Review Article: Molecular Beam Epitaxy of Lattice-Matched InAlAs and InGaAs Layers on InP (111)A, (111)B, and (110)

Christopher D. Yerino  
Yale University

Baolai Liang  
University of California

Diana L. Huffaker  
University of California

Paul J. Simmonds  
Boise State University

Minjoo Larry Lee  
Yale University
REVIEW ARTICLE

Review Article: Molecular beam epitaxy of lattice-matched InAlAs and InGaAs layers on InP (111)A, (111)B, and (110)

Christopher D. Yerino
Department of Electrical Engineering, Yale University, New Haven, Connecticut 06520

Baolai Liang and Diana L. Huffaker
California NanoSystems Institute and Department of Electrical Engineering, University of California, Los Angeles, California 90095

Paul J. Simmonds a)
Department of Physics and Micron School of Materials Science and Engineering, Boise State University, Boise, Idaho 83725

Minjoo Larry Lee b)
Department of Electrical Engineering, Yale University, New Haven, Connecticut 06520 and Department of Electrical and Computer Engineering, University of Illinois at Urbana-Champaign, Urbana, Illinois 61801

(Received 18 June 2016; accepted 29 November 2016; published 21 December 2016)

For more than 50 years, research into III–V compound semiconductors has focused almost exclusively on materials grown on (001)-oriented substrates. In part, this is due to the relative ease with which III–Vs can be grown on (001) surfaces. However, in recent years, a number of key technologies have emerged that could be realized, or vastly improved, by the ability to also grow high-quality III–Vs on (111)- or (110)-oriented substrates. These applications include: next-generation field-effect transistors, novel quantum dots, entangled photon emitters, spintronics, topological insulators, and transition metal dichalcogenides. The first purpose of this paper is to present a comprehensive review of the literature concerning growth by molecular beam epitaxy (MBE) of III–Vs on (111) and (110) substrates. The second is to describe our recent experimental findings on the growth, morphology, electrical, and optical properties of layers grown on non-(001) InP wafers. Taking InP(111)A, InP(111)B, and InP(110) substrates in turn, the authors systematically discuss growth of both In_{0.52}Al_{0.48}As and In_{0.53}Ga_{0.47}As on these surfaces. For each material system, the authors identify the main challenges for growth, and the key growth parameter–property relationships, trends, and interdependencies. The authors conclude with a section summarizing the MBE conditions needed to optimize the structural, optical and electrical properties of GaAs, InAlAs and InGaAs grown with (111) and (110) orientations. In most cases, the MBE growth parameters the authors recommend will enable the reader to grow high-quality material on these increasingly important non-(001) surfaces, paving the way for exciting technological advances.

I. INTRODUCTION

A. Motivation

III–V semiconductor layers grown on (111) and (110) surfaces offer a number of distinctive properties compared to those grown on (001) wafers. Examples include a unique alignment of the internal piezoelectric field, access to different zones of the electronic band-structure, compatibility with novel electronic materials, and different barriers to dislocation nucleation. Despite an initial surge of interest in the 1980s and 1990s, current understanding of growth on these orientations remains much less mature in comparison to growth on (001) surfaces. However, novel device opportunities continue to emerge whose realization depends on the ability to grow high quality (111) and (110) materials. In light of this recent resurgence of interest in (111) and (110) materials, a comprehensive review of the growth of III–V semiconductors with these orientations is long overdue.

The emerging applications that follow critically depend on growth on either the (111) or (110) surface for their success.

1. III–V field-effect transistors

(111)-oriented III–Vs have been proposed as a promising channel material for the scaling of n-channel metal-oxide-
Electronic field-effect transistors (MOSFETs). III–Vs offer higher electron mobilities \( \mu_e \) than silicon for increased drive current. However, III–V conduction bands typically have a low density-of-states (DOS), which reduces the sheet carrier density and may hinder the drive current in what is termed the “DOS bottleneck.” A proposed solution is to create III–V MOSFETs on the (111) surface to access the L-valleys for electron transport. The L-valleys of III–V semiconductors offer both high DOS and high injection velocities. Through the appropriate application of quantum confinement, strain, and III–V materials with low L-T valley separation, conduction in the L-valleys can occur in parallel with the \( g \)-valley, resulting in large drive currents. In particular, thin layers of GaAs (111) and GaSb (111) are predicted to have superior drive currents compared to Si. One proposed device structure relies on the growth of a tensile strained GaAs quantum well on InP(111), a material system that has been successfully grown for novel (111) quantum dots (QDs). These techniques require a deep understanding of growth methods, droplet epitaxy, and recently tensile-strained quantum dots (QDs) grown under tensile strain. QDs have previously been grown on (111) surfaces act as ideal sources of entangled photons for quantum information. Pairs of polarization-entangled photons are emitted by the decay of biexcitons. However, useful entangled photons can only be prepared if the QDs have negligible excitonic fine-structure splitting (FSS), which is difficult to achieve in QDs on the (001) surface. Low FSS requires high symmetry of the QD electronic states, which can only be reliably produced when the piezoelectric field is normal to the QDs, i.e., on the (111) surface. Growth of (111) QDs is a significant challenge, which has been addressed using etch-pit-assisted growth and recently tensile-strained self-assembly (described below). All of these (111) QD growth methods result in very low FSS compared to (001) QDs. These techniques require a deep understanding of growth on (111) surfaces, both to grow high quality buffer layers as a template for QD growth, and to understand the complex process of self-assembly on (111) surfaces.

3. Tensile quantum dots

Our groups have recently shown that (110) and (111) surfaces are ideal candidates for a new class of quantum dot (QD) grown under tensile strain. QDs have previously been limited to compressively strained Stranski–Krastanov growth or unstrained droplet epitaxy. This is because QDs grown on (001) surfaces under tensile strain tend to readily nucleate dislocations. In addition, QD growth has focused largely on the (001) surface, since previous attempts at compressively strained growth on (111) and (110) surfaces also resulted in highly dislocated material without QD formation. Instead, we have shown that combining tensile strain and a (110) or (111) surface inhibits dislocation formation in QDs. This principle has enabled us to demonstrate high-quality tensile GaAs/InAlAs and GaP/GaAs nanostructures on both (110) and (111) surfaces. Since tensile strain can strongly reduce semiconductor bandgaps, tensile QDs on these surfaces are promising for optoelectronic devices operating at long wavelengths. Moreover, tensile GaAs/InAlAs QDs grown on (111) surfaces are perfectly suited to entangled photon generation due to their high electronic symmetry.

4. Spintronics

(110) materials are of current interest for spintronics, due to the long spin-lifetime observed in (110) quantum wells relative to those on (001). This property has enabled a number of advances in spintronics to be made using (110) quantum wells, including gate-control of relaxation time, low-voltage spin manipulation, spin-injected light emitting diodes, and spin-injected lasers. Spintronics applications, smooth interfaces are essential for maximizing spin-lifetimes but III–V materials grown on (111) surfaces are often rough. Improvements in both interface and material quality of (110) III–Vs will thus enhance the performance of spintronic devices. For example, the optimization of high-quality InP-based materials on the (110) surface, as described in this paper, could potentially enable the creation of spintronic devices operating at telecom wavelengths.

5. Topological insulators

(111)-oriented III–Vs have been identified as useful templates for the growth of \( V_2–VI_3 \) topological insulators (TIs), including Bi\(_2\)Se\(_3\), Bi\(_2\)Te\(_3\), and Sb\(_2\)Te\(_3\). The hexagonal crystal symmetry of the \( V_2–VI_3 \) TIs is better matched to the (111) plane of zinc-blende materials than to the (001) plane. High quality TIs have been grown on both GaAs(111) and InP(111) substrates. InP(111) has only 0.16% lattice mismatch with Bi\(_2\)Se\(_3\). Bi\(_2\)Se\(_3\) grown on InP(111) therefore has superior crystalline domain size and defect density to Bi\(_2\)Se\(_3\) grown on GaAs(111), and both substrates produce better Bi\(_2\)Se\(_3\) crystalline quality than growth on Si(111). Advances in (111) III–V growth, particularly on InP, are likely to further improve TI quality and allow the integration of TIs with III–V devices.

6. Transition metal dichalcogenides

Growth on (111) III–Vs also offers a promising route for the synthesis of transition metal dichalcogenides (TMDs). TMDs can be grown in 2D monolayer or multilayer forms, and offer a broad range of properties, including semiconductivity, metalliclicity, and superconductivity. Novel electronic and optoelectronic devices have been demonstrated based on TMDs, such as field-effect transistors, phototransistors, and tunable \( p–n \) junctions. The
development of TMD devices typically relies on exfoliation,\textsuperscript{51} which is challenging to scale to wafer-level synthesis. In contrast, the epitaxial growth of TMD layers on GaAs(111) was reported many years ago, due to the unique termination of (111) surfaces.\textsuperscript{54} TMD materials are composed of 2D layers weakly connected by van der Waals bonds. High-quality layers can hence be grown if the substrate only interacts to support the TMD via van der Waals forces.\textsuperscript{55} The GaAs(111) surface is ideal for this purpose. Exposure to group-VI atoms, including sulfur, selenium, and tellurium, passivates the specific arrangement of dangling bonds found on (111) surfaces.\textsuperscript{55} Renewed interest in TMDs has led to recent molecular beam epitaxy (MBE) growth demonstrations of wafer-scale monolayer MoSe\textsubscript{2} on GaAs(111)B (Ref. 56) as well as MoTe\textsubscript{2}/MoSe\textsubscript{2} superlattices on GaAs(111)B with monolayer precision.\textsuperscript{57} Smooth GaAs(111)B buffer layers help facilitate the subsequent TMD growth.\textsuperscript{56} By understanding the growth of (111) III–V materials, new opportunities will arise for the integration of III–V and TMD devices.

\textbf{B. Challenges in III–V growth on (111) and (110) substrates}

The above examples highlight the growing demand for high-quality III–V s on (111) or (110) substrates. However, ideal conditions established for growth of III–V s on (001) surfaces often produce poor material quality on (111) and (110) orientations. To date, growth on (111) and (110) surfaces has largely focused on GaAs and AlGaAs. Growth of GaAs/AlGaAs (111) and (110) is much more difficult than growth of GaAs/AlGaAs (001), often resulting in large surface roughness, and poor electrical and optical properties. Nevertheless, by carefully exploring growth conditions, smooth surfaces with favorable electrical and optical properties have been demonstrated. However, very little information is available for growing other III–V s with (111) or (110) orientations, in particular for the technologically useful materials InGaAs or InAlAs grown on InP. This lack of a growth–optimization toolbox means that the electrical, optical, and crystallographic quality that is achievable in (111) and (110) materials is currently insufficient to meet the demands of the novel applications described above.

The atomic force microscope (AFM) images in Figs. 1(a)–1(e) show that surface roughening is the primary challenge for growth of (111) and (110) epilayers. Buffers grown on (111)A, (111)B, and (110) surfaces tend to form a high density of unwanted features. Most prominent among these are “hillocks” [the large 3D islands in Figs. 1(a) and 1(e)],\textsuperscript{58–64} and “pits” that arise from step bunching, particularly on offcut substrates [Fig. 1(c)]. To avoid hillock formation, many groups have adopted the use of offcut substrates, which promote step flow growth rather than 3D islanding,\textsuperscript{58,65–68} but the optimization of growth conditions is seldom reported, and smooth growth remains challenging.\textsuperscript{69–71} Furthermore, much of the early work on (110) and (111) growth was performed prior to widespread availability of commercial AFMs and so morphology studies were often limited to optical microscopy. The effect of growth conditions on atomic-scale morphology therefore remains largely unknown for most materials on non-(001) surfaces.

The purpose of this paper is twofold: first, to provide a comprehensive review of the literature surrounding growth on (111) and (110) surfaces; and second to fill in the gaps in the literature with new experimental data. The experimental work we present here focuses on the effect of growth conditions on material properties for InAlAs and InGaAs grown on InP(111)A, (111)B, and (110) with an emphasis on achieving smooth surface morphology. Challenges for homoepitaxy on InP(111)A and (111)B are also discussed. We show that each substrate orientation requires a different growth strategy to optimize material properties, which typically differs greatly from what is used for growth on the (001) face.

The paper is divided into separate sections describing growth on exact InP(111)A (Sec. III), offcut InP(111)B (Sec. IV), and exact InP(110) (Sec. V). Each section begins

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{fig1.png}
\caption{(Color online) AFM images (15 × 15 \textmu m\textsuperscript{2}, 15 nm z-scale) showing surface morphology of InAlAs on exact InP(111)A, InAlAs on offcut InP(111)B, and InGaAs on exact InP(110). (a), (c), and (e): Growth under nonoptimized conditions typically leads to rough surface morphologies. (b), (d), and (f): Optimized growth conditions arrived at in this work produce significantly smoother surfaces.}
\end{figure}
with a review of epitaxy for GaAs and other III–V materials on each of these orientations, which informs new growth strategies for InAlAs and InGaAs on InP explored in this work. In Sec. VI we compare and summarize growth across the different surface orientations.

We show that by careful selection of the growth conditions, it is possible to eliminate, or greatly mitigate, morphological defects on each orientation, resulting in atomically smooth surfaces [Figs. 1(b)–1(f)]. We present experimental data showing how the electrical and optical properties of these materials depend on growth conditions. We use trends in surface morphology, electrical and optical properties to identify optimal parameters for the growth of these alloys, and discuss cases where jointly optimizing all of these material properties is challenging. Taken together, this work offers a comprehensive guide to overcoming obstacles to III–V material growth on (110) and (111)-oriented InP substrates.

II. EXPERIMENT

For the new results presented in this work, samples were grown in a VEECO Modular GEN II solid-source MBE, using P$_2$ and As$_4$ as the group-V sources. As$_2$ was selected instead of As$_4$, for consistency with the body of literature on (111) and (110) III–V growth; many growth studies in the literature predated the widespread availability of the valved arsenic cracker for producing As$_2$, and hence used the As$_4$ species. Our fluxes were estimated using a beam flux monitor (BFM) located at the growth position. We calculated V/III flux ratios directly from BFM readings without sensitivity corrections. Growth temperatures $T_g$ were measured by optical pyrometer for $T_g \geq 400$°C. For $T_g < 400$°C, we measured temperature using a thermocouple located behind the substrate. We estimate that our thermocouple values are 5–20° lower than the true values for $T_g < 400$°C.

Wafers used in this study were all semi-insulating and included the following orientations: on-axis InP(110), on-axis InP(111)A, and vicinal InP(111)B offcut 2° toward $\langle 211 \rangle$, (110) and (111)A substrates were supplied by AXT, and (111)B substrates were supplied by SWI. All substrate orientations were specified to within ±0.5° by the manufacturers. Substrates were also specified as epiready, and no wet chemical treatment was performed, unless otherwise noted. Cleaved pieces were mounted on 4 in. diameter silicon handles using high-purity molten indium as an adhesive.

The conditions used for growth on (111)A, (111)B, and (110) InP are detailed in the corresponding sections of this paper. InAlAs growths on different InP surface orientations were performed separately. InGaAs samples were grown at a fixed rate of 0.17 μm/h by coloading (110), (111)A, and (111)B InP wafers onto the same backing wafer and testing growth conditions on the three samples simultaneously. In situ desorption of the native oxide was performed under P$_2$ or As$_2$; our choice of deoxidation species was partially based on the availability of P$_2$ in our MBE system and does not necessarily reflect a preference for a particular species. Unless otherwise noted, comparisons in this study are only made between samples where the same oxide desorption procedure was used. Single layers of lattice-matched In$_{0.52}$Al$_{0.48}$As or In$_{0.53}$Ga$_{0.47}$As were directly deposited on the InP substrate to investigate material properties. We confirmed lattice-matching of alloy compositions to the InP with x-ray diffraction (XRD). Hereafter, the terms InAlAs and InGaAs refer to epilayers with alloy compositions lattice-matched to InP.

Surface morphology was characterized using Nomarski microscopy and AFM (Digital Instruments Nanoscope IIIa). Photoluminescence spectroscopy (PL) was performed with a 532 nm pump laser, an InGaAs detector for InGaAs samples, and a Si detector for InAlAs samples. Hall effect measurements were performed on InGaAs samples using the van der Pauw method to determine background electron density, $n_0$, and electron mobility, $\mu_e$. We used indium solder to create electrical contacts for Hall measurements. To ensure Ohmic behavior, we gently annealed the contacts with the soldering iron until the indium wet the surface. Hall measurement was not possible for most InAlAs samples, due to the rapid formation of insulating oxides in air and resulting high contact resistances. Transmission electron microscopy was performed in either cross-section (X-TEM) or plan-view (PV-TEM) using a FEI Osiris operating at 200 kV.

III. GROWTH ON InP (111)A

A. Review of growth on (111)A surfaces

Previous reports of III–V epitaxy on (111)A surfaces focused almost exclusively on GaAs, with very little work describing growth trends for other materials. As discussed above, the primary challenge for growth on (111)A is the formation of a high density of hillocks under a wide range of growth conditions. The hillocks are threefold symmetric and take the form of either a pyramid triplet with three mounds that are bounded by {010}-related facets, or a single triangular pyramid bounded by {110}-related facets.

Hillock formation is eliminated using a substrate offset by $\geq 5°$ toward (100), but it is also possible to grow smooth GaAs on on-axis (111)A substrates under certain growth conditions. Earlier reports showed decreasing hillock size and increasing hillock density with low $T_g$ and high V/III ratio, enabling hillock removal by shrinking them to zero size. In addition, Sato et al. found that the presence of hillocks was related to arsenic-limited growth regimes.

Their group showed from reflection high-energy electron diffraction (RHEED) intensity oscillations that the GaAs(111)A growth rate was arsenic-limited rather than gallium-limited at high $T_g$ and low V/III. This behavior was attributed to a low arsenic sticking coefficient for the (111)A surface. The arsenic-limited regime was shown to be closely correlated to hillock formation, suggesting that hillocks are related to excess Ga on the surface. During growth on (111)A substrates, researchers have observed surface defects arising from growth on both (100) and (110)-related facets. We hypothesize that if they offer higher arsenic sticking coefficients than (111)A, it is possible that preferential growth on these facets could lead to hillock...
formation. Substrate cleaning and control of ambient arsenic levels prior to growth have also been suggested as techniques to promote smooth epitaxy.\textsuperscript{73}

The surface morphology of GaAs(111)A has been surveyed extensively by Nomarski and scanning electron microscopy, but little has been published regarding its morphology at the atomic-level. According to RHEED, GaAs(111)A exhibits a (2 x 2) reconstruction before, during, and after growth, across virtually all $T_g$ values.\textsuperscript{72} The (2 x 2) reconstruction has been studied by scanning tunneling microscopy (STM), and arises from periodic gallium vacancies on the GaAs(111)A surface.\textsuperscript{75} These Ga-vacancies are expected to appear under the Ga-rich conditions common to GaAs(111)A growth, based on ab initio theoretical calculations.\textsuperscript{76} However, the effect of growth conditions on the atomic terrace morphologies of InP(111)A and its lattice-matched InGaAs and InAlAs buffers remain largely unexplored. Sections III B–III E address this gap in the literature.

B. InP(111)A homoepitaxy

We performed preliminary work on InP(111)A homoepitaxy, to determine if growing InP buffers on InP(111)A substrates before heteroepitaxy would lead to smoother surfaces. In fact, both InAlAs and InGaAs grown on bare InP(111)A substrates were smoother than when grown on InP(111)A homoepitaxial buffers (see Secs. III C and III D). Substrate deoxidation and InP growth were both performed using $P_2$, followed by InP growth at 0.26 $\mu$m/h. Similar to GaAs(111)A homoepitaxy, InP(111)A [as well as InGaAs(111)A and InAlAs(111)A discussed in Secs. III C–III E] exhibits a (2 x 2) streaky RHEED pattern during growth.\textsuperscript{72,75}

The effect of native oxide desorption temperature and $T_g$ on InP homoepitaxy is shown in Fig. 2. Both sample surfaces in Fig. 2 were covered with triangular faceted features that are gradually sloped on one side and vertically sloped on the two adjoining sides. The vertical slopes produced pronounced discontinuities across the growth surface, which appear as narrow trenches at the foot of the faceted structures. Lower $T_g$ and desorb temperatures caused the hillocks to increase in density and decrease in size. Under such conditions, the trench-features subsided and smoother regions appeared between the hillocks.

We attempted to use V/III ratio to lower the hillock density, but no effect was observed in the range V/III = 40–80. Substrates etched in $H_2SO_4:H_2O_2:H_2O$ followed by a bromine-methanol etch produced very similar growth morphologies to untreated epiready substrates (not shown), confirming that the hillocks are not induced by substrate preparation. As discussed in Secs. III C–III E for InAlAs and InGaAs growth on InP(111)A, very high and very low $T_g$ appear most promising for smooth growth. Such strategies may also yield smooth InP homoepitaxy, but further work is needed to demonstrate atomically smooth InP(111)A homoepitaxy on exact substrates.

C. InAlAs on InP(111)A

1. InAlAs(111)A: Surface morphology

In this section, we present new results on surface morphology optimization of InAlAs grown directly on exact InP(111)A substrates without an InP buffer. Pyramidial hillocks are the primary challenge for InAlAs and InGaAs growth on InP(111)A [Fig. 1(a)]. The reduction or elimination of hillocks for InAlAs and InGaAs on exact InP(111)A was found to be more difficult than growth on offcut InP(111)B (Sec. IV) or exact InP(110) (Sec. V). The triangular hillocks were typically ~500 nm wide and ~50 nm tall with densities ranging from the mid-$10^6$ cm$^{-2}$ to the mid-$10^7$ cm$^{-2}$. In addition, the smoother region between the hillocks consisted of atomic “wedding cakes,” where monolayers of concentric 2D islands appeared, rather than step flow growth. Such wedding cakes form when adatoms nucleate new 2D islands on top of existing ones, rather than attaching to the sides of other islands; similar wedding cake features form during MBE growth of certain other III–V materials including GaSb(001).\textsuperscript{77} We characterized the
roughness attributable to the wedding cakes by measuring rms roughness values in regions free of hillocks (unless otherwise noted). InP(111)A oxide desorption was performed under either As$_4$ or P$_2$, followed by growth of 100–200 nm of InAlAs.

First, we show the effect of As$_4$ versus P$_2$ overpressure during InP native oxide desorption (Fig. 3) where a beam equivalent pressure of $1.1 \times 10^{-3}$ Torr was used for both species. When InAlAs was grown under the same, near-optimal $T_g$ and V/III conditions, the P$_2$ desorb produced $\sim$40% fewer hillocks on the surface than the As$_4$ desorb. Surface roughness between the hillocks also improved by switching from As$_4$ to P$_2$ (Fig. 3). The AFM images show that using P$_2$ during the oxide desorption produces a triangular step-flow morphology, which better reflects the (111) crystal symmetry than the almost-circular wedding cakes observed when As$_4$ is used. Desorbing the native InP oxide under arsenic produces monolayers of compressive InAs$_{1-x}$P$_x$ on the surface, due to anion exchange, which may explain the changes in morphology observed here. P$_2$ is thus recommended as the ideal species under which to desorb the InP native oxide.

As Figs. 4(c) and 4(e) show, growth rate had a pronounced effect on InAlAs(111)A morphology. Increasing the growth rate from 0.17 to 0.5 $\mu$m/h raised the hillock density by $>3 \times$. Adatom diffusion length is reduced at higher atomic flux, increasing the rate of island nucleation. The lower adatom diffusion time at high growth rate allows 3D islands to form closer together, leading to a higher density of hillocks.

In addition, comparison of Figs. 4(d) and 4(f) shows that a slower growth rate lowered wedding cake height, reducing rms roughness from 0.92 to 0.66 nm over a 5 $\times$ 5 $\mu$m$^2$ area. Since island nucleation rates increase with adatom arrival rate, the use of higher atomic fluxes can encourage the nucleation of new 2D layers atop existing ones to promote taller wedding cakes. Due to the improved atomic morphology and lower hillock density at lower growth rate, all subsequent InAlAs(111)A growths were performed at 0.17 $\mu$m/h.

Comparison of Figs. 4(a) and 4(c) also shows the effect of V/III ratio on the InAlAs(111)A morphology. Nomarski microscopy shows that increasing the V/III ratio from 77 to 230 reduced the hillock density by 40%. This similarity with GaAs(111)A growth suggests that the hillocks on InAlAs(111)A are related to limited arsenic-availability caused by a reduced arsenic sticking coefficient.

The effect of reducing $T_g$ on hillock density is shown in Fig. 5. Hillock density increased from $5.6 \times 10^6$ cm$^{-2}$ at 480°C to $8.0 \times 10^6$ cm$^{-2}$ at 437°C, while hillock size decreased rapidly, such that the volume of material in the...

---

**Fig. 3.** (Color online) Effect of oxide desorption overpressure species on subsequent InAlAs(111)A morphology. (a) and (b): Substrates deoxidized in P$_2$. (c) and (d): Substrates deoxidized in As$_4$. Nomarski images, annotated with hillock densities. (b) and (d): 1 $\times$ 1 $\mu$m$^2$ AFM images (3 nm z-scale). Rms roughness values averaged over 25 $\mu$m$^2$ between hillocks.

**Fig. 4.** (Color online) Comparison of growth rate and V/III ratio effects on InAlAs(111)A morphology for $T_g = 500^\circ$C (substrates deoxidized in As$_4$). Images in same row are taken from same sample. (a), (c), and (e): 50 $\times$ 50 $\mu$m$^2$ Nomarski images, annotated with hillock densities. (b), (d), and (f): 2 $\times$ 2 $\mu$m$^2$ AFM images (3 nm z-scale), annotated with rms roughness. Rms roughness values averaged over 25 $\mu$m$^2$ between hillocks.
hillocks also decreases. At 405°C, the hillocks were eliminated almost entirely. While the removal of hillocks encourages the use of low \( T_g \), the atomic scale roughness shows the opposite trend. As \( T_g \) decreased, the terrace widths on the wedding cakes shrank from 49 to 12 nm on average, and the wedding cakes grew in height; rms roughness increased concomitantly from 0.33 to 0.92 nm. The relationship between \( T_g \) and wedding cake size indicates that the nucleation rate of 2D islands is lower at higher \( T_g \) due to faster detachment of adatoms from island pronuclei.\(^{29,80–82}\)

New surface defects appeared in the InAlAs grown at 405°C, including pits, hillock clusters, and irregularly shaped trenches. Close inspection of Fig. 5(f) reveals hillocks much smaller than previously observed, which form in clusters of three, arranged with threefold symmetry. The symmetric arrangement of these hillocks resembles the larger, threefold symmetric hillocks more commonly observed at high \( T_g \), suggesting that they have the same origin. The total density of threefold symmetric defects, including larger hillocks and small hillock-clusters, was \( 1.0 \times 10^7 \) cm\(^{-2} \) at 480°C, \( 3.8 \times 10^7 \) cm\(^{-2} \) at 437°C, and \( 2.3 \times 10^8 \) cm\(^{-2} \) at 405°C. The higher density of these features with decreasing \( T_g \) shows that formation of the surface defects is regulated by adatom diffusion length. We attribute this effect to indium adatoms, given previous observations of enormous migration lengths up to \( \sim 25 \) \( \mu \)m on III–V surfaces.\(^{23} \) This in turn allows the hillocks to form closer together as they compete for adatoms. Note that \( V/III = 230 \) is used for all samples in Fig. 5. The increasing distance between hillocks at higher temperature indicates that the diffusion length is still on the order of microns, even under high As-flux.

X-TEM and PV-TEM of InAlAs/InP(111)A show very few extended defects in the optimized InAlAs(111)A buffers. Figure 6(a) shows TEM images from the smoothest InAlAs(111)A samples obtained in this study [i.e., Fig. 5(a)]. Defects are only noticeable in the hillocks [Fig. 6(b)], which contain a high density of planar defects and dislocations. Thus, the hillock defects are likely to be electrically and optically detrimental: reduction in hillock density is expected to improve the overall material quality. The sample in Figs. 6(a) and 6(b) also includes a layer of InGaAs between the InAlAs and InP, the growth of which is discussed in Sec. III E. We conclude that apart from the hillocks, both InAlAs and InGaAs grow with minimal defect nucleation.

Finally, we investigated the effect of pregrowth InP substrate etching, to determine whether the hillocks are influenced by substrate preparation, as suggested by Ref. 73. An untreated epiready InP(111)A wafer was coloaded with InP pieces etched in H\(_2\)SO\(_4\):H\(_2\)O\(_2\):H\(_2\)O (5:1:1) followed by bromine: methanol (1:80). The wet chemical treatment reduced the hillock density from \( 9.4 \times 10^6 \) to \( 5.0 \times 10^6 \) cm\(^{-2} \),
implying that surface preparation is partially responsible for hillocks.\textsuperscript{73}

2. \textit{InAlAs(111)A: Photoluminescence}

PL from the \textit{InAlAs(111)A} is influenced by \(T_g\) (Fig. 7) but is relatively insensitive to other variables. The PL spectra in Fig. 7 show the InP substrate emission at 1.34 eV, next to the \textit{InAlAs} emission at 1.45 eV. As \(T_g\) was increased from 405 to 480 °C, the entire spectrum increased in intensity by 35%. The shape of the two spectra are identical, suggesting that the higher PL intensity may partially result from stronger light extraction in the 480 °C sample, due to the formation of larger hillocks. PL is largely insensitive to growth rate and V/III ratio, with nearly identical PL intensities observed across the parameters used in this study (not shown).

3. \textit{InAlAs(111)A: Summary}

In summary, the hillocks prevalent during growth of \textit{InAlAs(111)A} can be significantly mitigated using high \(T_g\), very high V/III ratio, and low growth rate, but, unlike the case of \textit{GaAs(111)A},\textsuperscript{59} it was not possible to completely eliminate them. Low \(T_g\) removed the large hillocks but only at the expense of smaller-scale surface defects and higher atomic roughness due to wedding cakes. TEM confirms that hillocks contain a high density of extended defects, so further work is needed to eliminate them. The photoluminescence of \textit{InAlAs(111)A} was relatively insensitive to growth conditions, with marginal change in the intensity attributed to light extraction due to surface roughening. We favor the use of high \(T_g\) since this produces low hillock densities with smoother surfaces in-between. Provided the hillocks are low enough in density, there will be sufficiently large areas of smooth, high-quality, defect-free \textit{InAlAs} in between them to suit many applications.

Overall, our recommended growth conditions for \textit{InAlAs(111)A} are high \(T_g\) (≥480 °C), very high V/III ratio (∼230), and low growth rate (0.17 µm/h).

D. \textit{InGaAs on InP(111)A}

1. \textit{InGaAs(111)A: Surface morphology}

As shown in Fig. 6, hillocks are the primary source of extended defects in both \textit{InGaAs} and \textit{InAlAs}, so it is highly desirable to minimize their density. In terms of controlling hillock formation and wedding cake roughness, \textit{InGaAs(111)A} obeyed broadly similar growth trends to \textit{InAlAs(111)A}. We grew 200 nm \textit{InGaAs} layers on \textit{InP(111)A} following the oxide desorb at 485 °C under 1.1 × 10⁻⁵ Torr As\textsubscript{4}. Based on the \textit{InAlAs(111)A} growth experiments in Sec. III C, we adopted a growth rate of 0.17 µm/h for \textit{InGaAs(111)A}. However, we found \textit{InGaAs(111)A} to be much less sensitive to growth rate than \textit{InAlAs} (not shown), and we believe the following results are equally applicable at higher growth rates.

The effect of V/III ratio on \textit{InGaAs(111)A} morphology is shown in Fig. 8 for samples grown at \(T_g = 485 \text{ °C}\). The \textit{InGaAs(111)A} surface is roughened by large hillocks and very tall wedding cakes when a moderate V/III ratio of 25 was used [Figs. 8(e) and 8(f)]. A V/III ratio of ∼75 or higher
was needed to reduce hillock size [Figs. 8(a) and 8(c)] and planarize the InGaAs surface in between [Figs. 8(b) and 8(d)]. Similar to InAlAs(111)A, the InGaAs(111)A morphology at higher V/III ratio was predominantly flat with triangular hillocks scattered about the surface. However, for InGaAs growth, the hillock density was several times lower than for InAlAs growth, typically in the low to mid-10⁶ cm⁻². On some samples, clusters of hillocks were observed in addition to isolated hillocks; we report hillock densities as the number of contiguous hillock clusters per unit area.

Increasing the V/III ratio from 75 to 195 doubled the hillock density, which is opposite to the trend for InAlAs(111)A and GaAs(111)A.⁵⁸,⁵⁹ For InGaAs(111)A, the arsenic-limited model²⁹ used to explain GaAs(111)A hillocks no longer appears to be applicable. Instead, the trend for InGaAs(111)A suggests that hillock density scales with adatom diffusion length within this V/III range. The higher V/III ratio lowers the diffusion length, allowing hillocks to form closer together while competing for adatoms.

As we increased V/III ratio, atomic wedding cakes decreased in height and increased in atomic step spacing (i.e., terrace width). This effect was more pronounced than the effect of V/III ratio on InAlAs(111)A growth. V/III ratio optimization of InGaAs(111)A differs from InAlAs(111)A in that a tradeoff must be made between atomic-scale wedding cakes and larger-scale hillocks. For minimum hillock density, V/III ~75 is recommended. However, for applications where access to atomically flat surfaces is more important (e.g., QD studies), a high V/III ~195 should be used to increase the terrace widths.¹⁰

The effect of $T_g$ on InGaAs(111)A morphology from 350 to 485 °C is shown in Fig. 9 at a fixed V/III of 75. As seen in Fig. 5 for InAlAs(111)A and Fig. 2 for InP(111)A, reducing $T_g$ decreased the size and increased the density of hillocks. The hillock density increased by 3 orders of magnitude as $T_g$ was reduced from 485 to 350 °C, confirming that hillock density scales inversely with adatom diffusion length for InGaAs(111)A. However, in contrast to InAlAs(111)A growth, low $T_g$ could not be used to completely eliminate hillock formation. The higher adatom mobility of Ga compared to Al may make it easier for Ga adatoms to reach hillock incorporation sites, even at low $T_g$.

Atomic scale wedding cake formation in InGaAs(111)A was suppressed as $T_g$ increased, lowering the rms roughness (Fig. 9). This reduction is consistent with our observations for InAlAs(111)A in Sec. III C and suggests that a reduction in the 2D nucleation rate promotes smooth growth in both cases. Increasing both $T_g$ to 510 °C and V/III ratio to 195 yielded terraces as wide as 100 nm, with hillock densities as low as $2 \times 10^6$ cm⁻² (Fig. 12). Increasing $T_g$ further did not yield additional surface smoothing, and the size and density of hillocks became larger for $T_g$ near the InGaAs decomposition temperature of ~550 °C.

2. InGaAs(111)A: Electrical properties

All InGaAs(111)A samples were unintentionally $n$-type, as commonly observed for InGaAs/InP(001).⁸⁴ The electrical properties of the InGaAs samples at 300 K were strongly dependent on V/III ratio [Figs. 10(a) and 10(b)]. For growths performed at $T_g = 485$ °C, V/III was optimized at ~75 to reach the lowest $n_0$ of $3.8 \times 10^{14}$ cm⁻³ and the highest $\mu_n$ of 5300 cm²/V s. $n_0$ increased and $\mu_n$ decreased substantially if V/III was increased to 195 or decreased to 25. In addition to higher background electron density, the sample grown under low V/III conditions also contained larger hillocks (see Fig. 8). Strong defect scattering due to the high hillock density likely contributes to the reduction in $\mu_n$ at low V/III. For the samples grown with high V/III, we speculate that arsenic-related defects may play a role in the higher $n_0$. This issue is also encountered in the case of InGaAs(110), which is grown at low $T_g$ and high V/III (Sec. V D).

$T_g$ also controls the electronic properties of InGaAs(111)A, albeit to a slightly lesser extent than V/III ratio [Figs. 10(c) and 10(d)]. For samples grown at V/III = 75, increasing $T_g$ from 420 to 485 °C increased $\mu_n$ from 3900 to 5300 cm²/V s at 300 K; we note that the dependence on $T_g$ observed at V/III = 75 is similar to that at V/III = 225. The value of $n_0$ is
almost independent of \( T_g \), and so the concurrent improvement in \( n_0 \) at high \( T_g \) cannot be attributed to a reduction in ionized impurities. Instead, the defects associated with hillocks likely dominate carrier transport since hillock density is strongly temperature sensitive. \( n_0 \) of the InGaAs(111)A samples is much lower than InGaAs(001) for which \( n_0 \approx 11500 \text{ cm}^2/\text{V} \text{s} \) in this electron density range,\(^8\) indicating significant room for improvement.

3. InGaAs(111)A: Photoluminescence

A monotonic improvement in PL intensity and FWHM linewidth was observed with increasing \( T_g \) for a given V/III ratio [Figs. 11(a) and 11(b)], likely due to the lower density of defective hillocks at high \( T_g \). Figure 11(c) shows that the InGaAs PL intensity increased and linewidth decreased with higher V/III ratio; the improvement in PL from V/III \( = 25–75 \) is correlated with a large improvement in surface morphology. However, hillock density increased at V/III \( = 195 \), which does not explain the accompanying increase in PL intensity. Instead, a 10\( \times \) increase in \( n_0 \) occurred at this V/III ratio [Fig. 10(a)], which would increase the radiative recombination rate. While V/III \( = 195 \) yielded the best PL, the highest Hall \( \mu_n \) values were found for V/III \( = 75 \). This further suggests that the improvement of PL intensity at higher V/III may not be related to crystalline quality.

4. InGaAs(111)A: Summary

In summary, the optimization of InGaAs on InP(111)A requires high \( T_g \geq 485 \text{ °C} \) and high V/III ratio to minimize surface roughness on the atomic scale and to alleviate hillocks. While hillock density is minimized at V/III \( \sim 75 \), roughness from atomic-scale features is minimized at V/III \( \sim 225 \). Optimization of electrical and optical properties largely overlaps with the conditions that minimize hillock density, allowing these properties to be jointly optimized.

Overall, our recommended growth conditions for InGaAs(111)A are: high \( T_g \) (\( \geq 485 \text{ °C} \)) and V/III ratio \( \sim 75–225 \).

E. InAlAs on InGaAs/InP(111)A

In Secs. III C–III D, we showed that InAlAs and InGaAs(111)A exhibit similar growth trends, but that InGaAs(111)A exhibited a smoother surface both in terms of hillock density and wedding cake roughness. For applications that require smooth InAlAs(111)A, we demonstrate that InAlAs grown on top of an InGaAs(111)A buffer largely inherits the atomic-level smoothness of the InGaAs. After desorbing the native oxide under As\( _4 \), we grew 100 nm InAlAs on 50 nm InGaAs. InAlAs and InGaAs were grown at 510 \text{ °C} using V/III ratios of 230 and 195, respectively. InAlAs grown on an InGaAs(111)A buffer exhibits a tenfold reduction in hillock density compared to InAlAs grown directly on InP(111)A [2.1 \times 10^6 \text{ cm}^{-2} and 1.5 \times 10^7 \text{ cm}^{-2} respectively, Figs. 12(a) and 12(c)]. However, the InAlAs/InGaAs hillock density was still higher than that of the bare InGaAs layer [8.8 \times 10^5 \text{ cm}^{-2}, Fig. 12(b)]. Under these growth conditions, both InGaAs(111)A and InAlAs(111)A have similar rms roughness away from the hillocks. Also, the average
terrace width for InAlAs grown on InGaAs was 95 nm, very close to the 100 nm measured on the optimal InGaAs samples, and double the terrace width for optimal InAlAs grown directly on InP(111)A. Despite the tendency for InAlAs to form a high density of hillocks and tall wedding cakes, the step-edges and hillocks of the InGaAs provide favorable incorporation sites for adatoms during InAlAs growth. We have already demonstrated the effectiveness of this approach by growing high-quality InAlAs barriers for tensile strained GaAs(111)A quantum dots.10

IV. GROWTH ON InP (111)B

A. Review of growth on (111)B surfaces

Previous III–V epitaxy work on (111)B surfaces has focused primarily on GaAs(111)B, although good progress has also been made with InP(111)B homoepitaxy. The primary growth challenge for GaAs(111)B exact substrates is the formation of wide triangular hillocks, which were found to be nearly unavoidable.60,85–87 These wide, short hillocks tend to cover the surface, in contrast to the taller, narrower hillocks for growth on (111)A surfaces (see Sec. III), degrading both the optical and electrical quality.88 For GaAs(111)B, surface morphology was correlated with RHEED observations, where the low temperature (2 × 2) reconstruction was always associated with hillock formation. It is possible to obtain smooth GaAs(111)B when growth is performed at higher temperature where the ( √19 × √19) surface reconstruction is stable, but only a very narrow growth window exists.86 GaAs(111)B growth on the (1 × 1) reconstruction (stable at high Tg and low V/III ratio) is also known to result in smooth surfaces.69 However, growth of smooth indium-containing materials such as InGaAs(111)B, under these same reconstructions, is difficult due to the high desorption rate of indium above Tg = 530 °C.85,86

Due to these challenges, the use of offcut substrates became the primary solution to achieve smooth GaAs(111)B growth.66,67,85 Yang et al. identified that the hillocks formed on GaAs(111)B were composed of three vicinal (111) “facets” inclined ~2° from the singular plane.85 They proposed that these vicinal facets were more energetically stable than the singular (111) plane and thereby demonstrated that tilting the surface toward these planes by 1°–3° in the <111> directions promoted smooth growth. Offcut substrates consequently resolved the problem of narrow growth windows. Growth in the ( √19 × √19) and (1 × 1) regimes both resulted in smooth GaAs(111)B, even at low Tg, where In0.3Ga0.7As/GaAs(111)B (e.g., x < 0.15) could be grown with a unity sticking coefficient for indium.69,85

While resolving the hillock problem, growth of GaAs(111)B on offcut substrates is still susceptible to roughening by step-bunching on the atomic scale. Under many conditions, growth on offcut surfaces forms large atomic steps typically several nanometers tall, referred to as “macrosteps.”69–71 These macrosteps are composed of parallel alternations between exact (111) facets and vicinal (111) surfaces.69,70 Proposed reasons for the step bunching include an energetically stable vicinal (111) face as well as arsenic desorption, which is a factor during high temperature growth.69 high V/III ratios and moderate growth temperatures can prevent step bunching.69,71

Smooth homoepitaxy of InP(111)B also requires a substrate offcut. A study of growth by metalorganic vapor phase epitaxy showed that faceted InP morphologies were
unavoidable on exact substrates and that smoother surfaces were possible using an offcut toward $(\overline{1}0\bar{1})$ than an offcut toward $(0\bar{1}1)$. Growth optimization of InP was also reported by Hou and Tu for gas-source MBE for a $(0.5\overline{1}1)$ offcut toward $(\overline{1}0\bar{1})$; note that this group and some others refer to the $(\overline{1}0\bar{1})$ offcut direction as the nearest $(0\overline{1}1)$, which are equivalent tilt directions. They found that high $T_g$ just below the InP decomposition temperature and low $V/III$ produced featureless surfaces by Nomarski, while elongated surface defects emerged if higher $V/III$ ratios were used.

Very few reports discuss InAlAs and InGaAs growth on InP(111)B, though smooth surfaces have been obtained. A $1^\circ$ offcut toward $(2\bar{1}1)$ was found to produce featureless surfaces by Nomarski with favorable bulk and two-dimensional electron gas mobilities, while a faceted surface was observed for growth on exact substrates. However, neither atomic-level surface characterization nor systematic study of growth conditions on electrical or optical properties for InAlAs and InGaAs have been reported. Similar to GaAs(111)B, we have found that pronounced step bunching occurs under a wide range of conditions, further necessitating careful growth optimization.

B. InP(111)B homoepitaxy

1. InP(111)B: Surface morphology

For our experiments, we begin with InP(111)B homoepitaxy, which, unlike the (111)A surfaces, provided a useful initial buffer for subsequent ternary growth. Based on the review of (111)B growth in Sec. IV A, we exclusively used InP(111)B substrates with a $2^\circ$ offcut toward $(\overline{2}11)$ to prevent faceting for all work in Sec. IV.

The InP(111)B deoxidation temperature must be carefully controlled to prevent substrate decomposition. We used $5 \times 10^{-6}$ Torr of $P_2$ overpressure to desorb the native oxide prior to InP growth at 0.26 $\mu$m/h. At a high deoxidation temperature of 515 $^\circ$C, indium droplets appeared in Nomarski due to InP decomposition [Fig. 13(a)]; this decomposition was not evident using in situ RHEED monitoring, which showed a bright, streaky $(1\overline{1}1)$ pattern throughout. Similar defects were noted by Hou and Tu resulting from decomposition at high temperature. Lowering the deoxidation temperature to 495 $^\circ$C prevented droplet formation [Fig. 13(b)]. Further optimization of InP growth was not required beyond the conditions reported by Hou and Tu, and we demonstrated smooth InP(111)B growth at 475 $^\circ$C with a low $V/III$ of 6.4. The AFM image in Fig. 13(c) shows that after removing the native oxide at 495 $^\circ$C, these growth parameters result in a very smooth surface with rms roughness of 0.23 nm across a $25 \times 25$ $\mu$m$^2$ area. The striated appearance of the AFM image resulted from closely spaced atomic steps due to the substrate offcut, and no step-bunching was observed.

Despite the absence of lattice mismatch in InP homoepitaxy, X-TEM of InP(111)B under optimum growth conditions showed a high density of dislocations at the initial growth interface [Fig. 14]; a layer of InAlAs was grown on top of the InP buffer for study in Sec. IV C. We note that InP(111)B wafers are not widely available, so the dislocations may affect the quality of the manufacturer’s substrate surface preparation. Cleaning and etching of the substrate is
recommended to improve the microstructural quality for future work on InP(111)B. In addition, fine-tuning of the deoxidation procedure may further reduce formation of extended defects.

2. InP(111)B: Electrical properties

We performed Hall measurements on the InP(111)B buffers to assess their electrical properties, finding all samples to be unintentionally $n$-type. A sample grown under higher $T_g$ and deoxidation temperatures of 515°C resulted in resistive for measurement. These conditions exacerbate InP decomposition, producing poor electrical properties. Lowering $T_g$ reduced the resistivity significantly, even with decomposition still present from the 515°C deoxidation. Growth at 490°C and $V/III = 3.2$ showed $n_0$ of $9.8 \times 10^{14}$ cm$^{-3}$ and $\mu_n$ of 1650 cm$^2$/V s. Lowering $T_g$ to 475°C and increasing the $V/III$ to 6.4 lowered $n_0$ to $5.3 \times 10^{14}$ cm$^{-3}$ and increased $\mu_n$ to 2500 cm$^2$/V s, consistent with better material quality in a weaker decomposition regime. However, lowering the desorption temperature from 515 to 495°C impaired the electrical properties, producing a high $n_0$ of $1.9 \times 10^{17}$ cm$^{-3}$ and a low $\mu_n$ of 850 cm$^2$/V s, despite the prevention of surface decomposition. In contrast, InP(001) has a $\mu_n = 4000$ cm$^2$/V s for $n_0 = 1 \times 10^{15}$ cm$^{-3}$, suggesting that further work is needed to lower the dislocation density and reduce scattering in the InP(111)B epilayers.

A possible explanation is that the low deoxidation temperature does not fully remove the surface oxide and contaminants from the as-received wafer. Researchers have shown that the temperature window within which the native oxide can be successfully removed from InP(001) spans less than 10°C. Within this narrow range, the position at which the Fermi level is pinned is extremely sensitive to deoxidation temperature. Fine-tuning of the InP deoxidation temperature and anneal time is therefore critical to optimize electrical properties while avoiding substrate decomposition.

3. InP(111)B: Photoluminescence

PL results from the InP(111)B buffer layers, measured at 300 K, are shown in Fig. 15. PL spectra for the bare, semi-insulating InP(111)B substrates are also provided as a baseline for intensity comparison, and in general, the homoepitaxial layers are expected to have higher integrated intensity due to the unintentional $n$-type doping. $T_g$ has a pronounced effect on the PL intensity, such that the PL intensity reduces by a factor of 8 as $T_g$ was lowered from 515 to 475°C [Fig. 15(a)]. All samples in Fig. 15 showed some degree of InP decomposition except for the 495°C sample in Fig. 15(c). The stronger decomposition at the highest $T_g$ likely increases...
the PL intensity due to higher $n_0$ and increased light extraction due to surface roughening. Figure 15(b) shows that PL intensity increased significantly at higher V/III ratio. Finally, the effect of lower deoxidation temperature is shown in Fig. 15(c). The 495 °C deoxidation condition is the only sample without decomposition. However, the much stronger PL intensity appears to be largely correlated to the much higher $n_0$ observed during Hall measurement. Growth conditions had little effect on the emission linewidth. In all cases, the
epitaxial InP films produced higher PL intensity and slightly narrower linewidth than the InP(111)B substrate.

4. InP(111)B: Summary

We have produced smooth InP growth on offcut InP(111)B substrates using a Tg of 475°C and a low V/III of 6.4. A low deoxidation temperature of 495°C is important to prevent substrate decomposition. Low n0 and reasonable μn values can be obtained near these conditions. However, further adjustment of the deoxidation temperature as well as improved substrate preparation is needed to remove defects indicated by Hall measurement and TEM.

C. InAlAs on InP(111)B

1. InAlAs(111)B: Surface morphology

Here, we present our optimization of MBE conditions for the growth of InAlAs on InP(111)B to achieve low surface roughness, and good optical and electrical properties. Since growth is performed on offcut substrates, the primary obstacle to smooth growth is step bunching rather than hillock formation, as discussed in Sec. IV A.

We removed the InP native oxide at 495°C before growing InAlAs on the InP(111)B surface. For some samples, a 100 nm InP buffer was grown prior to the InAlAs, but the presence of the InP buffer was found to have no effect on InAlAs growth trends. In either case, the InP is initially capped with 5 nm InAlAs grown at 480°C to protect the InP from decomposition during higher temperature growth. Without this 5 nm InAlAs cap, growth of InAlAs above 480°C resulted in rough surfaces. Next, 200–500 nm of InAlAs was deposited at 0.5 μm/h to study the effect of Tg and V/III ratio on surface morphology. Across all growth conditions studied, RHEED monitoring showed a (1 × 1) streaky pattern for both InP and InAlAs surfaces, similar to GaAs(111)B under certain growth conditions.95,96

Almost all InAlAs(111)B samples grown in this study appeared smooth when viewed with Nomarski (not shown). However, at the atomic scale it is clear that Tg strongly influences surface morphology (Fig. 16). Growth at Tg = 540°C yielded a smooth surface with a prevalence of monolayer-high steps [Fig. 16(a) inset], implying a step-flow growth mode. Reducing Tg to 490°C resulted in the appearance of tall step bunches on the surface [Fig. 16(b) inset]. Pits formed where many step edges bunched together at a particular location (i.e., step pinning), and the depth of the pits decreased with increasing Tg.

It is not possible to investigate whether InAlAs(111)B surfaces improve further at even higher Tg. Five hundred and forty degree Celsius is the hottest we can grow smooth InAlAs before thermal decomposition occurs and indium droplets appear. In addition, due to the high desorption rate of indium at 540°C, the InAlAs composition is sensitive to small temperature variations during growth. Thus the InxAl1−xAs composition in this study was found to vary among samples from x = 0.514–0.532, as measured by x-ray diffraction and PL.

### Fig. 18. (Color online) PL at 300 K for InAlAs. (a) Comparison between InAlAs grown on InP(001) and InP(111)B. (111)B sample contains a buried 3 ML GaAs layer for tensile nanostructure study, causing broad emission at 1.26 eV, as discussed elsewhere (Ref. 32). (b) Effect of V/III ratio on InAlAs(111)B PL. (c) Effect of Tg on InAlAs(111)B PL. InP emission at 1.34 eV is also present in (b), since a 100 nm InP buffer was grown beneath the InAlAs in this case.

InAlAs(111)B surface morphology is also very sensitive to V/III ratio. Figure 17 shows AFM images of InAlAs grown at Tg = 540°C with varying V/III. A clear optimum in the InAlAs roughness lies at V/III = 10–12, corresponding to rms roughnesses of 0.24–0.44 nm. At a higher V/III of 14,
step bunches became larger and deeper, while at a lower V/III of 6, small facets and pits covered the surface and the RHEED became faint.

As a result of this buffer optimization, we were able to obtain surfaces with atomic-scale smoothness at 540°C and V/III ~ 10 [Fig. 17(e)]. Small indentations in the sample surface were still visible by AFM due to a slight tendency toward step-bunching, but these features contribute very little to the overall roughness. The 1 x 1 μm² inset to Fig. 16(a) (grown at 540°C, V/III = 12) shows closely spaced, parallel monolayer steps running perpendicular to the [211] offcut direction (i.e., parallel to [011]). These monolayer steps are spaced 11 nm apart, consistent with the 2° substrate offcut.

TEM of the optimized InAlAs(111)B buffers shows that the lattice-matched InAlAs did not produce new defects when grown atop an InP epilayer (Fig. 14). When InAlAs was grown directly on the InP substrate (not shown), a similar density of dislocations was observed as that of the InP/InP(111)B homoepitaxial interface in Fig. 14, supporting our hypothesis that the dislocations originate from nonideal substrate preparation, rather than the growth conditions.

2. InAlAs(111)B: Electrical properties

We performed Hall measurements to compare InAlAs(111)B samples grown at 490 and 540°C. The InAlAs(111)B surface was found to produce Ohmic contacts after etching the native oxide, whereas contacts to InAlAs on (001), (110), and (111)A surfaces could not be obtained, due to rapid oxidation. Of the four orientations, only the InAlAs(111)B surface is aluminum-free. We speculate that the arsenic-terminated InAlAs(111)B surface hinders oxidation of the buried aluminum, enabling Ohmic contact to these samples. In addition to smoother surface morphology, higher $T_g$ was found to improve the electrical properties. The InAlAs(111)B samples had $\mu_n$ values ($n_0$ values) of 1100 cm²/Vs (1.3 x 10¹⁶ cm⁻³) and 3600 cm²/Vs (4.0 x 10¹⁷ cm⁻³) for $T_g$ of 490 and 540°C, respectively. Unexpectedly, the sample with $n_0 = 3600$ cm⁻³/Vs suggests that InAlAs(111)B could have higher $\mu_n$ than InAlAs(001); for InAlAs(001), Ref. 98 reports $\mu_n = 1350$ cm²/Vs for $n_0 = 3.7 \times 10^{15}$ cm⁻³.

3. InAlAs(111)B: Photoluminescence

Optimized InAlAs(111)B exhibited strong 300 K PL, with ~40% the PL intensity of InAlAs(001) grown in the same MBE system [Fig. 18(a)]. The weaker PL of InAlAs(111)B compared to (001) is likely due to the high density of extended defects that form at the initial growth interface, which we attribute to the substrate preparation.

The PL intensity and linewidth of InAlAs(111)B was relatively insensitive to the growth conditions investigated [Figs. 18(b) and 18(c)]. Even though increasing the V/III ratio from 12 to 14 significantly roughened the surface, the PL intensity decreased only slightly. Lower $T_g$, which was observed to produce large step bunches, also did not deteriorate the PL intensity. However, PL linewidth did correlate with surface morphology, with broader linewidths measured for the rougher samples grown with higher V/III of ~14 and lower $T_g$ of 490°C. The surface roughening may play a role in compositional inhomogeneity for InAlAs due to nonuniform adatom incorporation along the surface. As noted earlier in this section, small variations in $T_g$ slightly modulated the InAlAs composition, and hence the PL peak position varied from 1.42 to 1.47 eV.

4. InAlAs(111)B: Summary

InAlAs(111)B can be jointly optimized for surface roughness, optical, and electrical properties, and we have shown atomically smooth InAlAs(111)B surfaces with PL and electrical quality comparable to InAlAs(001). Elsewhere, we have shown that InAlAs(111)B grown under these conditions makes a suitable buffer and barrier material for high quality tensile GaAs(111)B quantum dashes with strong PL.9 Overall, our recommended growth conditions for InAlAs(111)B are high $T_g$ of 540°C and a narrow window of V/III ratio of 10–12.

D. InGaAs on InP(111)B

1. InGaAs(111)B: Surface morphology

We investigated growth of InGaAs(111)B by depositing 200 nm on offcut InP(111)B substrates at 0.17 μm/h, consistent with other InGaAs samples in this study. As for InAlAs(111)B growth, InGaAs(111)B maintains a (1 x 1) streaky RHEED pattern for all tested conditions.

Both $T_g$ and V/III ratio had similar effects on the morphology of offcut InGaAs(111)B as offcut InAlAs(111)B. From Nomarski images (Fig. 19), an overall improvement in surface roughness with increasing $T_g$ was observed. At the low $T_g$ of 350°C, a high density of kite-shaped features...
appear, and with increasing \( T_g \), these features changed shape and decreased in density, disappearing almost entirely by 485 °C. A low density of faint features remained on the surface at \( T_g = 485 \) °C, resembling oval-defects that often form during the growth of buffers on (001) surfaces.

AFM imaging shows that higher \( T_g \) also suppressed step bunching for InGaAs [Figs. 20(c) and 20(e)], just as it did for InAlAs(111)B. At \( T_g \leq 460 \) °C, pronounced macrosteps formed due to the merging of adjacent offcut steps into taller steps. In contrast to InAlAs(111)B, the InGaAs macrosteps were straight instead of wavy, and pit-shaped step-bunches were not observed. The macrosteps roughened the atomic surface significantly, and the rms roughness increased from 0.58 nm at 485 °C to 1.49 nm at 400 °C as step bunches became more pronounced. However, the surface was also roughened by elongated triangular features as \( T_g \) increased to 485 °C. These features may indicate the onset of faceting, as previously reported in (111)B growth.60,85–87

Optimization of the V/III ratio is also important for smooth InGaAs(111)B growth, again similar to offcut InAlAs(111)B. As we decreased V/III from 75 to 10 at \( T_g = 485 \) °C [Figs. 20(a)–20(c)], the triangular features became less pronounced, and then disappeared. However, a high density of pits emerged at V/III = 10, which was also observed for InAlAs(111)B grown at lower V/III. The optimal growth condition for surface smoothness lies between V/III = 10–25, a slightly larger window than we observed for InAlAs. In Nomarski, all samples were nearly featureless at 485 °C, independent of V/III ratio.

2. InGaAs(111)B: Electrical properties

Nominally undoped InGaAs(111)B exhibited an n-type carrier background. Depending on growth conditions, we measured \( n_0 \) values ranging from \( 2 \times 10^{16} \) to \( 1 \times 10^{17} \) cm\(^{-3}\), with \( \mu_n = 1300–4000 \) cm\(^2\)/V s. V/III ratio had a strong effect on electrical properties, where increasing V/III raised \( \mu_n \) and lowered \( n_0 \) [Figs. 21(a) and 21(b)]. The inverse relationship between \( \mu_n \) and \( n_0 \) suggests that ionized impurity scattering is one of the factors limiting \( \mu_n \). The relationship between V/III ratio and electrical properties is not monotonic, but instead \( \mu_n (n_0) \) is lowest (highest) at V/III = 25 and improves at both higher and lower V/III. However, atomic hills or pits appear at high or low V/III ratios [Figs. 20(a)–20(c)], indicating a small tradeoff between morphology and electrical properties during V/III optimization.

\( T_g \) is an equally important variable with \( \mu_n (n_0) \) increasing (reducing) with temperature [Figs. 21(c) and 21(d)]. Once
again, conditions that produced low \( n_0 \) also yielded high \( \mu_n \), confirming the role of ionized impurity scattering. Similar to the V/III trend, the improvement was not monotonic, but some degradation was observed for a moderate \( T_g \) of 460 °C. As before, the electrical quality was correlated with higher surface roughness, for reasons not currently understood. Overall improvement in \( l_n \) at high \( T_g \) was also found by Yeo et al., who reported Si-doped InGaAs(111)B with \( l_n \) as high as 5100 cm²/V s for \( T_g \) of 550 °C. We note that these values are still below the 8000–10 000 cm²/V s observed for InGaAs(001) in this electron density range, indicating significant scattering. This increased carrier scattering in InGaAs and InP growth on (111)B is likely linked to the high dislocation densities originating from the initial growth interface (Fig. 14).

3. InGaAs(111)B: Photoluminescence

The effect of \( T_g \) on the InGaAs(111)B PL is shown in Fig. 22(a). For all InGaAs(111)B samples observed, linewidths decreased under conditions where PL intensity increased. PL intensity [Fig. 22(b)] and linewidth [Fig. 22(c)] are optimized at \( T_g \sim 460 °C \) and V/III \( \sim 25 \), respectively, growth conditions that correspond to samples with lowest surface roughness. An increase in linewidth with surface roughness suggests that rougher surfaces have spatially nonuniform incorporation of indium and gallium adatoms. In some quaternary alloys, small adjustments in the surface orientation can significantly change the alloy composition, which can lead to compositional nonuniformity for rough surfaces. Comparing the PL in Fig. 22 to the Hall data in Fig. 21, PL intensity and \( n_0 \) are closely correlated. This indicates that the higher PL intensity at 460 °C is largely due to a faster radiative recombination due to higher \( n_0 \) and is less strongly related to material quality.

A similar assessment applies to the PL trend with V/III ratio where again, the optimum PL corresponded to the V/III condition that yielded the smoothest surface. At V/III \( = 25 \), both surface pits and faceting were avoided, which may lead to narrower linewidths due to better compositional uniformity. PL intensity and \( n_0 \) were closely correlated across V/III ratios (Figs. 21 and 22). As we increased V/III from 25 to 75, PL intensity decreased due to reduced \( n_0 \), even though material quality appears to be higher, based on higher \( l_n \) (Fig. 21).

4. InGaAs(111)B: Summary

In this section we have shown that the optimization of offset InGaAs(111)B morphology follows a similar trend to that of offset InAlAs(111)B. For InGaAs, moderate \( T_g \) of \( \sim 460 °C \) and V/III ratio of \( \sim 25 \) are needed to minimize surface roughness. A minimum rms roughness of 0.66 nm was obtained in InGaAs, which is somewhat higher than the 0.24 nm rms roughness achieved in InAlAs. The \( \mu_n \) of InGaAs(111)B has not reached the level of InGaAs(001) growth, but further improvements should be possible based on the observed trends. To optimize electrical transport properties, we recommend high \( T_g \) (≥485 °C) and V/III ratios (≈75). InGaAs PL intensity varies primarily due to \( n_0 \), while
the PL linewidth appears to correlate with surface smoothness. As a result, an acceptable compromise between the smoothness of InGaAs(111)B and its electrical and optical properties can be made, even though individually each property is optimized at different areas of the parameter space. Overall, our recommended growth conditions for InGaAs(111)B are $T_g = 485^\circ C$ and V/III ratio = 75.

V. GROWTH ON InP (110)

A. Review of growth on (110) surfaces

Previous work on (110) growth has primarily focused on GaAs homoepitaxy, although some studies exist for InGaAs, InAlAs, and other III–Vs. Early studies found that rough surfaces with elongated, faceted hillocks were formed under a wide range of $T_g$, V/III ratios, and growth rates. At the low V/III ratios typical for (001) growth, metal droplets formed on the (110) surface, indicating a low sticking coefficient for arsenic. Consistent with the poor morphology, carrier mobilities were very low and PL emission was dominated by defect levels.

Analysis of the hillocks created during on-axis GaAs(001) growth showed that they are elongated along [001] with a prominent (111)A facet. The problem of hillocks was thus resolved by using an offcut 6° toward the (111)A facet. Subsequently, the same offcut technique has been used to achieve smooth InGaAs, InAlAs, InGaAsP, and InP on InP(110).

For growth on exact (110) surfaces, Zhou et al. reported that by using relatively low growth rate, high V/III ratio, and low $T_g$ of 470–500 °C, they could create AlGaAs/GaAs(110) heterostructures with sufficiently good interface quality to form a high $\mu_n$ 2D electron gas (2DEG). Pfeiffer et al. subsequently confirmed that similar growth conditions produced smooth surfaces, and demonstrated a 2DEG with very high $\mu_n$ via growth on a cleaved (110) wafer edge. Wassermeier et al. later investigated GaAs(110) growth trends by AFM, and eliminated hillocks using elevated V/III ratio and reduced growth rate to produce atomically smooth surfaces with step-flow growth.

In contrast, InGaAs and InAlAs growth on exact InP(110) have received little study. Brown et al. found that smooth InGaAs and InAlAs could only be formed using an extremely low temperature of 300 °C and a high V/III of 80. Yasuda et al. confirmed this trend for InAlAs(110), showing the formation of large, triangular hillocks above this temperature. In this section, we present a brief study of InAlAs(110), followed by a detailed growth parameter study of InGaAs(110).

B. InP(110) homoepitaxy

The development of homoepitaxial, on-axis InP(110) buffers may promote the growth of smooth InAlAs(110) and InGaAs(110) epilayers. InP(110) could not be experimentally investigated by our group however, due to equipment problems. Previous work on InP(110) growth on exact substrates shows a highly faceted surface. Consistent with the growth of other (110) III–Vs (Sec. V A). Bhat et al. showed by MOCVD that specular surfaces nearly free of defects can be obtained for a 3° offcut toward (111)A or (111)B, and they suggest that further improvement is possible with a higher offcut. Mitsuhara et al. confirmed this result for chemical beam epitaxy growth, and found gradual improvement in the surface morphology with offcut angle up to 6°. Smooth growth of GaAs(110) as well as InGaAs(110) and InAlAs(110) can be achieved using very
and high V/III ratios (Secs. VA, VC, and VD), and a similar strategy may be applicable to the growth of InP(110) buffers in future work.

C. InAlAs on InP(110)

1. InAlAs(110): Surface morphology

Following the work of Brown et al., we utilized a low $T_g$ of 280 °C and high V/III ratio of 80 to achieve smooth InAlAs growth on InP(110) substrates. Following desorption of the native oxide from the InP substrate at 500 °C under As$_4$, we deposited 200 nm of InAlAs at 0.5 μm/h. During each InAlAs growth, a streaky (1x1) RHEED pattern was observed. This compares well with the (1x1) pattern reported for pristine GaAs(110) surfaces (before growth) where surface atoms are displaced, yet retain the (1x1) periodicity. Other reconstructions for growth of III–Vs on (110) surfaces have not been reported to our knowledge.

Nomarski images confirmed that the InAlAs(110) surface was smooth and featureless (not shown), while AFM images showed a smooth surface with 0.35 nm rms over a 15 x 15 μm$^2$ area (Fig. 23). The absence of faceted hillocks is consistent with previous reports that high V/III and low $T_g$ promote a smooth (110) surface. The atomic surface morphology was disordered, and defined neither by step-flow growth nor clear indications of 2D islanding. In addition, small pits were visible in the 1x1 μm$^2$ image [Fig. 23(a) inset] that are 1.0–1.5 nm deep. We attribute this morphology to the use of a low $T_g$ that prevents adatoms from diffusing to their lowest energy sites and forming clear terraces. We attempted to smooth the InAlAs(110) further using annealing at 500 °C under As$_4$, based on a report by Yoshita et al., but rms roughness was unchanged [Fig. 23(b)].

While the use of low $T_g$ produces an atomically smooth surface, the microstructure suffers from a high density of stacking faults (SFs) that emerge from the InAlAs/InP growth interface (Fig. 24). A high SF density was also observed for InAlAs(110) in Ref. 38 under similar conditions. We have confirmed by XRD that the InAlAs composition is nearly lattice-matched to InP, so the SF nucleation is not induced by the lattice mismatch. To prevent SF formation, we suggest that the process by which InAlAs growth is initiated could be optimized.

2. InAlAs(110): Photoluminescence

PL for the annealed and nonannealed InAlAs(110) samples at 77 K is shown in Fig. 25. The nonannealed sample
has an emission peak at 1.47 eV, with a FWHM of 48 meV. PL from the annealed sample was red-shifted to $\sim 1.39$ eV (overlapping the InP peak). The group-III composition from XRD was found to be 51.2% indium in both samples, so the expected bandgap at 77 K is 1.54 eV. The 1.47 eV peak for low-$T_g$ InAlAs is consistent with the report by Brown et al., who attributed the reduction in emission energy to a defect-assisted transition. The reason for the redshift of the InAlAs PL after annealing is not currently understood. However, Ohbu et al. observed a similar effect in low-$T_g$ GaAs, where high temperature annealing extinguished the near-band edge emission, while creating new luminescent defect levels. It is also possible that the InAlAs PL has been quenched by the anneal, while the shoulder on the InP peak results from a new defect-assisted transition near the InP-InAlAs interface.

The presence of defect levels within the InAlAs is not surprising, given that the low $T_g$ required for smooth growth also promotes incorporation of excess As and other point defects. The PL of both InAlAs(110) samples is much weaker than typical for InAlAs(001) (not shown). The relatively weak InAlAs(110) PL suggests that point and extended defects in the buffer contribute significantly to non-radiative recombination. Increasing $T_g$ regime is clearly desirable, to mitigate point defects and improve the optical properties. However, our attempts to increase $T_g$ to 320°C led to the creation of hillocks on the surface, similar to the case of InGaAs(110) discussed below.

### 3. InAlAs(110): Summary

The growth of smooth InAlAs on InP(110) is possible using low $T_g$ and high V/III, although defects from stacking faults and excess arsenic impair the optical quality. Overall, our recommended growth conditions for InAlAs(110) follow Brown et al.: $T_g = 280$ °C and V/III ratio = 80. However, this recommendation is tentative, since substantial improvements are needed to improve the radiative efficiency.
D. InGaAs on InP(110)

1. InGaAs(110): Surface morphology

We studied the effect of growth conditions for 200 nm InGaAs(110) grown at 0.17 μm/h. For a fixed V/III = 75, the effect of T_g on surface morphology is shown by the Nomarski images in Fig. 26. A high density of trapezoidal hillocks appeared at all T_g, increasing in size and decreasing in density as T_g was increased. The effect of T_g on InGaAs(110) morphology in Fig. 26 is very similar to the trend noted by Yasuda et al. for InAlAs(110); very low T_g is required to eliminate hillock formation. However, even at 280 °C, faint hillocks can still be seen by Nomarski, albeit with weak contrast [Fig. 26(d)]. We conclude therefore that InGaAs requires even lower T_g than InAlAs to achieve smooth growth. The high mobility of Ga adatoms means that even lower T_g is required to prevent their incorporation into hillocks. As for InAlAs(110), we observed a streaky RHEED (1 × 1) pattern for InGaAs(110) growths at 280 °C.

The faceted hillocks on both InGaAs(110) and InAlAs(110) exhibit trapezoidal/nearly triangular geometry over a wide range of T_g. The hillocks have mirror symmetry along the in-plane [001] direction, as reported for GaAs(110) buffers. For InGaAs and InAlAs, the asymmetric morphology is indicative of different adatom incorporation rates on different hillock facets, similar to GaAs(110). The formation of facets indicates that the (110) surface is less energetically stable than the new facets that emerge. Our results suggest that suppressing adatom diffusion kinetics through the use of low T_g prevents adatoms from incorporating into hillocks.

The influence of V/III ratio on InGaAs(110) morphology is shown in Fig. 27 for samples grown at T_g = 280 °C; the sample at V/III = 75 is the same as in Fig. 26(d). Small hillocks and hillock pairs clearly remain for all V/III ratios up to ~75. The presence and morphology of hillocks on GaAs(110) have been attributed to asymmetric adatom incorporation along the [001] direction, which may also produce the hillock pairs observed here. Hillock size and density both decreased with increasing V/III, disappearing entirely at V/III = 195 and yielding a smooth surface with a 15 × 15 μm^2 rms roughness of 0.35 nm. Elevated V/III thus has the same effect as reduced T_g, where lower adatom mobility prevents adatom incorporation into faceted hillock structures.

Although low T_g and high V/III ratio promote smooth InGaAs and InAlAs(110) growth, these conditions also create point defects that degrade the optical and electrical properties, as discussed below. We thus attempted to increase T_g of the InGaAs(110) to 320 °C, while maintaining a high V/III of 195 (not shown). Hillocks formed once again, with size and density similar to that of Fig. 27(b). We conclude that low T_g is essential for smooth InGaAs and InAlAs buffer growth on (110) surfaces.

2. InGaAs(110): Electrical properties

The effects of T_g and V/III ratio on n_0 are shown in Figs. 28(a) and 28(c). InGaAs(110) samples grown with T_g > 350 °C were too resistive to measure, and all other...
samples were unintentionally $n$-type. $T_g$ strongly affects $n_0$ values [Fig. 28(c)], raising $n_0$ from $1 \times 10^{15}$ to $4 \times 10^{16}$ cm$^{-3}$ as $T_g$ was reduced from 350 to 280°C, for V/III = 75. Similar trends in $n_0$ values have been reported for (001)-oriented III–Vs grown in this low $T_g$ range, including GaAs (Ref. 114) and InGaAs. As $T_g$ is reduced, the arsenic desorption rate falls, allowing a higher amount of arsenic to incorporate into the crystal than normally allowed by stoichiometry. Excess arsenic creates antisite defects (As$_{Ga}$), occupying cation sites in the crystal lattice and thereby functioning as electron donors. The density of As$_{Ga}$ defects increases, and hence $n_0$ in the InGaAs increases at lower $T_g$. We observed a similar increase in $n_0$ for InGaAs(110) when V/III ratio was increased, as shown in Fig. 28(a). For samples at 280°C, increasing V/III from 25 to 195 increased $n_0$ from $1 \times 10^{16}$ to $2 \times 10^{17}$ cm$^{-3}$. This behavior is again consistent with elevated levels of As$_{Ga}$ antisite defects in the crystal as a result of the higher As$_4$ overpressure.

The effect of growth parameters on $\mu_n$ is shown in Figs. 28(b) and 28(d) for samples grown at 280°C. As V/III ratio was increased we observed a monotonic decrease in $\mu_n$, which is likely caused by impurity and Coulomb scattering from elevated levels of As$_{Ga}$. Charged and neutral states of As$_{Ga}$ are typically present after low temperature growth, and both contribute to electron scattering.

The effect of $T_g$ on $\mu_n$ is more complex, such that the $T_g$ trend depends on V/III ratio. At V/III = 195, $\mu_n$ increases with increasing $T_g$, while for V/III = 75, $\mu_n$ decreases with $T_g$. For V/III = 195, the improvement in $\mu_n$ with $T_g$ corresponds to the observed decrease in $n_0$ shown in Figs. 28(c) and 28(d). This can be explained by a reduction in impurity scattering from charged As$_{Ga}$, since As$_{Ga}$ concentration decreases with increasing $T_g$. The unexpected $\mu_n$ trend at V/III = 75 is difficult to elucidate, but may be related to hillocks that become larger at lower V/III. Christou et al. have shown that antiphase domains can cause rough InAlAs/InGaAs(110) surfaces. Roughening and defects associated with the hillocks are scattering sources that may reduce $\mu_n$, but further work is needed to confirm this hypothesis. We note that the sample grown at 400°C and V/III = 75 was too resistive for Hall measurement, which could be caused by further degradation in $\mu_n$ as larger hillocks form. In summary, the electrical properties of InGaAs(110) are influenced by a combination of factors including arsenic antisite defects, and possibly hillocks.

Overall, the lowest $n_0$ value for InGaAs(110) was achieved under the lowest V/III ($\sim 25$) and the highest $T_g$ (350°C) conditions explored in this study. Meanwhile, the best $\mu_n$ values were obtained at higher $T_g$ (320°C) and high V/III to minimize As$_{Ga}$, or low $T_g$ (280°C) and low V/III, where hillocks play a weaker role. Under these guidelines, $\mu_n \sim 6500$ cm$^2$/V s and $n_0 \sim 1 \times 10^{16}$ cm$^{-3}$ were obtained in this work; these are the highest $\mu_n$ values obtained in the present work, but still substantially lower than achieved on the (001) surface.

3. InGaAs(110): Photoluminescence

InGaAs(110) PL is sensitive to $T_g$ and V/III ratio as shown in Fig. 29. As in the case of InAlAs(110), InGaAs(110) PL is weak at 300 K, so we performed measurements at 77 K. Since PL intensities were affected by sample positioning in the cryostat, the spectra in Fig. 29(a) are normalized. All InGaAs samples show a primary PL peak at 790–810 meV, while additional, low-energy peaks emerge for samples grown at higher $T_g$ [Fig. 29(a)]. Our PL measurements agree with the calculated band gap of 804 meV for lattice-matched InGaAs at 77 K. Unlike InAlAs(110), the InGaAs(110) emission appears less susceptible to shallow luminescent defects formed during low temperature growth. At V/III = 75, as $T_g$ was raised from 280 to 350°C a new peak emerges at 763 meV, and at 420°C, another peak at
730 meV is also present. At these higher temperatures, excess-arsenic incorporation in InGaAs is greatly mitigated, suggesting that the new peaks are not arsenic-related defects. These peaks instead form concurrently with large faceted hillocks on the InGaAs(110) surface. Facet-dependent incorporation rates have been observed for a number of materials including GaAs, InGaN, and InGaAsP. For the latter two materials, different incorporation rates of each atomic species produce orientation-dependent alloy compositions. Similarly, for InGaAs(110), indium and gallium are likely to incorporate at different rates on different facets, leading to the appearance of the additional PL peaks. Note that the increase in the PL linewidth with $T_g$ is also consistent with increased compositional nonuniformity.

The effects of growth conditions on PL linewidth are shown in Figs. 29(b) and 29(c). In Fig. 29(b), a strong increase in FWHM is associated with increasing $T_g$, reflecting the formation of multiple PL peaks as hillocks emerged. The V/III trend in Fig. 29(c) shows that higher V/III ratios are needed to maintain lower linewidths. In this case, separate peaks were not discernible in the spectra (not shown), but small hillocks were present at $V/III = 25$ that could be responsible for broadening the linewidth. Thus, the low $T_g$ and high V/III conditions corresponding to smooth surfaces are also most favorable for narrowing PL linewidths.

4. InGaAs(110): Summary

In summary, InGaAs on InP(110) requires very low $T_g$ ($\sim 280^\circ$C) and very high V/III ratios ($\sim 200$) to form smooth surfaces. Electrical properties can be improved outside of this growth window, but only at the cost of hillock formation. The highest $\mu_n$ and $n_0$ values were observed at low V/III $\sim 25$ with $T_g = 280^\circ$C, or at high V/III $\sim 195$ with $T_g = 320^\circ$C. These characteristics appear to be influenced by AsGa incorporation as well as hillock formation. The narrowest PL linewidths were achieved at conditions that promoted smooth morphologies. At present the morphological, electrical and optical properties of InGaAs(110) cannot be simultaneously optimized, and the choice of growth conditions must therefore be informed by the desired application for the material.

VI. EFFECT OF SURFACE ORIENTATION ON GROWTH PARAMETERS

In this section, we compare trends in material properties across different surface orientations. We compare our experimental results for InAlAs and InGaAs to previous GaAs results found in the literature (from Secs. III A, IV A, and V A). We present parameter space maps as a roadmap to the optimal growth conditions for InAlAs, InGaAs and GaAs. GaAs homoepitaxy on exact (111)A, (111)B, and (110) surfaces results in hillocks that share the symmetry of the crystal lattice, as discussed in Secs. III A, IV A, and V A. GaAs(111)B forms hillocks elongated along [001], reflecting the twofold surface symmetry, while the hillocks for GaAs(111)B and (111)A are threefold symmetric. Hillocks on GaAs(111)B are broader and shorter than those on GaAs(111)A, since the former hillocks are composed of vicinal surfaces. The major surface features observed for InAlAs and InGaAs in this work are reviewed in Fig. 30. The InAlAs(111)B and InGaAs(111)B morphologies are notably free from hillocks as a direct result of the 2° substrate offset. Instead, the (111)B surfaces are dominated by closely spaced...
offcut steps, which can group together into larger morphological
defects such as step-bunched pits and macrosteps.

Figure 31 shows the optimal growth windows to achieve
smooth surfaces for InAlAs, InGaAs and GaAs for each of
the low index surfaces discussed in this work. The darker
shaded areas indicate conditions where the surface is smooth-
est. Smooth GaAs (001) homoepitaxy is possible across a
wide range of temperatures and V/III ratios. In contrast, the
growth windows for other materials are typically much nar-
rower, outside of which we see surface roughening and crys-
talline defects. Note that the GaAs temperature range
extends to higher values than InAlAs and InGaAs for some
surface orientations. These high temperature ranges are not
compatible with indium containing compounds due to their
lower decomposition temperature of ~550 °C. Turning to
non-(001) surfaces, the optimal growth conditions have a lim-
ited range for most materials and generally do not overlap for
the different surface orientations. As a result, ideal growth
conditions for (001) surfaces often make poor starting points
for investigating growth on (110) or (111) surfaces.

Crystallographic orientation plays a prominent role in
determining the influence of growth conditions on morphol-
ogy, while the material itself (e.g., InGaAs versus InAlAs)
plays a lesser role. The three non-(001) substrate orientations
each require a different growth strategy to achieve a smooth
surface. For most (110) and (111)A materials discussed in
this work, hillocks increase in size and decrease in density as
adatom diffusion length increases. In these cases, the two
limiting approaches for achieving surface smoothness are (1)
to lower adatom mobility until hillocks shrink to zero size;
or (2) to raise adatom mobility to increase the spacing
between the hillocks as much as possible.

1. Growth on (111)A

For (111)A growth, low adatom mobility conditions fail to
smooth the surface for some materials because either new sur-
face defects form (InAlAs) or because the hillocks do not reach
zero size (InGaAs). Therefore we recommend adopting a high
adatom mobility strategy that uses high $T_g$ to lower the hillock
density as much as possible. In contrast, GaAs(111)A can be
grown using low adatom mobility conditions, since morpho-
logical problems were not observed at low $T_g$.\textsuperscript{59} Between
InAlAs and InGaAs, V/III ratio plays a much different role.
Smooth InGaAs(111)A requires lower V/III, consistent with
raising adatom mobility to remove hillocks. InAlAs(111)A
requires higher V/III ratios to smooth the surface, a trend that
suggests it behaves more like GaAs(111)A, in which hillocks
are associated with the low arsenic sticking coefficient.\textsuperscript{59}

The (111)A surfaces of InAlAs and InGaAs have the addi-
tional complexity of atomic wedding cakes that arise from the
nucleation of 2D islands. GaAs(111)A differs from InAlAs and
InGaAs in this respect, since wedding cakes are not observed
near the ideal GaAs growth conditions.\textsuperscript{27} For InAlAs and
InGaAs, growth conditions that reduce the occurrence of wed-
ding cakes, presumably by reducing the 2D nucleation rate,
include high $T_g$ and high V/III ratio. This trend nearly overlaps
the strategy for hillock reduction in these ternaries, though in
the case of InGaAs(111)A, V/III ratio requires a slight tradeoff between hillock density and wedding cake roughness.

2. Growth on (111)B

The use of offcut InP(111)B substrates in this work for InGaAs and InAlAs growth changes the relationship between morphology and growth conditions compared to on-axis substrates. Little is understood about the smoothening and roughening mechanisms of offcut (111)B, so we can only comment on empirical trends observed during growth optimization. For InAlAs(111)B and InGaAs(111)B, high $T_g$ is required to avoid step bunching, while in contrast, high $T_g$ causes step bunching for offcut GaAs(111)B. However, the effect of lower $T_g$ on offcut GaAs(111)B has not been reported, and this approach could potentially exhibit similarities with InAlAs and InGaAs(111)B. In addition, the ternary (111)B materials show different forms of roughening for low or high V/III extremes. InAlAs and InGaAs both form pits at low V/III, and facets appear at high V/III. Literature reports suggest that GaAs(111)B is less sensitive to high V/III than the ternaries studied here.

3. Growth on (110)

For (110) growth, high adatom mobility is less desirable because of the large hillock size and density observed at higher $T_g$. Thus, for all materials grown on (110) in Fig. 31, a low adatom mobility approach is preferred where low $T_g$
and high V/III ratio are used to minimize hillock size. The
temperature required to smooth InAlAs and InGaAs
(∼300 °C) is much lower than required for GaAs
(470–500 °C), which is a function of the higher surface
diffusivity of indium adatoms compared to gallium.

B. Effects of surface orientation on electrical
properties

With the exception of InAlAs(111)B, electrical measure-
ments on InAlAs films were not possible. As a result, the
discussion here is limited to InGaAs growth. The effect of
surface orientation on the electrical properties of InGaAs is
independent of the surface orientation to some extent (Figs.
32 and 33). First, all InGaAs samples measured were uninten-
tionally n-type, regardless of surface orientation; an n-
type background is also the norm for In-containing III–V
semiconductors on (001) surfaces. Though we have not fully
optimized conditions for electrical properties, \( \mu_n \) values
obtained for non-(001) growth are reasonably high at
4000–6500 cm²/V s, where \( \mu_n(110) > \mu_n(111)A > \mu_n(111)B \).

\( n_0 \) varies greatly depending on surface orientation and
growth conditions as shown in Fig. 32. The InGaAs(111)A
epilayers exhibited the lowest \( n_0 \) of 4 × 10¹⁴ cm⁻³ (Fig. 32),
a value that is comparable to typical MBE-grown GaAs(001). InGaAs(110) and (111)B had significantly
higher \( n_0 \) of 6 × 10¹⁵ cm⁻³. Trends in \( \mu_n \) as a function of \( T_g \),
V/III ratio, and surface orientation are shown in Fig. 33.
Similar growth conditions were found to optimize \( \mu_n \) for
InGaAs(111)A and (111)B, namely high \( T_g \) and high V/III
ratio. For both (111) surfaces, these conditions corresponded
to lower \( n_0 \) (Fig. 32), indicating that reduced charged impurity
scattering plays a role. In the case of InGaAs(111)A, improved
\( \mu_n \) also corresponded to reduced hillock density. Though we
have not fully optimized conditions for electrical properties,
\( \mu_n \) values obtained for non-(001) growth are reasonably high at
4000–6500 cm²/V s, where \( \mu_n(110) > \mu_n(111)A > \mu_n(111)B \). However, \( \mu_n \) values of 6000–10000 cm²/V s are typical for
InGaAs(001) at similar carrier concentrations, suggesting
that electron scattering is higher in non-(001) InGaAs. Further optimization of growth conditions and crystal quality
should improve the \( \mu_n \) values on each surface.

Hillocks on the (111)A surface produce substantial rough-
ness and also contain high densities of crystallographic
defects, making their elimination crucial to achieving high
\( \mu_n \). In contrast, InGaAs(111)B surface roughness does not
change dramatically with growth conditions, so surface mor-
phology is less of a consideration for electrical properties.

For InGaAs(110), Fig. 33 shows that much lower \( T_g \) is
required for high \( \mu_n \) than on the (111) surfaces. Low \( T_g \) is
essential for smoothing the (110) surface (see Fig. 31), which
reduces surface scattering and avoids crystallographic defects
associated with the hillocks. However, the optimum point in
Fig. 33(c) lies at slightly higher \( T_g \) and lower V/III ratio than
the InGaAs(110) morphology optimum in Fig. 31. At very low
\( T_g \) and high V/III ratios, high densities of arsenic-related
defects appear that also degrade \( \mu_n \). Figure 32 illustrates
the remarkable increase in \( n_0 \) as \( T_g \) was lowered from 320 to
280 °C, corresponding to the incorporation of arsenic antisite
defects. Therefore, the extreme conditions that smooth
the morphology do not necessarily improve the electrical
properties, and some compromise must be made between the two.
C. Effects of surface orientation on photoluminescence

PL from InAlAs (111)A and (111)B were relatively insensitive to growth conditions, while PL from InAlAs(110) was weak and dominated by defects; it is worthwhile to mention that InAlAs(111)B exhibited integrated intensity \( \sim 40\% \) of InAlAs(001). No strong trends were noted for InAlAs PL, and thus, as in Sec. VI C, we focus our discussion on InGaAs growth.

PL linewidths on (110), (111)A, and (111)B generally narrowed as the surface became smoother, which suggests that the uniformity of indium and gallium incorporation is associated with surface roughness. For (111)A and (111)B samples, PL linewidth was found to reduce with increasing PL intensity, such that both PL attributes improved simultaneously.

The effect of growth parameters on PL intensity at 300 K for InGaAs (111)B and (111)A is shown in Fig. 34; the low signal-to-noise ratio for PL of InGaAs(110) at 300 K prevented a meaningful analysis for this orientation. Similar to the morphology trends, PL intensity varied significantly with surface orientation. InGaAs(111)A required a higher \( T_g \) to optimize PL \((\sim 510^\circ \text{C})\) than InGaAs(111)B \((\sim 460^\circ \text{C})\). For (111)A, high \( T_g \) corresponded to low hillock densities and higher \( \mu_n \) for InGaAs, both of which provide evidence for higher crystalline quality. In contrast, the moderate \( T_g \) used to increase the PL intensity of InGaAs(111)B in Fig. 34(b) corresponded to elevated \( n_0 \) levels in Fig. 32. Thus, when interpreting PL intensities, it is important to consider the fact that higher \( n_0 \) increases the radiative recombination rate. InGaAs(111)A is influenced by \( n_0 \) to a weaker extent, in that the high V/III used to improve the PL also slightly increased the \( n_0 \) level. However, the \( T_g \) trend for InGaAs(111)A is not related to \( n_0 \) and instead indicates improvement in optical quality.

We compare the PL spectra of InGaAs (111)A, (111)B, and (110) in Fig. 35. The PL intensity of InGaAs(111)B is the strongest, four times more intense than that of InGaAs(111)A and nearly 20\( \times \) stronger than InGaAs(110). That PL from InGaAs(111)B is stronger than InGaAs(111)A is largely attributed to the former’s much higher \( n_0 \) (Fig. 32). The PL of InGaAs(110) is weaker than the others due to the low \( T_g \) and concomitant arsenic-antisite defects. InGaAs(111)A and InGaAs(111)B have good optical quality, while further work is needed to minimize nonradiative recombination in InGaAs(110) and improve luminescence.

D. Overview of buffer optimization

Finally, we compare the effect of surface orientation on the simultaneous optimization of the morphology, electrical, and optical properties of InGaAs. Figure 36 compares the points in parameter space that optimize the surface morphology (rms roughness), the PL intensity, and \( \mu_n \) for InGaAs on each surface orientation. InGaAs(111)A has the best overlap of properties in parameter space, where the highest \( \mu_n \), strongest PL, and lowest hillock density all occur at V/III \( \sim 75 \) and \( \sim 510^\circ \text{C} \). For InGaAs(111)B, some separation exists between optimal surface roughness and the highest \( \mu_n \), where a higher \( T_g \) and V/III ratio were needed for better electrical properties. InGaAs(110) is the most difficult to jointly optimize, since surface morphology is anticorrelated with \( \mu_n \). Optimal morphology was produced at low \( T_g \) and high V/III ratio to suppress hillocks, while higher \( T_g \) and low V/III...
were needed to mitigate excess-arsenic for improvements in $\mu_n$. The combined trends for each surface show the extent to which compromises must be made when optimizing material properties. While precise growth conditions are expected to vary between different research groups and different MBE chambers, we believe that the trends we have reported will be robust and useful to other researchers.

VII. CONCLUSIONS

We have presented a literature review and experimental investigation into the optimal conditions for MBE growth of InAlAs and InGaAs on InP (111)A, (111)B, and (110) substrates. We report trends in growth parameters toward the optimization of surface morphology, electrical properties, and optical quality. We have emphasized morphological optimization, and for each surface, we have shown that atomic level smoothness is possible, and that large morphological defects can either be eliminated or minimized. Growth conditions for optimal smoothness vary substantially with surface orientation. However, we report that different materials appear to have similar growth trends for a given surface orientation [e.g., InGaAs(110) behaves similarly to InAlAs(110)], meaning that insights learned from a particular surface orientation could perhaps be used to develop a range of materials on that same surface.

Strong electrical and optical properties can be obtained from most of the materials studied in this work, despite the challenges with achieving flat surface morphologies. We measured electron mobilities at 300 K as high as 4000, 5300, and 6500 cm$^2$/V s for InGaAs on InP (111)B, (111)A, and (110), respectively. InAlAs(111)B was particularly noteworthy in its optoelectronic properties, exhibiting higher $\mu_n$ and comparable PL intensity to InAlAs(001). We anticipate that the trends in electrical and optical properties we report here will enable the growth of device quality InGaAs and InAlAs on each of these surfaces.

This work provides a foundation for the development of novel materials and devices on the (111) and (110) surfaces of InP. As an example, the InAlAs and InGaAs layers explored here have already been employed to facilitate growth of highly tensile quantum dots on (111) and (110) surfaces.\cite{9,10,30} This achievement allows strong bandgap tuning for long wavelength optoelectronics. Furthermore, symmetric QDs with ideal electronic characteristics for entangled photon emitters have been demonstrated using the InAlAs/InP(111)A buffers achieved in our experiments.\cite{10,21} GaAs(110) quantum wells have been used to make encouraging spintronic devices,\cite{33-36} and the developments we report here may soon help enable InP(110)-based spintronic devices. Smooth buffer layers on InP (111) surfaces are anticipated to provide ideal templates for the growth of topological insulators and transition metal dichalcogenides. Finally, MOSFETs that employ smooth, high-mobility III–V materials grown on (111) surfaces are predicted to have characteristics that rival silicon. III–Vs based on (111) and (110) materials are anticipated to drive a host of new technologies in the upcoming years.

ACKNOWLEDGMENTS

Microscopy facilities used in this work were supported by the Yale Institute for Nanoscience and Quantum Engineering and National Science Foundation MRSEC DMR 1119826. C.D.Y. acknowledges support from the Department of Energy Office of Science Graduate Fellowship Program (DOE SCGF), made possible in part by the American Recovery and Reinvestment Act of 2009, administered by ORISE-ORAU under Contract No. DE-AC05-06OR23100. Additional support was provided by the University of California Lab Fees Research Program (Grant No. 12-LR-238568).

\begin{thebibliography}{10}
\bibitem{1} P. M. Solomon and S. E. Laux, \textit{IEDM Tech. Dig.} \textbf{2001}, 95.
\end{thebibliography}
124T. P. Pearsall, GaInAsP Alloy Semiconductors (Wiley, New York, 1982).