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A Combinatorial Approach to TCO Synthesis and Characterization

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ABSTRACT

New and optimized metal-oxide thin films for both transparent-conductor and antireflection coatings have the potential to significantly improve photovoltaic devices. One approach to this task of discovery and improvement is to use a parallel optimization, i.e., a combinatorial, approach. Using internal seed funds, we have developed the initial deposition, characterization and analysis tools necessary for implementing a combinatorial approach to thin-film metal oxides, with a special focus on transparent conducting oxides (TCOs).

1. Introduction

Transparent conducting oxides [1] play a key role in a number of thin-film opto-electronic devices, including photovoltaics, flat panel displays, low-e windows, electrochromic devices, and anti-static coatings [2]. The bulk of these applications rely on the established n-type transparent conducting oxides (TCOs), such as SnO_2:F, ZnO:Al, and indium-tin-oxide (ITO) [3]. For many of these technologies, next-generation devices would be significantly enhanced with improved or new TCOs [4]. This is leading to the investigation of new n-type materials such as Cd_2SnO_4, $Zn_2In_2O_5$, and $In_{2-2x}Sn_xZn_xO_3$. These more complex materials are generally alloys of the simpler established transparent conducting metal oxides such as CdO, SnO_2, ZnO, In_2O_3, and Ga_2O_3. Combinatorial

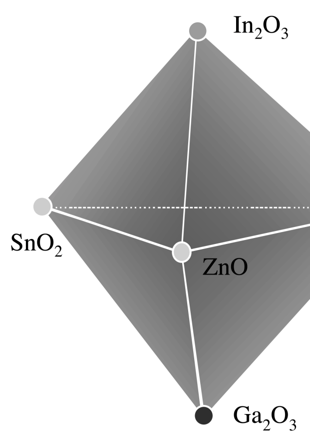

Fig. 1. Pictorial representation of the potential phase space of current interest for new TCOs [1].

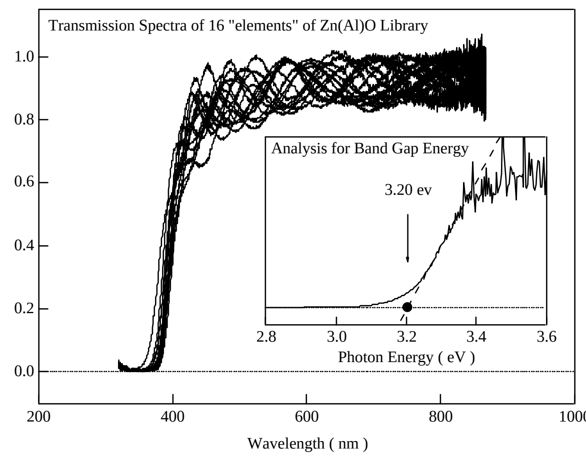

Fig. 2. Optical transmission spectra of 16 elements of a sputtered Zn(Al)O library.

approaches are thus needed to explore realistically the large compositional space represented by Figure 1 [5]. Combinatorial approaches are composed of three components: library deposition, critical-parameter analysis, and data mining to extract useful information and conclusions.

2. Combinatorial Deposition and Characterization

Zn(Al)O and Zn-Sn-O libraries have been deposited by co-sputtering using a three-gun sputtering system specifically built for combinatorial deposition. Cd-Sn-O libraries have been grown by chemical vapor deposition on an existing system by utilizing system-generated compositional gradients.

Figure 2 shows the optical transmission spectrum for 16 elements of a Zn(Al)O library. These spectra are measured using a UV/VIS/NIR CCD-based fiber-optically coupled spectrometer with measurement times of ~ 1 sec. per full spectrum. Figure 3 shows the infrared reflectivity vs. photon energy for an 18-element Cd-Sn-O library grown by CVD. All elements show the increasing infrared reflectivity due to the free-electron plasma oscillations indicative of a conductor. In the inset, the reflectance at 3000 cm^{-1} (circles, left axis) and the Hall-measured carrier concentration (triangles, right axis) are both plotted vs. the sample number. The good correlation shows that the infrared reflectivity is a viable noncontact diagnostic for

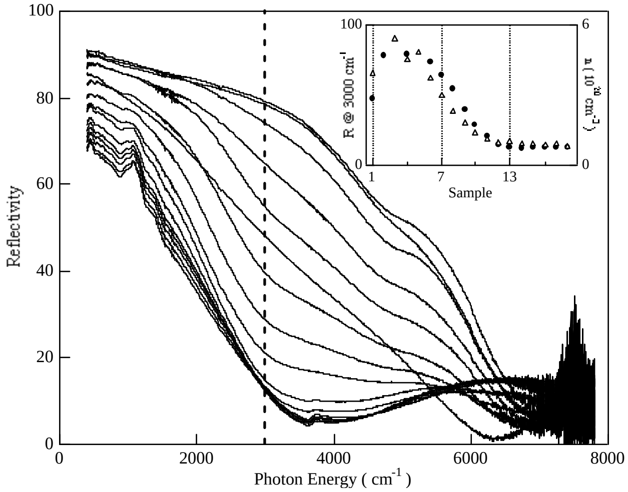

Fig. 3. FTIR reflectivity measurement of a Cd-Sn-O library.

combinatorial characterization of the relative carrier concentration in TCO libraries.

3. Additional Libraries

Using gray-scale intensity maps, Figure 4 summarizes several properties of a Zn(Al)O library that was used for technique verification. The upper left map, which shows the infrared reflectance, indicates a higher electron density on right side of the sample. The thickness, upper right map, shows good uniformity with thicknesses between 2000 and 3000 Å. The bulk conductivity map is shown in the bottom left. The bottom right graph shows the conductivity, reflectance at 2000 cm^{-1}, and the relative aluminum content, measured using electron microprobe, vs. column position for row 3. For this sample, the conductivity generally scales as expected with Al content.

We are now applying the developed combinatorial tools to the Zn-Sn-O binary TCO system. Initial results (10 libraries) suggest that $ZnSnO_3$ may be a substantially better TCO than Zn_2SnO_4, which is presently used as an interface layer in CdS/CdTe solar cells. If sufficiently conducting Zn-Sn-O can be developed, it may be possible to eliminate the current Cd_2SnO_4 TCO layer in these CdS/CdTe cells. These early results from our initial Zn-Sn-O libraries demonstrate the great potential for combinatorial approaches.

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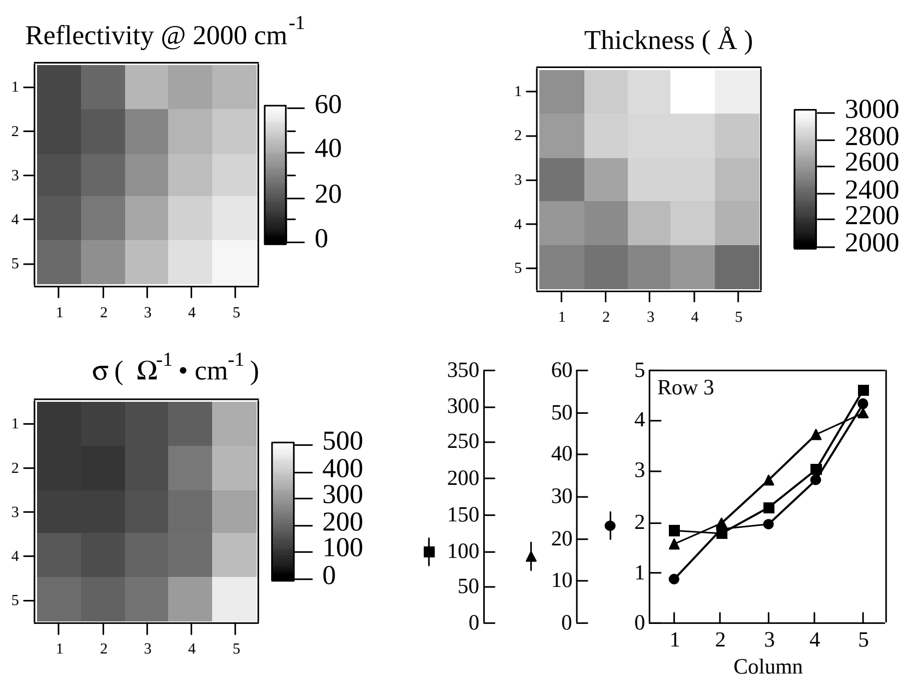

Fig. 4. Gray-scale intensity maps of relevant materials properties in a sputtered Zn(Al)O library.