Identifying the Origins of Heterogeneity in Ultra-thin Films by LEEM-IV Analysis

by **Gary Kellogg**

Motivation—Surface alloy formation plays a key role in many technologies. An application of current interest in the microelectronics industry is the potential use of surface alloys as a means to reduce the detrimental effect of electromigration on Cu interconnects. This process is mediated by surface diffusion, and the search is on for coating metals that reduce surface diffusion but do not increase bulk resistance – i.e., metals that do not alloy. Pd films have been shown to reduce electromigration, but since Pd and Cu are miscible, bulk alloying is a potential problem. Consequently, understanding how, and at what temperature, Pd intermixes with Cu is of considerable interest.

Accomplishment—Our low energy electron microscope (LEEM) images show clearly that the growth of Pd on Cu(001) is *not* uniform (Fig. 1). The surface contains a number of single-atomic layer steps, identified by sharp changes in image contrast. In these regions, one sees dendritic-type growth outward from the advancing step and a continuous decrease in imaging intensity in the opposite direction towards the original step position. Precisely how the surface is inhomogeneous and the extent of the intermixing during growth, however, cannot be determined from the images alone. The key information needed to understand the heterogeneity that develops during surface alloying is a quantitative measure of the spatial and depth distribution of the deposited Pd. To obtain this information, we have developed a new LEEM-based technique to determine the three-dimensional composition profile of a surface with 8-nm spatial resolution. In our "LEEM-IV" analysis, we measure reflected electron intensity vs. incident energy curves (so-called "IV" curves) pixel-by-pixel for the entire LEEM image. We then analyze the reflectivity data using multiple-scattering low energy electron diffraction (LEED) calculations (in analogy with conventional LEED-IV analysis). With this new technique, we have measured the spatial and depth distribution of Pd on Cu(001) as a function of deposition time. Color-coded maps indicating how the Pd concentration varies in the vicinity of a surface step are shown in Fig. 2a. From the time-evolution of such 3-d compositional profiles, we show that the heterogeneity observed in Fig. 1 can be explained with a conceptually simple "step-overgrowth" model (Fig. 2b) in which step flow converts mobile Pd in the second layer into fixed Pd in the third layer.

Significance—Understanding how chemical heterogeneity develops at surfaces is a key challenge in the engineering of thin-films. By carefully mapping the three-dimensional compositional profile of Pd near surface steps, we have identified the exact process – step overgrowth coupled with inhibited bulk diffusion – that gives rise to heterogeneity in Pd/Cu(001) surface alloys. Although the model was developed specifically for Pd/Cu(001), step overgrowth requires only fast surface diffusion and relatively slow bulk diffusion. This state of affairs is quite common in thin-film growth, and the same mechanism should be operative generally.

Sponsors for various phases of this work include: DOE Office of Basic Energy Sciences/Center for Integrated Nanotechnologies (CINT)

Contact: Gary L. Kellogg, Surface & Interface Sciences, Dept. 1114
Phone: (505) 844-2079, Fax: (505) 844-5470, E-mail: glkello@sandia.gov

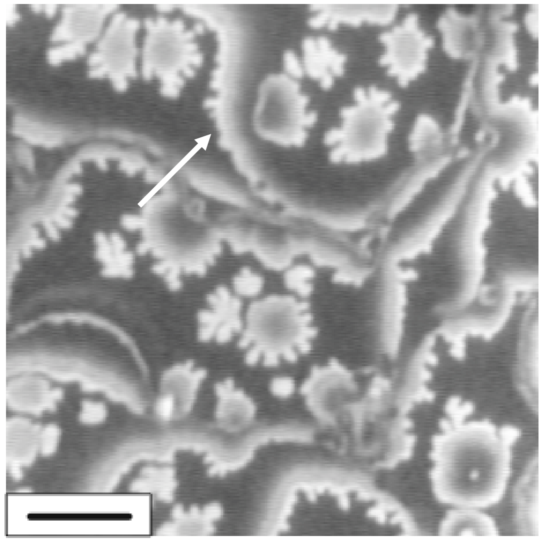
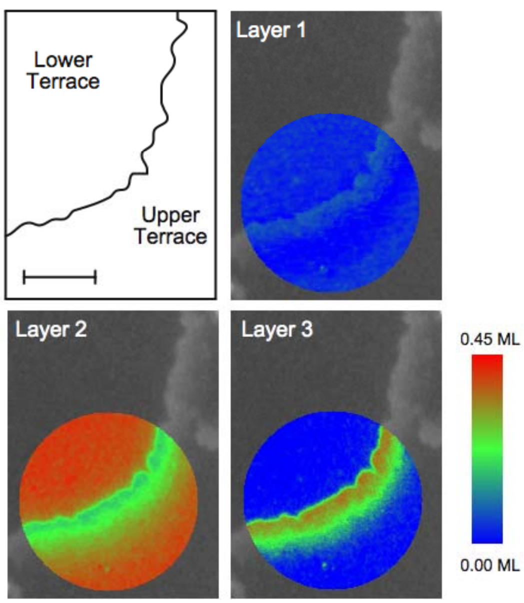
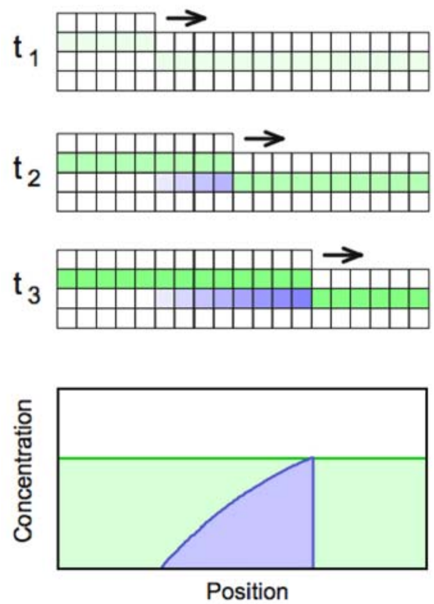


Figure 1. LEEM image (electron energy = 13.1 eV) recorded after the deposition of ~0.6 ML Pd on Cu(001) at 200 ºC. The gradual decrease in reflected electron intensity moving away from surface steps (white arrow) is unusual as is the dendritic-like growth along the steps. Scale bar = 1 μm.



(a)



(b)

Figure 2. (a) Three-dimensional maps of the Pd concentration near a surface step. The images were constructed from the analysis of 17,665 individual pixels. The maps are superimposed on the corresponding LEEM image at 13.1 eV. Scale bar = 500 nm. (b) Schematic illustrating how heterogeneity arises during step-flow overgrowth. (top) 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. (bottom) Spatial dependence of the Pd concentration in the second (green) and third (blue) layers at the end of growth.