Materials Science and Technology
Ultra-thin Films

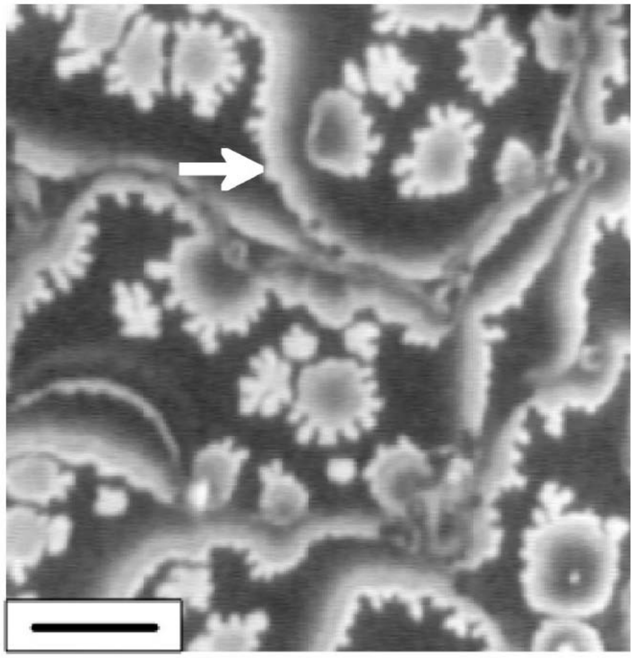
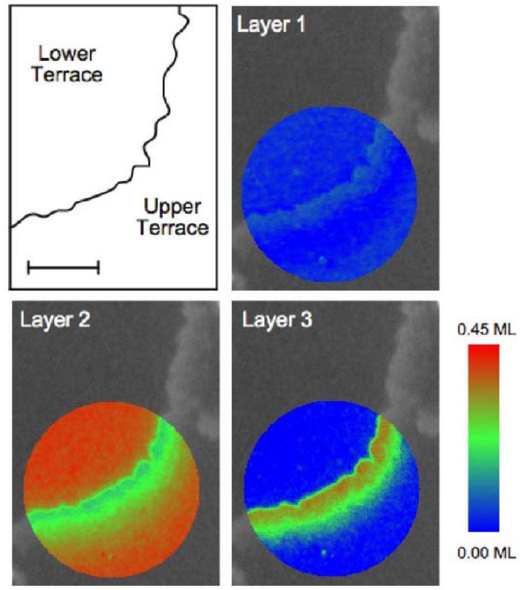


Figure 1: (Left) Low energy electron microscope (LEEM) image of a Cu(001) surface recorded after the deposition of ~0.6 ML Pd at 200ºC. The white arrow points to a surface step. The continuous variation in intensity away from the step and its dendritic shape indicates highly non-uniform growth. (Electron energy = 13.1 eV; Scale bar = 1 μm).

Figure 2: (Right) Three-dimensional, color-coded map of the Pd concentration near a surface step. The images were constructed from the current-voltage (IV) analysis of 17,665 individual pixels. The maps are superimposed on the corresponding LEEM image at 13.1 eV. There is essentially no Pd in the top layer, maximum concentration in the second layer away from the steps, and a high concentration in the third layer near the steps. (Scale bar = 500 nm.)



Identifying the Origins of Heterogeneity in Ultra-thin Films

New technique provides three-dimensional maps of surface chemical composition.

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Thin films play a key role in many technologies. Applications range from promoting chemical reactions at surfaces for improved sensor, catalytic, or anti-corrosion properties to more exotic ones in nanotechnology, where reduced dimensionality gives rise to unique electronic or magnetic properties. It is well known that the mechanical, electrical, and catalytic properties of thin films are intimately related to structure and composition. However, understanding this relationship in detail is challenging because thin films are often inhomogeneous, especially at the nanoscale. Historically, measuring structural and compositional heterogeneity at these small length scales has proven difficult because high spatial resolution must be combined with sub-surface chemical sensitivity.

In collaboration with scientists at IBM, Yorktown Heights, and the University of New Hampshire, Sandia researchers have developed a technique to determine the three-dimensional (3-D) composition profile of a surface with 10-nm lateral resolution and single atomic-layer depth sensitivity. The technique combines the high-spatial resolution of the low-energy electron microscope (LEEM) with the chemical sensitivity of quantitative electron reflectivity measurements. Using this technique, we have investigated how heterogeneity develops during the growth of ultra-thin Pd films on Cu(001). Such films have been identified as potential inhibitors to the undesired effect of Cu electromigration in microelectronic devices, but little is known about the initial stages of film growth. Figure 1 shows a LEEM image of a Cu(001) surface recorded during Pd deposition at 200ºC. The spatial variation of the intensity shows directly that the surface is heterogeneous, but to determine how and why the inhomogeneity occurs requires detailed information on the distribution of Pd in the atomic layers just below the surface. From our analysis, we obtain color-coded maps (Figure 2) showing how the Pd concentration varies in the vicinity of a surface step. From the time-evolution of such 3-D compositional profiles, we discovered that

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the heterogeneity observed in Figure 1 can be explained with a conceptually simple "step-overgrowth" model (Figure 3) in which step flow, caused by movement of the Cu atoms, converts mobile Pd in the second layer into fixed Pd in the third layer.

These results highlight the important role of surface steps in surface alloy formation and demonstrate how our new technique can give powerful insight into complex morphological evolution at surfaces.

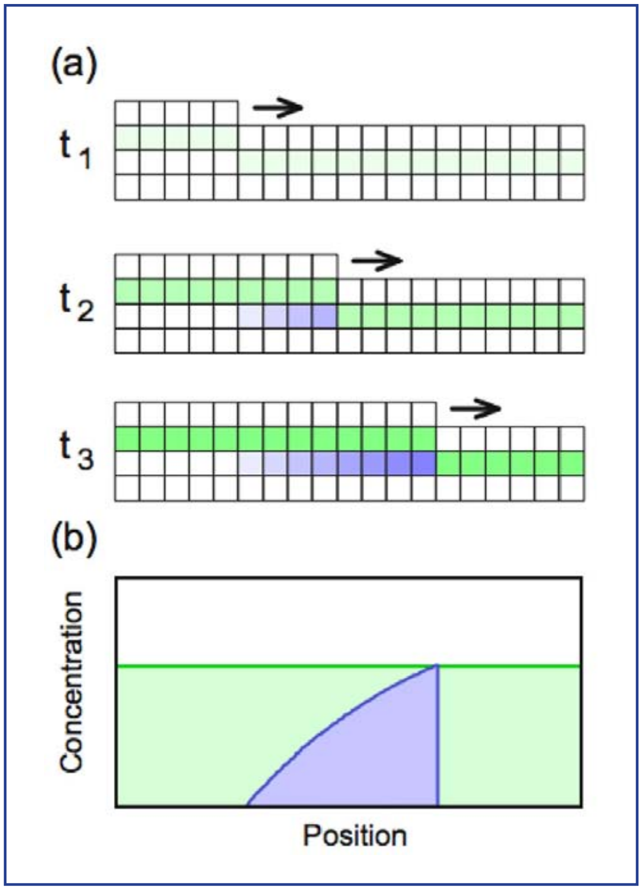

Figure 3: Schematic drawing illustrating how heterogeneity arises during step-flow overgrowth for Pd on Cu(001). (a) Side views of the Cu surface. The Pd composition in the second layer is shown in green, in the third layer blue. Step flow overgrowth converts mobile Pd in the second layer into fixed Pd in the third layer. (b) Spatial dependence of the Pd concentration in the second (green) and third (blue) layers at the end of growth.

Publications:

J. B. Hannon, J. Sun, K. Pohl, and G. L. Kellogg, " Origins of Nanoscale Heterogeneity in Ultra-thin Films," *Phys. Rev. Lett.* 96, 246103 (2006).

J. Sun, J. B. Hannon, G. L. Kellogg, and K. Pohl, " Local Structural and Compositional Determination via Electron Scattering: Heterogeneous Cu(001)-Pd Surface Alloy," *Phys. Rev.* B 76, 205414 (2007).

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