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The Atomic Structure of Amorphous Oxide Semiconductors

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ABSTRACT Amorphous oxide semiconductors have important industrial applications in optoelectronic devices due to their transparency and high electrical conductivity. It is important to understand the arrangements of the atoms in these semiconductors since they determine the electrical properties of the materials. To investigate the atomic structure of indium oxide semiconductors, a series of nine thin films were deposited at different temperatures. Wide-angle x-ray scattering data were collected at Argonne National Laboratory. The images for the partially amorphous and fully crystalline films were calibrated, integrated, and analyzed. Structural results include the lattice parameters and positions of the atoms in the bixbyite phase. The lattice parameters directly correlate with the measured electrical properties of the samples.

INTRODUCTION

Transparent semiconductors are used in the forefront of modern technology. These transparent semiconductors are crucial to technology due to their ability to conduct electricity while also allowing light to pass through. These properties make them optimal for use in LCD and touch screens. Scientists have found that changing the deposition temperatures of thin film semiconductors can greatly affect the atomic structure and electrical properties of these oxides [1]. At high temperatures, the atoms of oxide semiconductors typically arrange themselves in a crystalline pattern. An example of this crystalline periodic pattern for indium oxide (In$_2$O$_3$) can be seen in Figure 1. However, at lower deposition temperatures, the atom arrangements may become more randomized. This project aimed to discover both how and why these atoms order themselves at certain temperatures, and determine how the temperature variance affected the electrical properties.

This research was part of a collaborative project between DePaul University and Northwestern University (Interdisciplinary Research Group 2 (IRG2) of the Materials Research Center). The main mission of IRG2 is to understand the processing, structure, and properties of amorphous oxide semiconductor (AOS) materials, leading to improved materials and enabling their applications. The González

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research group at DePaul University focused on determining the atomic structure of materials that were prepared and characterized at Northwestern University. The samples consisted of nine thin films of indium oxide prepared by the Chang Group (IRG2) via pulsed laser deposition at temperatures ranging from −50 °C to 600 °C. The thickness of the films was approximately 400 nm. Details of the procedure for similar but thinner films (~60 nm) can be found in reference [1]. The DePaul University research group collected two-dimensional diffraction data of these samples at Sector 1 of the Advanced Photon Source (APS) at Argonne National Laboratory. The results of this particular research project herein focused on calibrating, integrating, and performing a preliminary analysis of the diffraction data.

![Figure 1: The periodic crystal structure of indium oxide. The green and blue spheres represent indium atoms, and the red spheres represent oxygen atoms. The structure is cubic, and each side of the cube is equal to \( a = 10.117 \) Å (\( a \) is also known as the lattice parameter). [2]](image)

The data sets were collected in glancing incidence geometry using very shallow angles of the incident x-ray beam. Three different incident angles were used to probe different depths within the sample. The incident angles corresponded to \( \alpha_c \) (critical angle), \( 2\alpha_c \), and \( 4\alpha_c \). As the angle increases, the penetration depth of the x-ray beam into the sample increases, and a bigger volume of the sample is probed. The energy of the x-ray beam was approximately 70 keV. For each penetration depth the exact energy of the beam was carefully measured. The results presented here corresponded to the data collected at the critical angle using an x-ray energy of 69.958 keV. Each data collection consisted of four individual images, which required calibration and integration. The four images in the group were then combined to obtain complete diffraction rings. Approximately 500 images were collected and required analysis.

![Figure 2: The two-dimensional x-ray diffraction detector panels and the thin film experimental setup at the Sector 1 of the APS.](image)

**METHODS**

**DATA COLLECTION**

A four-detector panel was used to collect the wide-angle x-ray scattering data. A photograph of the experimental setup is shown in Figure 2.

**DATA ANALYSIS**

GSAS II [3], an open-source program, was utilized to perform precise calibrations and integrations of image data. Before the analysis of \( \text{In}_2\text{O}_3 \) samples could be processed, wavelength calibrations and image integrations...
of standard reference powders was necessary. Two standard materials, LaB₆ and CeO₂, were used to determine the wavelength of the x-rays and the resolution of the instrument. These standards were obtained from the National Institute of Standards and Technology [4,5]. GSAS-II is optimized for dealing with full diffraction patterns. Due to the fact that ¼-sized sections were being used, internal adjustments and conventions had to be implemented in order to obtain reasonable calibration and integration results. The calibration for one image containing ¼ of the diffraction pattern is shown in Figure 3, where the multicolored rings are the auto-fits projected by the software. The program fitted the location of the x-ray beam center and corrected for any tilt in the detector that can result in asymmetric rings. Once a calibration was complete, GSAS-II superimposed fitted rings over the diffraction image data to give an estimate of the goodness of fit.

Figure 3: This image displays a sample calibration done with GSAS-II. The scale at the right represents the intensity.

Since the thin film samples were deposited on fused silica substrates, an empty substrate was also tested through the same conditions. The contribution of the empty glass substrate was subtracted from each sample image. As displayed in Figure 3, hundreds of diffraction rings were collected. It was crucial to fit as many rings as possible in order to obtain accurate atomic information for the In₂O₃ samples.

After the calibrations were performed, the integration function of GSAS-II was used. A radial integration was done to obtain the intensity peaks across the 2-theta range. With these intensity vs. 2 theta graphs it was then possible to continue the analysis with the software FullProf [6] to study the atomic structures and arrangements for that particular material. For these integrations, an accurately calibrated image resulted in sharp and well-defined intensity peaks.

RESULTS AND DISCUSSION

Numerous calibrations and integrations were performed for all samples. The integration of data for a specific series of nine samples is shown below.

Figure 4: A compilation of integration images at the critical angle. The colored diffraction patterns correspond to different deposition temperatures.

As seen in Figure 4, more sharp peaks were obtained at higher deposition temperatures, which meant that the films had a higher number of crystallites than at lower deposition temperatures. Since broad peaks corresponded to amorphous states, the films deposited at ~50
°C and 0 °C were completely amorphous. At room temperature, small crystals formed, resulting in the appearance of sharp peaks. This indicated that near this temperature the atoms of the semiconductors began arranging themselves in a periodic manner. As the deposition temperature increased, the samples became even more crystalline. These integrations were the input for the software package FullProf [6], which applied inverse Fourier transformations of the data to give information about the atomic positions, the lattice parameters, the different bond lengths, and the coordination numbers. Some of the lattice parameter results are shown below.

![Figure 5](https://via.library.depaul.edu/depaul-disc/vol3/iss1/9)

**Figure 5**: The lattice parameter data obtained from Figure 4 against the deposition temperature. The red line corresponds to the lattice parameter of fully crystalline indium oxide. The error bars for the data points are too small to be seen.

Figure 5 displays how the lattice parameter changed as the films became more crystalline. The drastic change in the lattice parameter suggested that during the initial crystallization stages, nano grains with large distortions were formed. More random arrangements corresponded to a higher lattice parameter. As the deposition temperature increased, the lattice parameter dropped and settled around the typical lattice parameter for crystalline indium oxide (shown as a horizontal red line). Furthermore, for the higher temperature films, the analysis showed that positions of the indium and oxygen atoms matched well those for crystalline indium oxide.

Due to the collaborative nature of this research, the work completed was only a mere fraction of the large puzzle of amorphous oxide semiconductors. Recently, the Chang group of the IRG 2 recorded electrical properties of these materials, and interesting results have been observed. The conductivity is displayed in Figure 6.

![Figure 6](https://via.library.depaul.edu/depaul-disc/vol3/iss1/9)

**Figure 6**: The conductivity of the samples as deposition temperature increased. Data collected by Dr. D. Bruce Buchholz (Chang’s group, IRG2).

These data are particularly interesting mainly due to the astounding change in conductivity and its direct correlation with the change in the lattice parameter (Figure 5). The conductivity is the product of the mobility and electron concentration. Correlating Figures 6 and 4, the best electrical properties were obtained just before crystallization began. The conductivity decreased for higher deposition temperatures. As mentioned earlier, the results presented here were obtained from the data collected at the critical angle, which means that the structural information corresponds mainly to the surface of the thin film. In the future, the data collected at higher critical angles will be analyzed to determine the homogeneity of the samples and...
to study any structural differences as a function of penetration depth in the films.

One possible explanation for the change in conductivity is that as the film began to crystallize, the interfaces between the small crystal particles with the amorphous matrix disrupted the electrons’ paths. These small crystal particles acted as obstacles and scattered the electrons, lowering the mobility. The overall behavior of conductivity as a function of crystallinity shows why amorphous oxide semiconductors would be a much more efficient and electrically superior to crystalline materials.

Through this research, the electrical behavior of these semiconductors was correlated to the atomic structures. Though there is still much work to be done, qualitative predictions about the structural properties of amorphous oxide semiconductors can be made, which can lead to more efficient and more cost-effective materials. Since semiconductors are vital in technologies, such as microprocessors, computer screens, and virtually all modern technologies, an increased understanding of the atomic arrangements and their effects on the electrical properties will benefit the fast-paced market.

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