

Metal Oxide Thin Films for LED Applications

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Abstract:

Electroluminescence (EL) of silicon nanoparticles (Si NPs) holds promise for a new generation of light-emitting diodes (LEDs). While recently reported values for EL efficiency max out at around 1.6% [1], these numbers could be increased via improvement of charge delivery to the particles—optimization of the electron and hole transport layers (ETL, HTL). For the HTL, thin films of ZnO and Al-doped ZnO (AZO) were deposited on glass and silicon substrates via atomic layer deposition (ALD). Films' physical and electrical properties were determined by means of Auger electron spectroscopy (AES), atomic force microscopy (AFM), UV-VIS spectroscopy, profilometry, ellipsometry, and four-point probe measurements; resistivities in the range of $1.75 \times 10^{-3} \Omega\text{cm}$ were achieved while maintaining an 85% transparency across the visible spectrum. For the ETL, thin films of NiO and WO_3 were deposited in an RF plasma sputtering system. Deposition rates for each oxide were determined and tuned to be within range of each other for future co-deposition experiments. Oxygen concentration in process gases was found to have a significant impact on NiO film quality and deposition rates. Further investigation of the films and their properties is necessary to begin device construction.

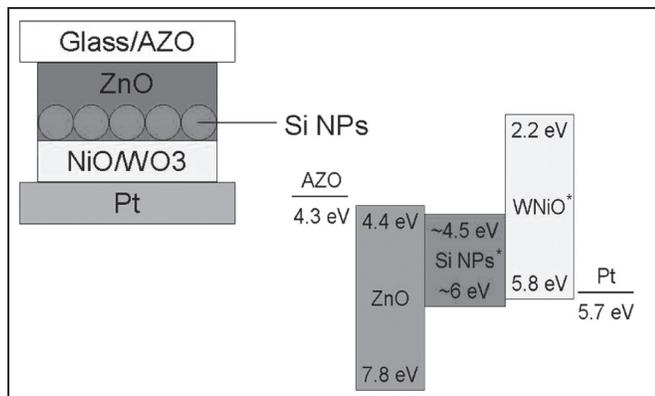


Figure 1: Theoretical device construction, including band gap data (some projected values).

Device Structure:

Electroluminescence requires hole-electron recombination within a semiconducting material. Thus, producing an efficient LED necessitates a series of layers to generate holes and electrons and transport them to the luminescent material for recombination. Figure 1 demonstrates a theoretical construction of this device. It should be noted that the band gaps for WNIO and Si NPs are projected values that will hopefully be obtained through further experimentation.

Experimental Procedures:

Atomic Layer Deposition. Thin films of Al-doped ZnO (AZO) were deposited on glass and silicon substrates via atomic layer deposition (ALD) in a Cambridge Nanotech Savannah ALD system. All laboratory work took place in a class 10 clean room. Substrates were sonicated for 10 min in each of the following solvents: deionized water, acetone, methanol, and isopropanol, then cleaned in an ozone atmosphere for 5 minutes (to produce a hydrogen-terminated surface) before

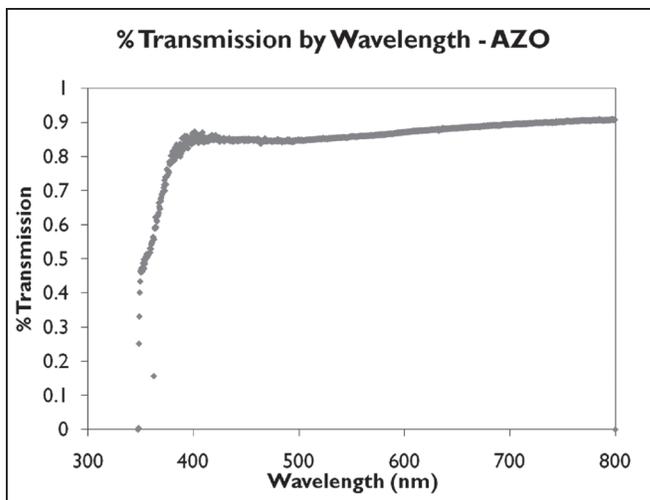


Figure 2: Optical transmittance of a ~50 nm AZO film (30:1) measured with a UV-VIS spectrometer.

being transferred to the ALD system. The precursor gases—trimethyl aluminum (TMA), diethyl zinc (DEZ), water vapor (H_2O)—were pulsed at 0.015 s, with a purge time of 5 s in 40 sccm of N_2 at 180°C . Doping ratios were varied from 10:1 to 50:1 ($\text{ZnO}:\text{Al}_2\text{O}_3$). Samples were characterized with single-wavelength ellipsometry, AFM, four-point probe, and AES.

RF Sputtering. Thin films of NiO and WO_3 were deposited on quartz and silicon substrates via RF-regulated plasma sputtering in an AJA ATC 2000 sputter system. Substrates were cleaned via the method detailed above, with the exception of the ozone cleaning. Deposition rates were determined by measuring film thickness via single-wavelength ellipsometry.

Results and Conclusions:

Figure 2 shows light transmission through the AZO thin film, which is 85% or greater throughout the visible spectrum. A 30:1 doping ratio maintained this transparency while having resistivities as low as $1.75 \times 10^{-3} \Omega/\text{cm}$. This is comparable to low resistivity values found in the literature for these films, $9.7 \times 10^{-4} \Omega/\text{cm}$ [2]. Films produced by this method are very smooth, exhibiting RMS roughness values of ~ 1 nm. Doping ratios and film purity were verified by AES, showing virtually no contamination in the deposition process. These results are very promising for both charge transport and device integration.

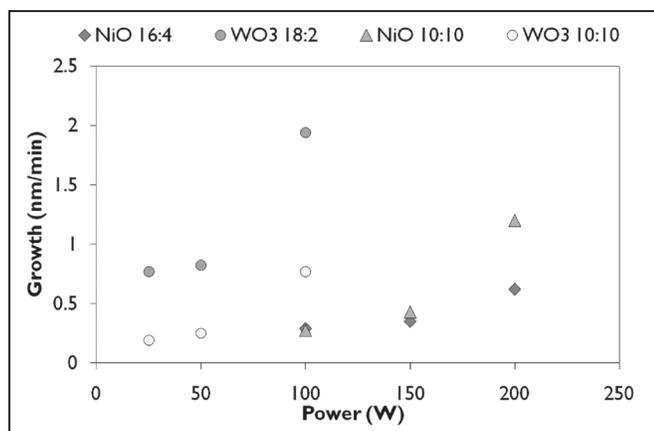


Figure 3: Deposition of metal-oxide films by an RF sputtering process. Atmosphere ratios ($\text{Ar}:\text{O}_2$) given in sccm.

Figure 3 illustrates deposition rate trends for the NiO and WO_3 films sputtered in different atmospheres of $\text{Ar}:\text{O}_2$. Ideally, NiO and WO_3 deposition rates would be identical within a certain range of wattages. With higher Ar concentrations, we found high rates of WO_3 deposition and low rates of NiO deposition. When the O_2 concentration in the sputtering atmosphere was increased, the WO_3 deposition rate decreased and the NiO deposition rate increased. This brought the two rates more in line with each other, below our system's upper limit of 200 W.

Future Work:

The deposition rate tuning detailed above paves the way for deposition and characterization of various WO_3/NiO alloys. Tuning the alloy ratios should lead to variations in the HTL material's band gap, which can then be optimized for the device. Further work should lead to a functional inorganic device, which will, in the best scenario, exceed previously disappointing electroluminescence efficiencies for Si NPs.

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References:

- [1] K. Cho, N. Park, T. Kim, K. Kim, and G. Sung, *Appl. Phys. Lett.* 86, 071909 (2005).
- [2] S. Kwon, H. Lee, Y. Seo, H. Jeong, *J. Kor. Phys. Soc.* 43; 5. p. 709-713 (2003).