564b Atomic-Scale Analysis of Radical Precursor and Hydrogen Surface Diffusion on Plasma-Deposited Amorphous Silicon Thin Films

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Hydrogenated amorphous silicon (a-Si:H) thin films grown by plasma-assisted deposition from silane-containing discharges are technologically important semiconductor materials, used widely in the fabrication of solar cells and flat panel displays. Film properties, such as surface roughness and film crystallinity, affect device performance significantly. A crucial factor in determining these properties is the mobility of reactive species, such as the deposition precursor, that impinge on the film surface during deposition. The dominant precursor for deposition of smooth device-quality a-Si:H films is the SiH_3 radical. In addition, post-deposition exposure of plasma deposited a-Si:H films to a flux of H atoms from an H_2 plasma can lead to crystallization of the amorphous films at temperatures much lower than those required for thermal annealing. In this context, the surface transport of SiH_3 radicals and H atoms plays a key role in determining film smoothness and affecting film crystallinity, respectively.

This presentation focuses on detailed atomic-scale analysis of diffusion of SiH_3 and H on the a-Si:H surface. Using molecular-dynamics (MD) simulations of impingement of SiH_3 radicals on growth surfaces of smooth a-Si:H films, we studied the diffusion mechanism of the SiH_3 radical over the temperature (T) range 475 T 800 K. In similar manner, we studied the diffusion mechanism of atomic H during H_2 plasma post-deposition treatment of the a-Si:H films by repeatedly impinging H atoms on the film surfaces. Our MD simulations employed a many-body interatomic potential that has been tested extensively by comparing its predictions with experimental measurements and accurate first-principles calculations based on density functional theory (DFT). In addition, we have carried out analysis of SiH_3 surface migration pathways based on DFT calculations using crystalline Si surfaces as representative models of atomic bonding at film growth surfaces. In our DFT calculations, we used the generalized gradient approximation, plane-wave basis sets, ultra-soft pseudopotentials, slab supercells, and the nudged elastic band method for determining surface migration pathways.

We have found the MD-grown a-Si:H film surfaces to be remarkably smooth due to valley-filling mechanisms mediated by diffusion of SiH_3, the mobile precursor. The mobility of the SiH_3 radical was determined through the evolution of the mean squared displacement of the radical's center of mass, as monitored from the MD trajectory. The temperature dependence of the calculated surface diffusion coefficient yielded an average activation barrier of 0.16 eV for radical diffusion on the a-Si:H surface. This low barrier for diffusion is due to the weak adsorption onto the a-Si:H surface of the radical and its migration predominantly through overcoordination defects. The DFT analysis revealed that the SiH_3 surface migration barrier is reduced to 0.18-0.25 eV when the radical is weakly bonded to surface Si atoms that are overcoordinated, consistent with the MD simulations on the a-Si:H surface. Furthermore, we have found that exposure of MD-grown a-Si:H films to H atoms from an H_2 plasma leads to formation of nanocrystalline regions in the films. Analysis of the corresponding MD trajectories revealed that H atoms diffuse on the a-Si:H surface and into the bulk film by hopping from one five-fold coordinated Si atom to another. This fifth bond, formed between the surface Si atom and the migrating H, introduces a floating bond that accompanies the H atom in its surface migration. The diffusing H atom inserts into strained Si-Si bonds, occupying a bond-centered-like Si-H-Si position. Structural relaxation of these strained Si-Si bonds, following the migration of H from the bond-centered configuration, triggers disorder-to-order transitions that lead to structural transformation of the film from amorphous to nanocrystalline.