

WHY DOES A THICK FILM DE-WET?

Kevin F. McCarty¹, John Hamilton¹, Angela Saá², Yu Sato¹, Juan de la Figuera^{2,3},
A.K. Schmid⁴, Konrad Thürmer¹, and Norman C. Bartelt¹

¹Sandia National Laboratories, Livermore, CA 9455, USA

²Centro de Microanálisis de Materiales, Universidad Autónoma de Madrid, Madrid 28049,
Spain

³Instituto de Química-Física Rocasolano, CSIC, Madrid 28006, Spain

⁴Lawrence Berkeley National Laboratory, Berkeley CA 94720, USA

Juan.delafiguera@iqfr.csic.es

Growing a flat, uniform film of a material that has a substantial lattice mismatch with a given substrate remains a complicated problem. To start with, many film/substrate systems are unstable if the film uniformly covers the substrate – the film will tend to break apart into discrete, 3D islands. The energetic driving force for forming 3D islands typically arises because the surface energy of the substrate is less than the sum of the film/substrate interface energy and the film's surface energy. Even in systems that will eventually de-wet and form 3D islands, low-temperature deposition can yield relatively flat films. This approach for producing uniform films, which many technologies depend upon, works if the kinetic processes that de-wet the film are practically slow. Since the atomic processes that convert a flat film into 3D islands are poorly known, predicting the time needed for film de-wetting is currently not possible. Well-defined 3D structures, such as dots and wires, on surfaces also have great technological potential. Hence, research has been devoted to determining whether de-wetting is a viable route to synthesizing self-organized nanostructures[1-4]. To understand the competition between smoothing and film de-wetting, we perform real-time observations by low energy electron microscopy (LEEM).

Here we reveal a process by which a continuous film simultaneously thins to expose the wetting layer in local regions and thickens in adjacent regions, eventually forming 3D islands. We study the system of single-crystal Cr films on a W(110) substrate [5], a system free of threading dislocations. Cr films grown near room temperature are rough. When annealed, the films first smooth and can uniformly cover the substrate. With further annealing, the films de-wet. Because of this system's large anisotropy, the trenches that expose the stable wetting layer only form along one crystallographic direction, the W [001] direction. The Cr islands formed by de-wetting are arrays of 3D, unidirectional stripes. Fe also forms stripes when de-wetting W(110) [6,7]. This directionality allows the nucleation of individual trenches to be observed with sufficient temporal and spatial resolution to see the motion of individual atomic film steps.

The 3D islands shape of unidirectional stripes greatly simplifies observing the de-wetting mechanism. Our microscopic observations reveal that de-wetting starts at points of the surface with a high density of atomic steps. We observe that a section of an atomic film step advances (downhill) relative to the substrate steps, making this local region thicker, and, therefore, more stable than surrounding regions. This thicker region then draws material from adjacent areas where steps retract (uphill). These instabilities grow and eventually form trenches that reach down to the wetting layer. With time, the Cr stripes between the trenches become continuously narrower and thicker. Eventually, the Cr film is converted into reasonably well-organized patterns of Cr stripes as the trenches that expose the wetting layer lengthen along the W[001] direction (see Figure).

The thick Cr films are largely not strained by the substrate as detected by low energy electron diffraction (LEED). Instead, we propose that surface stress provides the driving force for the observed morphological instability. Atomistic simulation and analytic elastic models show that surface stress can cause the dependence of film energy on thickness required for simultaneous thinning and thickening. We observe that de-wetting is also initiated at bunches of substrate steps in two other systems, Ag/W(110) and Ag/Ru(0001). Thus, we expect that the observed de-wetting mechanism of cooperative motion of film steps relative to substrate steps is general.

This research was supported by the Office of Basic Energy Sciences, Division of Materials Sciences, U.S. Department of Energy under Contract No. DE-AC04-94AL85000, by the Spanish Ministry of Science and Technology through Project No. MAT2006-13149-C02-02 and by the Comunidad Autónoma de Madrid and the CSIC through Project No. CCG07-CSIC/MAT-2030.

References:

- [1] Y. Ishikawa, Y. Imai, H. Ikeda and M. Tabe, *Appl. Phys. Lett.* **83** (2003) 3162.
- [2] A. L. Giermann and C. V. Thompson, *Appl. Phys. Lett.* **86** (2005) 121903.
- [3] K. F. McCarty, *Nano Letters* **6** (2006) 858.
- [4] P. Sutter, W. Ernst, Y. S. Choi and E. Sutter, *Appl. Phys. Lett.* **88** (2006) 141924.
- [5] P. J. Berlowitz and N. D. Shinn, *Surf. Sci.* **209** (1989) 345.
- [6] R. Zdyb, A. Pavlovska, M. Jalochofski, E. Bauer, *Surf. Sci.* **600** (2006) 1586.
- [7] N. Rougemaille and A. K. Schmid, *J. Appl. Phys.* **99** (2006) 8S502.

Figures:

Figure 1. Successive low energy electron images acquired while annealing a Cr/W film at 620°C.

