206b *in Situ* Real-Time Diagnostics for Studying the Structural Evolution of Nanocrystalline Silicon Thin Films during Plasma Deposition

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The knowledge of the gas-surface interactions and film structure evolution during growth is crucial for understanding the growth mechanisms of thin films. We used *in situ* real-time attenuated total-internal reflection Fourier transform infrared spectroscopy (ATR-FTIR) and spectroscopic ellipsometry (SE) to monitor the growth of nanocrystalline silicon thin films at room temperature by plasma enhanced chemical vapor deposition (PECVD). The nanocrystalline silicon films were grown on a double side polished GaAs ATR crystal, which has two of its parallel side-faces cut to a 45^{o} beveled angle. The IR beam incident perpendicularly on one of the beveled edges is reflected multiple times from the bottom and the top of the ATR crystal. During this multiple reflection process, the IR beam passes through the growing film multiple times which increases the sensitivity of the technique. ATR-FTIR was used to monitor the evolution of silicon hydrides trapped in the growing film and in the grain boundaries between the nanocrystals. ATR-FTIR was also used to monitor the temperature of the growing film to ensure that the films were indeed grown at room temperature; during plasma deposition, the substrate can receive considerable heating from the plasma even in the absence of intentional substrate heating. Previous studies without substrate heating had shown growth of nanocrystalline silicon but whether the substrate temperature remained at room temperature was an open question. In fact, we show that even at a fraction of the plasma power used in those studies the substrate can be heated to nearly 200 ^{o}C just by ion bombardment from the plasma. In this study, we used very low intensity plasma and made sure that the substrate temperature remained <50 ^{o}C even during long deposition experiments. Spectroscopic ellipsometry was used to determine the thickness and the optical properties of the nanocrystalline silicon film. The optical properties of the film were simulated using a 3-layer model; this model consisted of a nanocrystalline Si layer sandwiched between two amorphous silicon layers. The crystalline fraction in the film was approximately 40%. The three-layer model was confirmed by obtaining bright-field cross-sectional image of the film. The thicknesses of each layer obtained by ellipsometry were comparable to the direct measurement made using transmission electron microscopy. From the real time spectroscopic ellipsometry data obtained during film growth, we concluded that the nanocrystalline thin films nucleate and grow through the recently proposed hydrogen-induced crystallization mechanism in the bulk under an amorphous crust. That is, amorphous silicon is deposited from plasma and then converted to nanocrystalline silicon through H-induced rearrangement of strained Si-Si bonds in the bulk of the film. Using ATR-FTIR and spectroscopic ellipsometry, we were able to follow the formation of the amorphous crust in the film and the appearance of crystals under this crust as well as the appearance of silicon hydrides that decorate the grain boundaries between the nanocrystals. These *in situ*, real-time techniques provided a quick assessment of the microstructure of the films, which was confirmed with *ex-situ*, and sometimes more time consuming, characterization techniques such as transmission electron microscopy and Raman spectroscopy.