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Changing planar thin film growth into self-assembled island formation by adjusting experimental conditions

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Abstract

Illustrated in this paper are two examples of altering planar growth into self-assembled island formation by adapting experimental conditions. Partial oxidation, undersaturated solution and high temperature change Frank–Van der Merwe (FM) growth of $Al_{0.3}Ga_{0.7}As$ in liquid phase epitaxy (LPE) into isolated island deposition. Low growth speed, high temperature and in situ annealing in molecular beam epitaxy (MBE) cause the origination of InAs/GaAs quantum dots (QDs) to happen while the film is still below critical thickness in Stranski–Krastanow (SK) mode. Sample morphologies are characterized by scanning electron microscopy (SEM) or atomic force microscopy (AFM). It is suggested that such achievements are of value not only to fundamental researches but also to spheres of device applications as well.

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1. Introduction

The epitaxy modes of thin films can usually be categorized into three types: (a) Frank–Van der Merwe (FM) [1], (b) Stranski–Krastanow (SK) [2] and (c) Volmer–Weber (VW) [3]. These modes are deduced from equilibrium considerations of the surface and interface energies for lattice matched or mismatched systems. The FM mode is simply a layer-by-layer growth, and it usually occurs in lattice well-matched or slightly mismatched systems. In the former case, one may achieve high-quality flat films. In the latter case, films grow excellently until they reach a critical thickness, at which the accumulated elastic strain energy introduces the formation of dislocations. The situation of SK growth, however, is more complicated. In a moderately mismatched system, such as InAs/GaAs, initially, the deposition follows layer-by-layer mechanism which leads to perfect wetting of substrate. Subsequently, when the thickness of pseudomorphic film reaches a certain value d_1, the epitaxial layer is potentially ready to undergo a two-dimensional–three-dimensional (2D–3D) transition. While a further value d_2 (mentioned in literature as "critical thickness") is arrived at, the nucleation and growth of 3D islands will begin, and the system's total energy decreases sharply. Apparently, strain and strain-relaxation by the 3D island formation offer thermodynamic driving force for coherent SK growth. Finally, highly mismatched material combinations, then again, will form islands on the bare unwetted surface, crystallizing in the VW mode.

Modern epitaxial growth techniques, e.g., molecular beam epitaxy (MBE), can fabricate thin films to the atomic dimensional precision. However, in spheres like micro-electromechanical system (MEMS), molecular biology and nanometer micrology, there is also an urgent need to directly control the *lateral* dimension of microstructures and nano-

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structures. Hence, selective epitaxy, such as island crystal growth, gradually becomes a focus of attention. Self-organized island growth is particularly attractive, since many requirements for device manufacturing are satisfied, such as less process-induced defects, low cost and easy fabrication, as well as full compatibility with traditional device growth and processing techniques [4]. These self-assembled islands can be accomplished with great skill via SK [5] or VW [6] growth of epilayers, as elucidated above. However, little attention has been concentrated on *changing* planar thin film growth into self-assembled island growth by properly adjusting experimental conditions. In this paper, we show that SK film growth, with the thickness still being less than critical value d_2, and FM layer-by-layer homogeneous epitaxy can both be evolved into self-assembled island formation under certain conditions. Systematic disquisition of altering the epitaxy which ought to have deposited in layer-by-layer manner into self-organized island growth is too complex a topic. In the following discussions, two typical examples are experimentally studied: (a) liquid phase homoepitaxy of $Al_{0.3}Ga_{0.7}As$ islands (FM); (b) molecular beam heteroepitaxy of InAs/GaAs dots (SK).

2. Changing planar growth in FM modus

Generally speaking, during liquid phase epitaxy (LPE) progress on a smooth substrate, the formation of a new monolayer (ML) begins as soon as the monolayer beneath it covers the surface. A flat thin film therefore can be obtained in the FM modus. Nevertheless, in this section, we will focus on micron-sized AlAs–GaAs alloy islands formed by LPE under conditions of partial oxidation, undersaturated solution and high temperature. That research is of great interest from not only a fundamental point of view, but a technological one as well. For instance, in the promising scheme of a certain kind of integrated scanning near-field optical microscopy (SNOM) sensors manufacturing, the fabrication of GaAs microtips plays a very critical part [7]. Although the wet etching process can also yield good quality microtips, it is hard to fulfill monolithic integration of microtips on top of other optical elements because of technological difficulties [7]. Akiyama et al. [8] suggest that one can fabricate microtips on an independent substrate and transfer them onto the target wafer with the tips protected by a resist layer, but that technique seems to be too tedious. In our case, the pyramid-like probe can be conveniently grown onto the facet of the optical element by LPE, thus, there is no problem of the monolithic integration. The reason we choose $Al_{0.3}Ga_{0.7}As$ as test material, however, lies in its easier oxidation and the higher stability of the oxides.

The $Al_{0.3}Ga_{0.7}As$ patterns are created by employing a universal LPE system. The substrate is a 400-μm-thick intrinsic (100) GaAs wafer. After organic abstersion, it is chemically treated in a sulphuric acid cleaning system ($H_2SO_4/H_2O_2/H_2O$=6:1:1). Subsequently, it is washed by deionized water, dried by inert gas, and finally coated with a flat $Al_{0.3}Ga_{0.7}As$ epitaxial layer which serves as a transitive stratum. That newly prepared layer is exposed in extrapure air for 20 min. In order to grow $Al_{0.3}Ga_{0.7}As$ islands, the growth solution should be composed of 500 mg Ga, 25 mg GaAs, 0.6 mg Al and 8.5 mg Ge. In the reducing atmosphere of H_2, a 200-μm-thick layer of growth solution is smeared onto the $Al_{0.3}Ga_{0.7}As$ surface (the temperatures of different samples range from 800 to 810 °C) by a graphite scraper. Afterwards, the wafer temperature is decreased at a rate of 1–1.5 °C min^{-1}. Finally, the $Al_{0.3}Ga_{0.7}As$ hills are turned out when the samples are cooled down to room temperature and scanning electron microscope (SEM) is employed to study their surface conformations.

Under our conditions, 20 min of exposure of the $Al_{0.3}Ga_{0.7}As$ epilayer in the open air results in partial oxidation and many micrometer-sized windows are created due to the chemical property of the oxides. A similar $Al_xGa_{1-x}As$ native oxide matrix is observed by Ghita et al. [9]. Ideal positions for homogeneous nucleation are thus offered by those tiny oxide-free windows. Once stuck onto the windows, clusters of adsorbed particles will act as growth nuclei and grow further by the gathering of migrating particles, becoming large islands consequently by the direction-dependent arrangement of atoms. Nevertheless, the abortion of island growth may occur if the hills are fed by supersaturated sources, for they may overgrow onto the oxides, joint to each other, and finally fuse into a plane. The circumstances of inadequate growth sources should be necessarily produced by maintaining only a skinny layer of undersaturated gallium solution on the $Al_{0.3}Ga_{0.7}As$ surface. It should be pointed out that there is still a small probability for the sources to nucleate on the oxides, generating poor-quality poly-crystalline deposits [10]. Growth at relatively higher temperatures can result in the rapid surface diffusion of materials across the oxides to an opening and favor the desorption of extraneous growth materials from the oxides [11]. Therefore, in our case, high temperature (800–810 °C) has urged self-organized islands

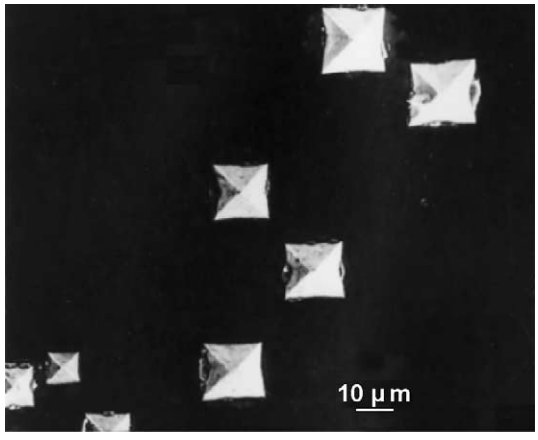

Fig. 1. Several of as-prepared $Al_{0.3}Ga_{0.7}As$ pyramids.