Bridging the Nano-Micro Length Scale in Surface Science

by ***M. L. Anderson, N. C. Bartelt, B. S. Swartzentruber, P. J. Feibelman and G. L. Kellogg***

Motivation—The ability to control the nanometer-scale structure of solids requires a fundamental understanding of physical phenomena occurring over multiple length scales. This is especially true for processes at surfaces, where often a delicate interplay between atomic-scale kinetics and macro-scale thermodynamics leads to stable nanoscale structures. To understand this interplay, experimental tools and modeling approaches that probe *both* the behavior of individual atoms as they migrate across surfaces *and* the collective motion of thousands of atoms as they organize into the desired surface structures are required.

Accomplishment—We are the first to combine scanning tunneling microscopy (STM) and low energy electron microscopy (LEEM) to understand the atomistic mechanisms controlling the 10-1000 nanometer-scale structure of a surface as it spontaneously self-assembles. The STM with its single-atom resolution can image surface features with dimensions of Angströms to tens of nanometers. The LEEM with a lateral resolution of 7-8 nanometers can image features from tens of nanometers to tens of microns. Together, the two instruments can view surface features spanning dimensions covering six orders of magnitude. The phenomenon that we are investigating is a particularly dramatic example of surface self-assembly—the spontaneous ordering of two-dimensional Pb islands on Cu(111) into stable patterns with submicron periodicities. In addition to a proper balance of thermodynamic forces, pattern formation in this system requires that two-dimensional islands containing tens of thousands of atoms have sufficient mobility to move across the surface and find their equilibrium positions. To understand the atomistics of this large-scale, collective motion, we are investigating the thermal decay (shrinking) of two-dimensional Pb overlayer and Pb-Cu alloy islands (Fig. 1). By applying both STM and LEEM to the same surface process we are able to measure decay rates covering 6-7 orders of magnitude (2-3 orders of magnitude are typical) and show that the mechanisms and energetics of mass transport remain the same from atomic to micron length scales (Fig. 2). From the temperature dependence of the decay rates, we determine that it is the migration of just one atomic species (Cu) that controls the overall kinetics of island decay. Combined with LEEM studies of Cu deposition on Pb overlayers and first-principles calculations of the formation energy for vacancies and interstitials within the Pb overlayer, these studies are providing a self-consistent picture of how two-dimensional islands, approaching 100 nanometers in diameter, move intact across Pb-Cu alloy surfaces.

Significance—A comprehensive understanding of pattern formation on surfaces requires quantitative information on both the thermodynamic forces that drive self-assembly and the kinetic processes that allow the patterns to evolve. Here, we find that the surprisingly large mobility of large two-dimensional islands (key to pattern evolution) is the result of mobile defects within the Pb overlayer—a totally unanticipated discovery. The new understanding is the result of our ability combine complementary surface microscopies and bridge length scales from atoms to microns.

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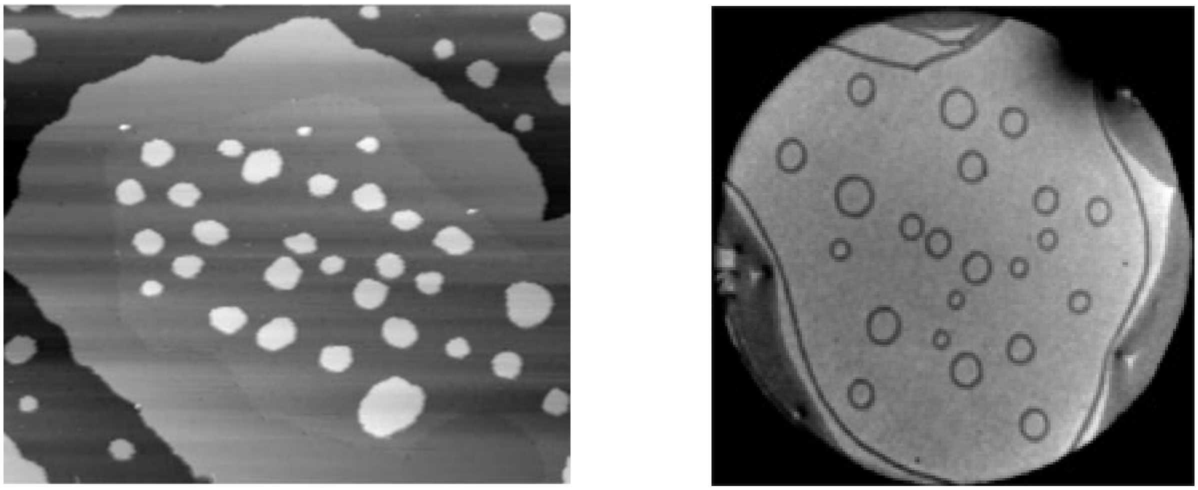
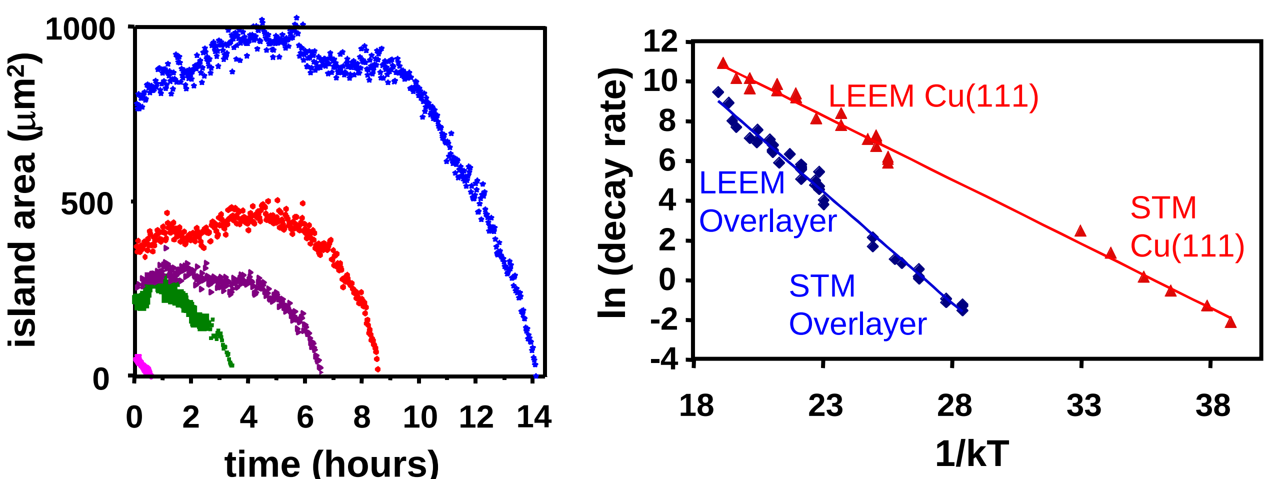
Contact: Gary L. Kellogg; Surface & Interface Sciences, Dept. 1114
Phone: (505) 844-2079, Fax: (505) 844-1197, E-mail: glkello@sandia.gov



(a) STM (260 nm field of view) (b) LEEM (3 micron field of view)

Figure 1. Images of Pb overlayer islands on Pb-Cu surface alloy [(a) STM image and (b) LEEM image]. Note that the field of view in the two images differs by more than an order of magnitude. In both cases, the islands decay (shrink) as a function of time. Measurements of the decay rates probe the mechanisms and energetics of atom diffusion.



(a) (b)

Figure 2. (a) Decay curves for selected Pb overlayer islands on a Pb-Cu surface alloy. (b) Arrhenius plots of island decay rates for pure Cu islands on Cu(111) and Pb overlayer islands on Pb-Cu surface alloy. Note that the data fall on a single straight line for the LEEM and STM measurements. All data are from this work except STM measurements of pure Cu islands, which are from: G.S. Icking-Konert, M. Giesen, H. Ibach, Surf. Sci. 398, 37 (1998).