,[1] in which a planar wetting layer is initially formed through coherent two-dimensional growth, followed by the nucleation of coherent three-dimensional (3D) islands above some critical film thickness (t_{SK}). This growth mode has been observed experimentally for a variety of systems, including $In_xGa_{1-x}As$ films on GaAs(001) or InP(001) substrates,[2–14] which are of particular interest for quantum dot applications. Their use relies in large part on the ability to control the SK transition and on island shape and size, which impact quantum confinement.

Several key observations have been made from the previous studies of these systems. For As-rich growth, t_{SK} increases with decreasing misfit strain (decreasing In content) and the transition to 3D growth can be completely eliminated at In compositions below 20%.[2,3] Increasing the deposition temperature decreases t_{SK} due to the increased diffusion lengths, which facilitate island formation,[4,5] and to the In enrichment of the film surface, which increases the driving force for 3D islanding.[6] The converse, an increase in t_{SK} with temperature occurs when severe In desorption decreases the misfit strain in the wetting layer.[5] Very low As fluxes can completely suppress 3D islanding when the growth occurs in group III terminated conditions, presumably due to an alteration in the surface energy.[7] An increase in t_{SK} was observed for InAs/GaAs(001) films at As:In flux ratios <2:1,[8] concurrent with the appearance of metal terminated regions on the surface. Above t_{SK}, increasing the As flux delays the growth of islands and the formation of surface pits.[9]

This letter presents observations concerning less understood aspects of the transition to 3D growth for InGaAs alloys. It is shown that for moderate misfit strains and over a narrow range of temperatures, t_{SK} can be increased substantially by increasing As flux during deposition. This effectively provides an additional parameter to control the growth of thick but planar pseudomorphic layers and has important potential applications in device growth. Additionally, the detailed morphology of 3D islands formed at moderate misfits is revealed by scanning tunneling microscopy (STM). Facets near the {114}, {216}, and {138} orientations are observed for the islands.

All films were grown using solid source molecular beam epitaxy. The film composition, deposition rate R, and As flux F_{As4} were calibrated using reflection high energy electron diffraction (RHEED) oscillations, and the temperature was measured using an optical pyrometer. Following oxide desorption, a 0.5 μm GaAs buffer layer was grown at T=580 °C, $F_{As4}\approx 2.0$ ML/s, and R_{buffer}=0.54 ML/s, which resulted in a smooth, (2×4) reconstructed surface. The substrates were then cooled under the same As flux to the film growth temperature. $In_{0.27}Ga_{0.73}As/GaAs(001)$ films (1.9% compressive strain) were grown in the range of 485 °C<T<495 °C at R_{film}=0.74 ML/s at various F_{As4}. The critical thickness for 3D growth was determined from the increase in the intensity of the RHEED specular spot, followed by the appearance of a spotty RHEED pattern. The STM chamber is connected to the growth chamber which enables high resolution imaging of fresh surfaces transferred *in vacuo*. All STM images shown are filled states taken at typical imaging parameters of −3 V and 100 pA. The surface morphology was also examined at larger scales by *ex situ* tapping mode atomic force microscopy (AFM).

Figure 1 shows a plot of t_{SK} as a function of As flux for $In_{0.27}Ga_{0.73}As/GaAs(001)$ films grown under As-rich conditions at T=485 and 495 °C. The critical film thickness decreases with increasing temperature at a given As flux as expected during growth at temperatures for which the In desorption is insignificant.[4,5] Figure 1 also shows an increase in t_{SK} with F_{As4} at constant temperature. The effect is more pronounced at T=485 °C and for V/III flux ratios greater than 2:1, and is limited to low misfit strains. $In_{0.53}Ga_{0.47}As/GaAs(001)$ films, not shown, transitioned to 3D growth at $t_{SK}\approx 3$ ML regardless of the As flux.

The t_{SK}-F_{As4} dependence arises from the effects of the As flux on the kinetic factors limiting the nucleation and growth of 3D islands, which may be evident only at low or moderate strains. It has been shown that high As flux decreases the equilibrium adatom density at the film surface,[10]

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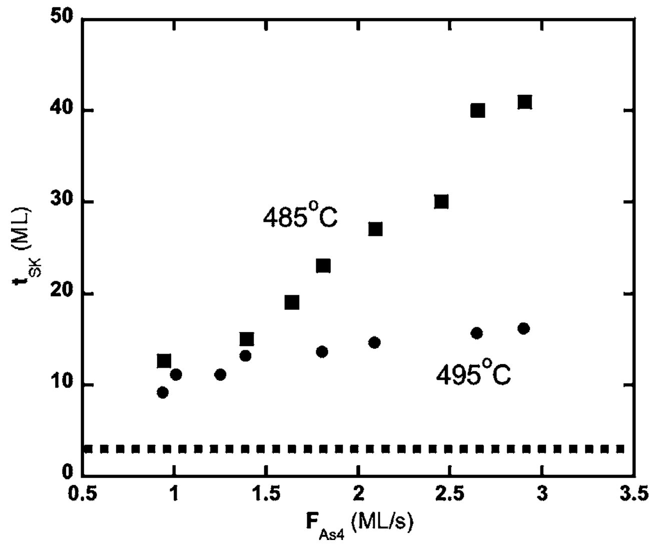

FIG. 1. Critical film thickness for 3D island growth vs As flux for $In_{0.27}Ga_{0.73}As/GaAs(001)$ films (1.9% compressive strain) at different growth temperatures. The dashed line indicates the transition to 3D growth for higher misfit $In_{0.53}Ga_{0.47}As$ films, which is ≈3 ML regardless of the As flux.

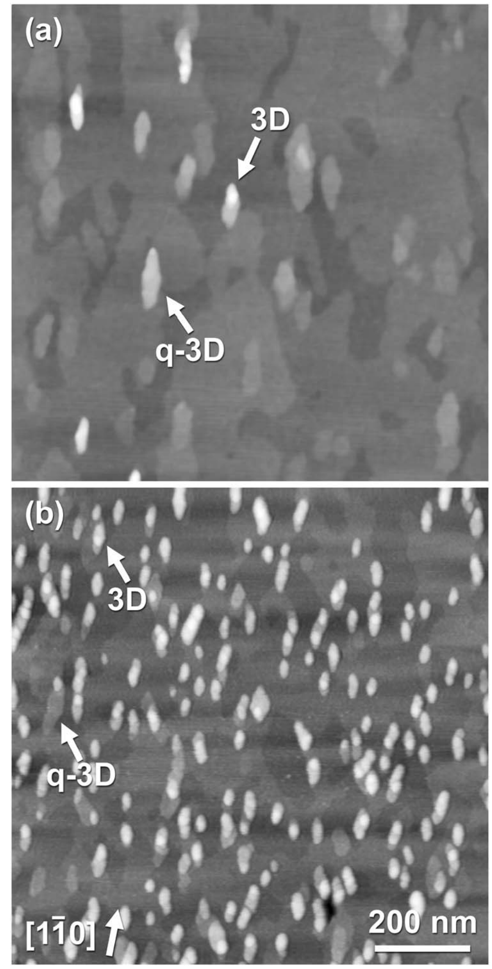

FIG. 2. 1×1 μm^2 AFM images of 25 ML $In_{0.27}Ga_{0.73}As/GaAs(001)$ films grown at T=490 °C using (a) F_{As4}=2 ML/s and (b) F_{As4}=1.8 ML/s. Island shapes, quasi-3D (q-3D) and 3D, are identified by arrows. The height scale is 10 nm. A surface pit is also observed within a group of 3D islands in (b).

as well as the diffusion lengths of cations.[11,12] Both these factors indicate that the nucleation rate of 3D islands should decrease with increasing F_{As4}, consistent with the present results. The disappearance of this effect at higher deposition temperatures is expected, as the adatom density and diffusion length increase exponentially with temperature, while the adatom density has only a power law dependence on As pressure.[10] At higher misfit strains, the increased driving force for 3D islanding could overcome the kinetic limitations imposed by the As flux, thereby diminishing the dependence of t_{SK} on F_{As4}. Other factors may also contribute to the observed t_{SK}-F_{As4} dependence. Ternary $In_xGa_{1-x}As$ alloys have mixed reconstructions, which vary with the surface composition and strain.[13,14] While a clear dependence of t_{SK} on these reconstructions has not been observed for As-rich surfaces, it is likely that their relative coverage affects the surface energy, as well as the surface diffusivity and the attachment rate of adatoms at island edges, thus affecting island growth.

The surface morphology was investigated by *ex situ* AFM and by *in vacuo* high resolution STM. Figure 2 shows large scale AFM images of films grown near t_{SK}, i.e., at the initiation of 3D growth. The films were deposited at the same temperature (490 °C) and to the same thickness (25 ML) but using slightly different As fluxes, such that t_{SK} was somewhat higher ($t_{SK}\sim 25$ ML) in Fig. 2(a) than in Fig. 2(b) ($t_{SK}\sim 22$ ML). Both surfaces show 3D features formed on a planar wetting layer that had undergone some two-dimensional roughening prior to the SK transition. The morphology of these films follows the behavior expected from the t_{SK}-F_{As4} dependence observed by RHEED, i.e., a higher island coverage is observed for the film grown at the lower As flux [Fig. 2(b)]. This agrees with an observation showing that increasing the P overpressure smoothens the surface during the growth of InP,[15] but contrasts a result from the InGaP/InP system, which shows enhanced ripening of islands at high P overpressures.[16]

Two island morphologies are observed in Fig. 2. Large but short quasi-3D islands are typically present at t_{SK} [Fig. 2(a)], while taller 3D islands appear at thicknesses greater than t_{sk} [Fig. 2(b)]. Figure 3(a) shows a STM image of a typical quasi-3D island formed on the same surface shown in Fig. 2(a). The quasi-3D islands are 10–20 Å tall, 1000–1500 Å in length, and are elongated along [1$\bar{1}$0]. Their tops are planar, consisting of a single (001) facet or overlapping (001) terraces.[17] Many quasi-3D islands also have side facets from the {216} and {138} families,[18] which are close to the orientations observed for fully developed InAs[19] and $In_xGa_{1-x}As$[20] quantum dots. At the atomic scale, the tops of quasi-3D islands and the surface of the wetting layer are both comprised of $\alpha2(2\times4)$ and $(n\times3)/(4\times3)$ reconstructions, similar to planar surfaces at this composition.[13,14] The quasi-3D islands formed at t_{SK} are similar in morphology to the large and elongated islands which form during the annealing of planar subcritical films,[17,21] i.e., also in a regime where the driving force for 3D islanding is low.

The island shape evolves at film thicknesses further above t_{SK}. Figure 3(b) shows a STM image of typical 3D islands formed on the surface shown in Fig. 2(b), at approximately 3 ML above t_{SK}. Narrow islands having (001) tops and side facets near the {216} and {138} orientations are often observed. These islands usually form on the top of larger elongated platelets that are similar in size and height to the

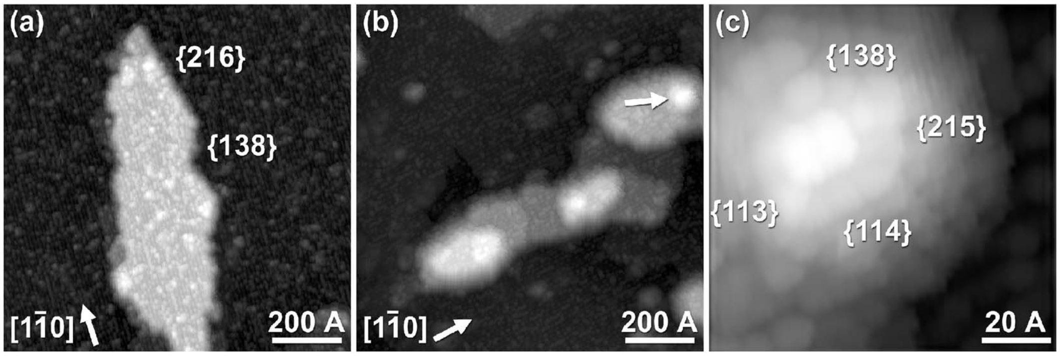

FIG. 3. Series of STM images of 25 ML $In_{0.27}Ga_{0.73}As/GaAs(001)$ films grown at $T=490$ °C using (a) $F_{As4}=2$ ML/s and (b) $F_{As4}=1.8$ ML/s. The small pyramidal island arrowed in (b) is shown at higher resolution in (c).

quasi-3D islands [Fig. 3(a)]. Often, small 3D islands develop on the top of the tallest features [Fig. 3(c) and arrowed in Fig. 3(b)], which have a pyramidal shape with facets near the {215}, {138}, and {113} orientations. These small islands resemble in shape and size, the quantum dots formed at higher In composition and strain,[19] and their formation is likely related to the In enrichment occurring at the island tops during growth.[22]

The transition between planar and 3D growth has been examined for intermediate misfit strain $In_{0.27}Ga_{0.73}As/GaAs(001)$ films. For a narrow range in growth temperature and misfit strain, the critical thickness is strongly dependent on the As flux. The increase in t_{SK} at high As flux is attributed to a lower adatom density and a shorter diffusion length, which slows down the island nucleation rate. This effect provides an additional and valuable growth parameter that can be used to manipulate the surface morphology of strained InGaAs layers, with applications in device growth.[23] The detailed morphology of 3D islands formed at moderate misfit strains has also been characterized by AFM and high resolution STM. Large quasi-3D islands form at, or just below, t_{SK}, and appear to act as a base for the formation of more discrete 3D islands above t_{SK}.

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