## The role of long-range interactions and critical behavior in determining surface morphologies: a combined LEEM/SXRD study

Raoul van Gastel

University of Twente, MESA+ Institute for Nanotechnology, P.O. Box 217, 7500AE Enschede, The Netherlands

In this presentation I will detail how long-ranged elastic interactions that occur in metal heteroepitaxy can be used to achieve shaping and sizing of nanostructures. Pb/Cu(111) is first used as an example to illustrate how Low Energy Electron Microscopy (LEEM) can be applied to quantify the thermodynamic parameters that are driving the self-assembly of two phases, a surface alloy phase and an overlayer phase [1-3], illustrated in figure 1. We show how those parameters can be manipulated to induce size [3] and shape [4] changes of individual domains.

For the similar Bi/Cu(111) system we have combined surface X-ray diffraction (SXRD) and LEEM to investigate the atomic structure and pattern formation. Although it might appear identical to Pb/Cu(111) at first sight, critical behavior of the Bi surface alloy leads to dramatic changes in the morphology of the deposited Bi layer as can be seen in figure 2. Using a combination of SXRD and LEEM measurements we investigate the dynamics and transitions of the different surface phases [5], and pinpoint the origin of the morphological changes to the atomic structure of the different phases. A rich variety of surface phases, covering the whole spectrum, from solid to liquid to lattice-gas-like and from ordered to disordered, is observed. In LEEM movies, structural contrast between e.g. solid and liquid Bi overlayers lets us image the phase transitions directly in real space.

The pattern formation and the dramatic changes that occur during the transitions are again analyzed to extract information and quantify the thermodynamic parameters that control the rich phase behavior in this system. In the particular case of the morphological changes shown in figure 2, we employ the hard hexagon model [6] to interpret the nature of the phase transition and the changes in the thermodynamic parameters. In our analysis we find that the Bi/Cu(111) system exhibits near-perfect hard hexagon behavior and the predicted value for the hard-hexagon critical coverage is recovered from our experiments.

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Figure 1: Surface domain self assembly evolution during Pb deposition on Cu(111). LEEM brightfield micrographs of the Cu(111) surface with a) a 2D-droplet, b) a striped, and c) an inverted 2D-droplet pattern. Images were taken at an electron energy of 18 Volts where the overlayer phase appears bright and the surface alloy phase appears dark. (Field of view is 1.8 μm.)



Figure 2: A small change in temperature induces significant morphological changes in the Bi/Cu(111) overlayer morphology. The Bi overlayer phase is imaged bright and the alloy phase dark. Images were recorded at a temperature of 680 K, field of view is 4.0 μm.