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Invited Paper

Development of lateral epitaxial overgrown InAs microstructure on patterned (100) GaAs substrates

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Backscattered electron Kikuchi pattern-based orientation imaging was used to investigate the origin of the improved epitaxial alignment that is realized when a lateral epitaxial overgrowth approach is used for the growth of InAs on GaAs. The island size at coalescence appears to be critical in determining whether a single or multi-fold tilted epitaxial orientation relationship(s) is (are) present in the film. Sub-micron (~ 0.5 μm or less) island sizes at coalescence appear to lead to a single orientation aligned with the GaAs. This work shows that spatial constraints imposed at the early stages of growth, in this case through use of a mask-patterned substrate, can be used to promote coalescence at small island size as an alternative or parallel approach to setting growth conditions (temperature, precursor stoichiometry, etc) in order to control the nucleation and growth kinetics.

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1. Introduction

The potential for new high-performance devices that is offered by the small band gap arsenide and antimonide semiconductors [1,2], together with the current lack of commercially-produced, lattice-matched, semi-insulating substrates for these materials, has stimulated renewed interest in heteroepitaxy of cubic compound semiconductors on GaAs and InP. To their detriment, the approximately 7% lattice mismatch between the "6.1 Å" materials (GaSb, InAs, InSb) and GaAs generally translates to high defect densities and complex microstructures when these materials are grown directly on as-received GaAs wafers using standard growth techniques. In the case of MOVPE-grown InAs on GaAs, not only the crystal quality and microstructure, but also the orientational purity of the film is sensitive to the gas-phase stoichiometry and growth temperature. Films grown at high temperatures (700 °C) and high V-III ratio tend to consist of columnar grains, each of which is tilted a few degrees relative to the substrate in one of several directions. The mean-linear-intercept grain size of the columnar grains in the plane of the film is one to a few micrometers. Films grown at lower temperature (500°C) or at a lower V-III ratio have singular orientation as evidence by a single, albeit rather broad for applications in electronics, InAs peak in (400) rocking curves. Parallel studies to those reported in this paper have shown that the tilt forms during the island stage of film growth [3]. Multiple tilted regions are not only present in each island, but their arrangement within the islands appears to be systematic.

The tilting that occurs at higher temperatures can be reduced or even ameliorated through use of a lateral epitaxial overgrowth (LEO) method [4]. Fig. 1a schematically illustrates the basic concept of LEO as a means to grow high crystal quality epitaxial films on appreciably lattice mismatched substrates. The LEO substrate consists of a wafer on which an amorphous mask material has been deposited, into which a pattern is etched to expose the original substrate surface in 'windows.' The pattern commonly consists of holes, or, as in the present case, stripes oriented along a favorable crystallographic direction. The epi-layer nucleates in and initially grows to fill the windows. This material typically has the epitaxial orientation and defect density characteristic of growth on an unmodified substrate. As the deposit grows out over the amorphous mask, however, it is no longer constrained to match the substrate lattice parameter, and thus significant defect density reduction is realized over the volume of the film. In studies leading up to the one reported here [4], two improvements in InAs film quality were observed for films grown on LEO substrates compared to those grown on unmasked substrates using the same nominal growth conditions. First, the treading dislocation density in the LEO InAs material decreased by more than an order of magnitude and perhaps as much as three orders when compared to the conventional film. Second, a singular orientation was realized at 700 °C when mask-patterned substrates with stripe-shaped windows were used for LEO growth of InAs on GaAs, provided the windows were less than a micrometer in width. The magnitude of the tilt obtained in LEO films grown under the same conditions but with wider windows increased as the width increased from 2 μm to 5 μm to infinity (unpatterned substrate). Fig.

1b demonstrates that surfaces roughness is a possible cost of the improved crystal quality obtained via the LEO process. Whether the surface can be planarized for subsequent device growth though optimization of growth conditions or post growth planarization remains an open question

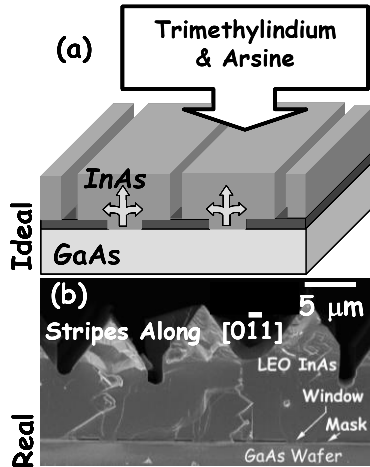

Fig. 1. (a) Schematic of the LEO process. (b) Secondary electron SEM micrograph of cross-sectional fracture surface of LEO InAs grown on a mask patterned GaAs substrate with 0.8 µm wide windows.

The conditions described above that lead to singular orientation all correspond to circumstances under which the InAs nucleation rate is expected to be high: low temperature, low V-III ratio, and a large mask area to window area ratio on a LEO substrate. All point toward control of the nucleation kinetics, be it chemically or configurationally, as a key to growth of films with singular orientation. To further explore how substrate configuration can be used to control nucleation and/or promote the orientational purity of epitaxial films, this study investigated the spatial distribution of pre-coalesced islands formed in short time growths and their crystal orientations in InAs films grown on GaAs by LEO.

2. Experimental methods

The LEO substrates used in this work consisted of 0°-miscut (100) GaAs wafers onto which ~120 nm of SiO_2 were deposited by low-pressure chemical vapor deposition. Stripe-shaped windows of width 0.8 µm and 5 µm were etched into the SiO_2 at 10 µm pitch using standard photolithographic and etching techniques. The long axis of the stripes was oriented along the $[01\bar{1}]_{GaAs}$ to maximize the lateral growth rate of the InAs. InAs epilayers were grown by metalorganic chemical vapor deposition in a horizontal quartz reactor at 700 °C and 78 Torr. The substrates were annealed in arsine for 10 minutes at 700 °C to desorb the native surface oxides prior to growth. Trimethylindium and arsine were used as precursors in a Pd-diffused H_2 carrier gas at a V/III ratio of 80. Films were grown to various thicknesses, ranging from un-coalesced islands to thick coalesced films in order to study the evolution of the microstructure and crystal orientation. Each growth run included both LEO and unpatterned (100) GaAs (i.e., control) substrates. InAs was grown directly on the GaAs/SiO_2 composite surface; no buffer layers were introduced. SEM, x-ray diffraction and cross-sectional transmission electron microscopy (TEM) techniques were used to characterize the growth topography, crystal quality and internal defect microstructure. Backscattered electron kikuchi patterns (BEKPs) collected in a scanning electron microscope (SEM) were used to determine local crystal orientation with 0.5° precision at a spatial resolution of 100 nm [5]. Both the as-deposited surface and polished cross-sections were examined.

BEKPs were collected in spot and in scanning modes. In spot mode, the electron beam is positioned at a point of interest in the sample, for example, the dot in the Fig 2a and a Kikuchi pattern, Fig. 2b, is collected, stored and later analyzed. In scanning mode, the electron beam is rastered, for example over the area outlined in 2a. BEKPs are collected and analyzed automatically as the beam scans. Spatial coordinates and Euler angles that describe the location and orientation of the crystal, respectively, are stored, and orientation images like those shown in Figs. 2-4 are constructed. The colors in the map reflect the angle of rotation between the orientation of the phase at that point and a reference orientation, in this case the GaAs orientation measured at a point in the magenta region that corresponds to the underlying GaAs wafer. This angle is often referred to as the misorientation.

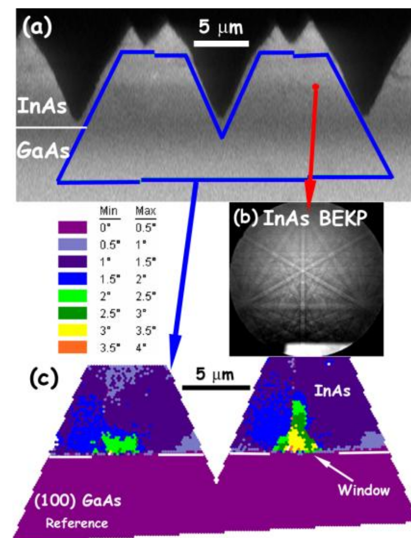

Fig. 2. (a) Secondary electron SEM micrograph of the area from which b and c were obtained. Sample was LEO InAs grown on a substrate with 5 µm wide windows. (b) A sample BEKP obtained from the InAs layer in spot mode. (c) Orientation image constructed from BEKPs for the area shown in a. Colors represent the magnitude of the local tilt relative to the orientation of the GaAs, the reference orientation for this image. White lines indicate the location of the mask material.

3. Results

The orientation images in Figs. 2-4 show the evolution from the tilted-grain microstructure characteristic of the InAs films grown on un-patterned substrates to a singular orientation as the LEO window width is reduced to below 1 μm. Fig. 3, from a film on an un-patterned wafer, shows that the multiple preferred orientations detected in x-ray rocking curves from this sample originate from columnar grains with mean diameters on the order of a micrometer. The image also suggests that the tilt is established at the substrate/ film interface and then propagates directly through the film. No tendency for evolution of the pattern or distribution of tilted material with thickness is suggested by the orientation image. TEM characterization of this film revealed the same microstructural features, albeit with considerably more demanding experimental methodology.

The orientation image in Fig. 2c was obtained from a sample with 5 μm wide windows and a 1:1 ratio of exposed GaAs to SiO_2 at the surface of the substrate. Three features distinguish this image from that obtained for the pattern-free wafer, Fig. 3. First, the magnitude of the InAs tilt relative to the GaAs substrate has decreased from ~4° for most grains on the pattern-free wafer (clearly evident when the GaAs is used as the reference orientation and from x-ray rocking curves, not shown here) to less than 2° in the bulk of the LEO material. Second, the angle of misorientation between the GaAs and the InAs in and directly above the centers of the windows is greater (~2-4°) than it is near and above the edges of the window and over the mask (≤2°). And third, the distribution of misorientations within a plane parallel to the substrate changes with distance from the substrate as the regions with the larger misorientations near the center of the window do not propagate through to the film surface.

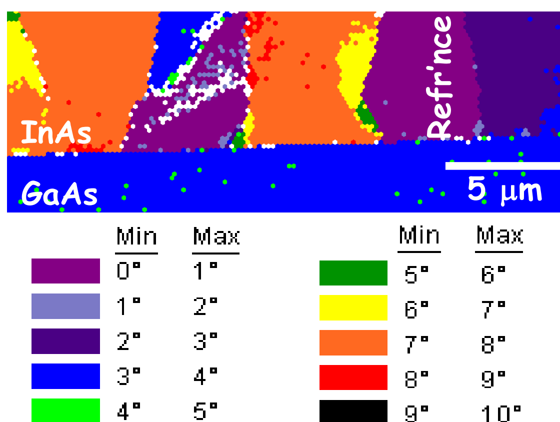

Fig. 3. Orientation image of polished cross-section of InAs grown on bare GaAs. Orientation values are relative to a point in the crystal labeled Refer'nce.

Fig. 4 shows the orientation image of a cross-section of LEO InAs grown on a substrate with 0.8 μm wide windows and an approximately 1:10 ratio of exposed GaAs to SiO_2 at the surface of the substrate. The InAs in the window, above the window, and above the mask *all* is oriented within 0.5° of the GaAs reference orientation. Singular orientation was indicated by x-ray rocking curves from this sample and as well as in a comparable sample for which the coalesced thickness was 3 μm, shown in Fig. 1b. Thus, although the InAs "bars" have not grown to impingement in the sample used for the BEKP study, we believe the results are representative of coalesced films due to the comparable x-ray results.

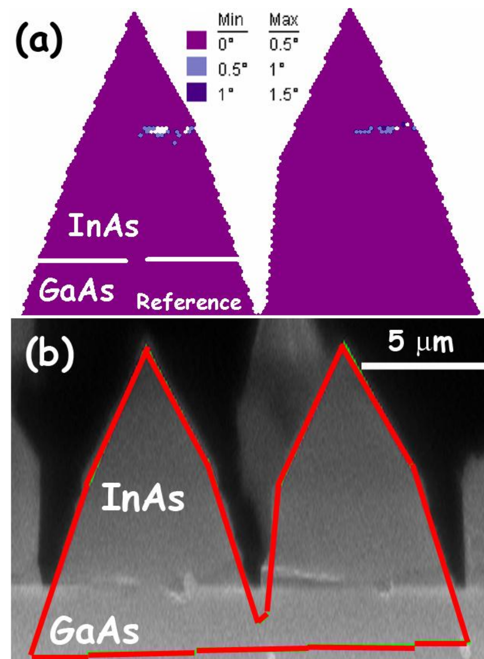

Fig. 4. (a) Orientation image for the LEO InAs grown on a substrate with 0.8 μm windows shown in (b). Note the singular orientation of the InAs and its parallel orientation to the GaAs, indicated by the monotone image. (b) Secondary electron SEM image with area used for (a) outlined.

Fig. 5a is a secondary-electron SEM micrograph of a 5 μm wide window in a LEO substrate upon which a small volume of InAs has been deposited and formed isolated islands. The wafer was tilted considerably (70°) relative to the imaging electron beam to the experimental geometry typically used to record BEKPs. The SEM image clearly reveals the propensity for islands to nucleate preferentially on the GaAs and along the edges of the mask. At this stage of growth, the island size at the mask edge is about a third of a micrometer, and the islands appear to have just coalesced. A few larger but sparse islands are seen in the center of the window.

Fig. 5b presents the results of BEKP orientation measurements made along the line shown in Fig. 5a (island row 1) and along a similar line in a neighboring window (island row 2). Each of these line scans includes approximately 300 orientation measurements from a section of the row containing about twenty islands. The

reference orientation is the measured orientation of the GaAs near the center of the window. Not only are the islands parallel to one another, but they are also essentially parallel to the GaAs. Furthermore, spot mode measurements of crystal orientation in the 0.5 μm islands in the center of the window suggested that they are crystallographically aligned with the GaAs as well. This behavior dramatically contrasts both the multiple orientations observed in larger (10 μm) sized islands on pattern-free substrates [6] and the orientation microstructure that developed in all of the other films with window widths larger than 1 μm.

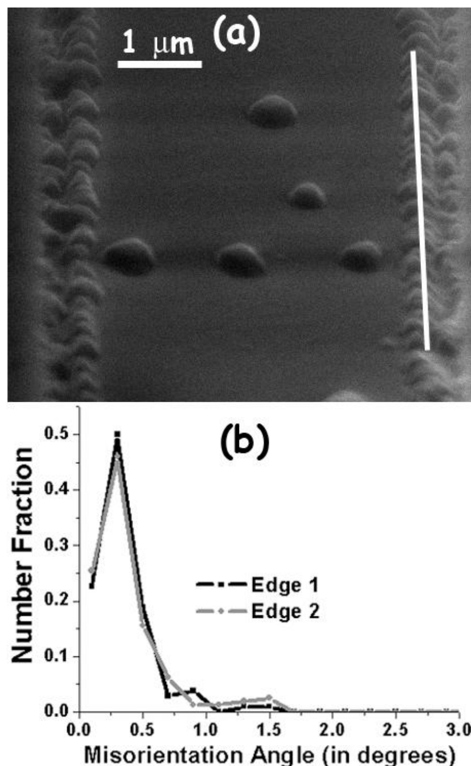

Fig. 5. (a) Secondary SEM micrograph of the distribution of InAs islands in the substrate window at an early stage in the growth process. (b) distribution of misorientation angles measured in two line-scans obtained from the islands located along the window edge, as indicated in a.

4. Discussion

The small islands at the edge of the window show singular orientation. Either the presence of the mask edge or the small size of the islands could be the underlying reason for the orientational purity at this stage and location of growth. The tendency for singular orientation to arise in films grown at lower temperatures and lower V/III ratios suggests that it is the island size at coalescence that is crucial, rather than the presence of the mask edge. Both of these conditions enhance the nucleation rate, and thus likely diminish the size of the islands at coalescence.

The islands along the window edge in Fig. 5 are approximately a third of a micrometer in diameter. Thus, if the window width were approximately two-thirds of a micrometer and the same growth pattern were to persist, at this stage of growth the window would be full of InAs with a singular orientation and a continuous film of singular orientation would be expected to as the deposition proceeded. In fact, precisely this result was obtained on windows of width 0.6 μm and 0.8 μm. The observation of singular orientation on the 0.8 μm but not for a 2 μm wide window suggests that the critical island size at which tilted domains form within an isolated island is about 0.5 μm. When the window width decreases at fixed stripe pitch, the nucleation rate is expected to increase as well as the increased SiO_2:GaAs surface area ratio will lead to increased supersaturation of the reacting species in the window region.

5. Conclusions

Control of the island size upon coalescence appears to be critical to establishing monolithic orientation in the InAs films grown on GaAs. Two effective routes to achieving this control are suggested by the present results when viewed in the context of the broader project of which they are a part. The first is to control the nucleation and growth kinetics, as could be done by utilizing a low temperature nucleation layer on an unmodified substrate. The second is to incorporate geometric constraints into the substrate, as is done, in conjunction with increasing the nucleation rate at the window edge, in the LEO process.

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