

Thin film technology has greatly advanced over the last few decades and plays an important role in advancing electronic and spintronic-based technology. On the fundamental side, efforts have been focused on controlling the formation of novel nanostructures through manipulation of strain energy, adsorbate-modified kinetics, light irradiation, etc. [1-5] For ultra-thin metal epitaxy, emerging research over the last decade has demonstrated that quantum confinement of electronic states can have profound effects on the growth of various metal nanostructures as well as their physical and chemical properties [6-13]. This is commonly referred to as “quantum” or “electronic growth [14].”

Among various systems exhibiting quantum growth behavior, Pb on Si(111) is probably the most thoroughly investigated. This is due to a nearly 4:3 matching between the (111) lattice constant and the Fermi wavelength (λ_F), resulting in a bi-layer quantum oscillation of the density of states [15, 16]. This thickness-dependent modulation of the quantum well states (QWS) has been utilized to study the interplay between quantum confinement and surface diffusion of Fe adatoms as well as QSE-induced oscillations of surface reactivity for adsorbed O atoms [17, 18]. However, for these specific cases, there is no experimental evidence showing that adsorbates induce modifications to the underlying quantum growth phenomena. Furthermore, this thickness-dependent modulation, where the interplay between classical surface energy (which favors a smooth (111) surface) and the quantum size effect (QSE) energy (which favors bilayer stability), gives rise to rich phenomena in atomic processes of film growth [19-22].

I am interested in studying how adsorbates modify the overall energy landscape of quantum thin films. As has been demonstrated on metal surfaces, adsorbates can influence the formation of novel nanostructures. However, there are no experiments that investigate this effect when QSE are strong. How do adsorbates modify the surface energy when QSE are present? For the case of Pb films, will the bi-layer oscillation in surface energy be altered by impurities? Using Scanning Tunneling Microscopy/Spectroscopy (STM/STS), I will utilize atomic scale imaging to characterize how dilute levels of impurities affect the morphology of the film and the local electronic density of states. Furthermore, STM can reveal how different adsorbates will distribute on the surface. Is there preferential thickness-dependent adsorption resulting from the quantum well state modulation?

I plan on using Cs, Oxygen, Au, and Xe adsorbates. The first two elements have tendencies to donate/accept electrons. This should result in different preferential binding to specific layer thicknesses (as demonstrated for O). I want to confirm this effect and see if changing to a typical donor, like Cs, can alter the preferential binding. Au is a common transition metal that shows a tendency to alloy with Pb. This would be an interesting case since it might be possible that Au does not prefer to sit on the Pb surface but sit inside the Pb film somewhere. Xe is a noble gas and therefore should not have a strong tendency to donate/accept electrons. Thus, with these 4 adsorbates, if adsorbate-adsorbate interactions are small, there is a nice spectrum of possibilities.

STM experiments alone cannot reveal the binding properties of these adsorbate species. I have begun a collaboration with the Materials theory group at Oak Ridge National Laboratory (ORNL) with Professor Zhenyu Zhang. For my project, I will perform an internship at ORNL where I will receive hands on training with theoretical modeling/calculation based on first-principles techniques. The purpose of the collaboration will be to model the binding properties of different adsorbates on these Pb films in conjunction with experiment. For example, what is preferential binding site of a specific adsorbate (on the surface or inside the film)? Do adsorbates preferentially bind to stable or unstable films? Are there strong adsorbate-adsorbate interactions that might lead to clustering? How do adsorbates modify the formation energy of quantum stable or unstable structures? Do quantum stable structures become more energetically favorable with the introduction of adsorbates? This collaboration provides a nice way answer an important scientific question utilizing an interplay of both theory and experiment.

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