Stress Evolution During Volmer-Weber Growth of Thin Films

Personnel

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We have carried out experimental characterization of stress evolution during polycrystalline film formation, using micromachined piezocantilevers and using laser reflection from the tips of cantilevers. Polycrystalline thin films form through nucleation of isolated islands that grow until they impinge and coalesce to form a continuous film. A compressive stress develops at the earliest stages of film formation, before island coalescence, and a tensile stress develops as islands coalesce. After coalescence is complete, so that a continuous film has formed, the tensile stress decreases, and, in some cases, the stress state evolves back into the compressive regime as the film is thickened.

The tensile stress that develops during island coalescence is thought to be associated with grain boundary formation. When islands first come into contact, grain boundary formation can proceed from the point of contact, and leads to the reduction of the high-energy surface area of both coalescing islands. For small islands, with high surface-to-volume ratios, the energy-per-unit-volume released due to the replacement of two surfaces by one lower-energy grain boundary can be very high. The fastest way for this process to occur is for the islands to strain to close the gap between them, as the grain boundary 'zips' up from the substrate surface. The strain energy that can be accommodated is balanced by the accompanying surface energy reduction [Figure 6].

We have analyzed the zipping process using finite element models and molecular dynamics simulations. Finite element models have been used to calculate the strain energy as a function of zipping distance, z_0. The total energy associated with zipping (the strain energy plus the grain boundary energy, minus the energy of the replaced surfaces) is calculated and the zipping distance that minimizes the total energy is determined. The stress associated with the energy-minimizing zipping is then calculated. This allows calculation of the stress as a function of the size of the coalescing islands,

and leads to predicted stresses that are in better agreement with experimental observations than analytic models developed by others [Figure 7]. Finite element analyses have been combined with simulations of island nucleation and growth to predict the average stress as a function of the mass-equivalent thickness. These calculations are quantitatively consistent with experimental observations for Ag films deposited on oxidized silicon. Finite element calculations can be readily modified to account for the effects of variable traction at the island-substrate interface, for different island-substrate contact angles, and for coalescence of islands with different sizes.

Molecular dynamics simulations of the zipping process have also been carried out. So far, only islands bound to substrates *without* interface traction have been investigated. Islands composed of up to 360 Ag atoms have been modeled using embedded atom potentials. Island-substrate interactions are modeled using a Leonard-Jones potential. We find that grain boundary formation occurs over time scales that are accessible through MD simulations, in that zipping occurs at very high rates [Figures 8 and 9]. The observed zipping height is consistent with the results of finite element modeling, suggesting that the physical model on which the finite element calculations are based, grain boundary formation through island straining, is valid.

We are further developing molecular dynamics simulations that will account for traction at the island-substrate interface. We also plan to carry out experiments involving *in-situ* Transmission Electron Microscope (TEM) observations of the early stages of polycrystalline film formation, using micromachined membranes.

In addition to studies of the origin of the tensile stresses associated with island coalescence, we are also investigating the origins of pre- and post-coalescence compressive stresses. It has been previously demonstrated

that growth interruptions during post-coalescence deposition lead to a reversible relaxation of a large component of the compressive stress that develops during deposition. We have recently found that this is also true in the pre-coalescence regime. We believe that the reversible component of the compressive stress observed during film deposition is associated with either changes in the adatom population, or changes in the shapes of island and grain surfaces, or both. We are currently investigating the effects of substrate temperature and deposition rates on the magnitude and on the rate of reversible stress changes that occur during growth interruptions.
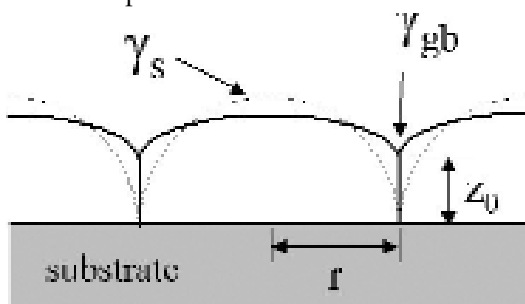

Fig. 6: When islands impinge, grain boundaries form as islands strain to close the gap between them, zipping a distance z_0 to replace two surfaces of energy γ_s with a boundary of energy γ_{gb}.
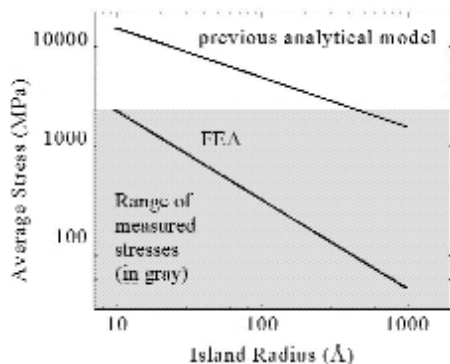

Fig. 7: Comparison of Finite Element Analyses (FEA) of the stress in coalesced islands, with results from previous analytic approximations. The gray region represents typical tensile stresses measured in experiments.
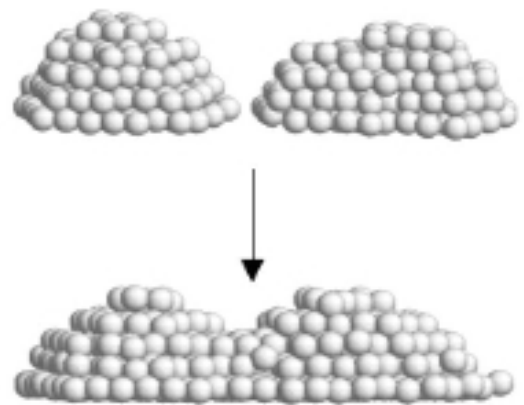

Fig. 8: Molecular dynamics simulation of grain boundary formation during island coalescence.
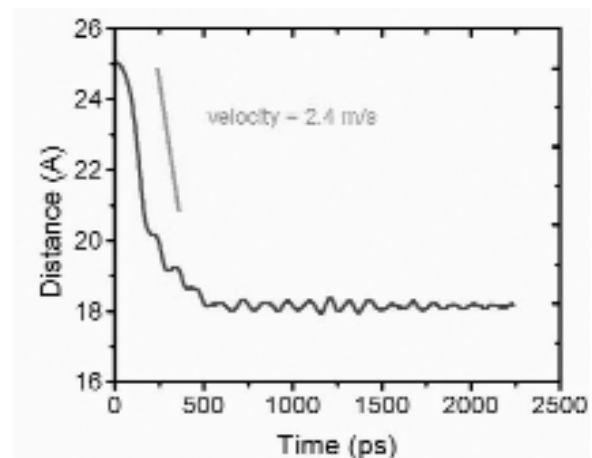

Fig. 9: Molecular dynamics results showing the distance between the centers of mass of coalescing islands as a function of time. A distance of roughly two atomic diameters is closed in approximately 0.5 nanoseconds.