Uncovering thin film growth dynamics from in situ and real-time diagnostics

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Abstract:

The understanding of morphological and microstructural development during thin film growth is of particular relevance to control islands shape, nucleation and growth of nanoparticles, phase transformation, texture or surface roughness. Due to oversaturated vapour fluxes employed in physical vapor deposition (PVD) techniques, dynamics usually prevails over thermodynamics in dictating growth and microstructural evolution in PVD films. Depending on the material mobility, different growth modes occur, driven by kinetically limited surface diffusion processes and/or interfacial reactions. This has important implications in areas where nanoscale materials are involved, such as plasmonics, spintronics or next-generation of nano-devices.

In this presentation, we will provide some examples of *in situ* and real-time diagnostics based on optical techniques (wafer curvature, surface differential reflectance spectroscopy) and electrical resistance measurements to probe with high sensitivity the early growth stages of a variety of metal films on Si during sputter-deposition. In particular, we will show by coupling simultaneously two optical sensing techniques that the tensile stress generated upon island coalescence exhibit a maximum value at a film thickness that coincides with the onset of film continuity, for films growing in a Volmer-Weber mode (Ag, Au, Cu, Pd). The tensile stress magnitude is found to increase with decreasing material mobility, in relation with a lower percolation threshold, revealed from in situ resistivity measurement.

For films with lower adatom mobility (e.g., Mo, W), interfacial reaction with silicon favors a 2D growth mode and the initial formation of an amorphous film, followed by a phase transition to the equilibrium bcc phase above a critical thickness. In the case of Mo and Fe films, our *in situ* measurements reveal a structural transition at a film thickness of ~ 2-3 nm manifested by a concomitant tensile stress variation and decrease in electrical resistance. Insights on the kinetics of the amorphous-to-crystalline phase transformation were gained from *in situ* synchrotron studies, coupling simultaneously X-ray diffraction, X-ray reflectivity and wafer curvature during sputter-deposition of a series of $Mo_{1-x}Si_x$ alloys. These unique measurements evidenced an interface-controlled crystallization process, taking place spontaneously above a composition-dependent critical thickness, with a constant growth front velocity (~13 nm/s).