

In situ growth studies during expanding thermal plasma deposition of doped and undoped ZnO

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The extensive research on ZnO films focuses on the application of this versatile transparent conductive oxide in applications ranging from photovoltaics to organic light emitting diodes. Many deposition techniques have been explored with varying success. In this talk we will discuss a remote-Plasma Enhanced Chemical Vapour Deposition technique, the Expanding Thermal Plasma (ETP) technique, which uses mixtures of diethyl zinc and oxygen as the primary feed gases to deposit ZnO from an argon-fed ETP system. The ETP technique has been successfully applied previously for high rate deposition of silicon oxide, hydrogenated amorphous carbon, silicon and nitride.

An overview of the work performed at the Eindhoven University of Technology in recent years will be given. We will discuss the successful implementation of highly conductive natively textured ZnO films in amorphous silicon solar cells and which have a similar efficiency as solar cells with Asahi fluorine doped tin oxide front contacts. In addition we will discuss the doping of ZnO by means of nitrogen and aluminum. The doped ZnO layers are deposited using an Ar-fed ETP system in which oxygen, diethylzinc, nitrogen gas (for N doping) and trimethylaluminum (for Al doping, respectively), are admixed downstream in the expanding beam. Apart from studies of the bulk material properties and implementation in devices, *in-situ* growth studies of surface morphology are presented.

A particularity of the ETP technique is that typical downstream ion energies in this remote plasma are < 2 eV, therefore energetic ion bombardment does not contribute/assist the film growth. We use *in-situ* spectroscopic ellipsometry to monitor the refractive index, thickness and roughness development during the film growth. Atomic force microscopy, Hall and X-ray diffraction measurements are used to determine the morphological, electronic and structural properties of the films, respectively. During nitrogen doping, infrared spectroscopy and mass spectrometry measurements point towards the incorporation of nitrogen as $-C\equiv N$ which is segregated to the grain boundaries. Hall and XRD measurements indicate that CN behaves as impurity and not as dopant. By means of Al doping we can obtain conductive films ($5 \cdot 10^{-4} \Omega\text{cm}$) with native roughness of ~ 45 nm for $1 \mu\text{m}$ thickness, suitable for solar cell applications. The film conductivity, however, shows a strong inhomogeneous depth profile, which could affect the suitability of this material for applications requiring thinner (< 200 nm) films, i.e. diodes and thin film transistors. To manipulate the growth process, the (doped) ZnO film growth is investigated by studying the overall effect of the substrate temperature, working pressure, and ion bombardment (as delivered by an external RF bias).