East Asian Journal on Applied Mathematics
doi: 10.4208/eajam.040411.030611a

Vol. **1**, No. 4, pp. 297-371
November 2011

Review Article

Interface Dynamics and Far-From-Equilibrium Phase Transitions in Multilayer Epitaxial Growth and Erosion on Crystal Surfaces: Continuum Theory Insights

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Received 4 April 2011; Accepted (in revised version) 3 June 2011

Available online 23 September 2011

Abstract. We review recent theoretical progress in the physical understanding of far-from-equilibrium phenomena seen experimentally in epitaxial growth and erosion on crystal surfaces. The formation and dynamics of various interface structures (pyramids, ripples, etc.), and also kinetic phase transitions observed between these structures, can all be understood within a simple continuum model based on the mass conservation law and respecting the symmetries of the growing crystal surface. In particular, theoretical predictions and experimental results are compared for (001), (110) and (111) crystal surfaces.

PACS (2006): 81.15.Aa, 89.75.Kd, 05.90.+m

Key words: Interface dynamics, epitaxial growth, non-equilibrium phase transitions.

Contents

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1. Introduction

There is extensive research interest in the dynamics of the surfaces of thin films undergoing molecular beam epitaxy (MBE) growth. The MBE technique is considered the best method for the growth of thin solid films, of great importance in applied and experimental studies [1]. It is a unique way to grow high quality crystalline materials; and in particular, to form structures with very high precision in the vertical direction perpendicular to the substrate, such as monolayer-thin interfaces or atomically flat surfaces. However, multilayer epitaxial growth may exhibit complex surface features that cannot easily be controlled experimentally. In epitaxial multilayer growth regimes, surface morphology dynamics involves a subtle interplay between the depositing molecular flux and the relaxation of the surface profile through the surface diffusion of adatoms. Of paramount significance are adatom interactions with steps that form on the multilayered surface, particularly the energy barriers occurring near the step edges that inhibit adatom transitions between layers of the growing interface. These so-called step edge barriers were found by Ehrlich and Hudda [2], and further elucidated by Schwoebel [3] and Villain [4]. Ehrlich-Schwoebel-Villain (ESV) energy barriers produce up-hill adatom surface currents, from the terrace of the lower layer towards the terrace of the upper layer. The ESV effect produces instabilities in the evolution of the surface morphology, which ultimately lead to the formation of fascinating structures such as mounds and pyramids across the growing interface [5, 6]. The ESV instability is a non-equilibrium effect, which is present only if the adatom density on a terrace is higher than in equilibrium. The deposition process by molecular beams raises the adatom density well above its equilibrium value, and surface currents are generated that depend on the local slope of the growing film. From a study of the diffusive dynamics of adatoms on vicinal surfaces with step edge barriers, Villain [4] found that the surface non-equilibrium current $\mathbf{J}^{\rm NE}$ has the same direction as the slope, and consequently tends to increase the local slope of the interface. However, once the interface attains sufficiently large slopes, other processes that counterbalance the destabilizing ESV effect also become significant, so the net current in *non-equilibrium* states vanishes for certain slopes (slope selection). Indeed, funneling and knockout processes [7, 8] can lead to zeros of $\mathbf{J}^{\rm NE}(M)$, even for small values of the interface slope M [9, 10]. Due to slope selection, the interface restructures into facets with pyramid-like objects. The slopes of these facets correspond to stable zeros of the non-equilibrium surface current.

Many experiments have shown that the surface evolution of films grown by MBE frequently involves the formation of pyramids or pyramid-like structures, even for homoepitaxial growth. Pyramid-like structures have often been observed — e.g. on the homoepitaxy of GaAs [6, 11], Cu [5, 44], Ge [12] and Fe [13, 14], all grown on singular (001) substrates, and on the Rh(111) surface [15]. The lateral size λ and height w of these pyramids grow in time as power laws with the *same* exponent. Thus the ratio w/λ corresponding to the pyramid slope eventually approaches a constant value, so there is