Advancements in Transparent Conducting Oxides

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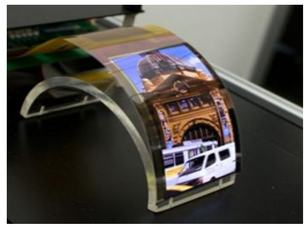
Abstract The purpose of this research was to develop a methodology to analyze x-ray pair-distribution function data for amorphous and crystalline transparent conducting oxides. The samples consisted of indium-based oxide thin films ranging from fully amorphous to fully crystalline. X-ray scattering data were collected at the Advanced Photon Source of Argonne National Laboratory. The data were analyzed with existing software packages and new Matlab code. The analysis enables the extraction of information about the short-range ordering of the atoms in the materials.

Motivation

Transparent Conducting Oxides (TCOs) are semiconductors that have high electrical conductivity and are transparent in the visible region. TCO crystalline electrodes are used in the electronics industry in flat-panels, solar cells, and energy-efficient windows. Some amorphous TCOs benefit from having increased electrical properties as well as improved mechanical properties.



TCO electrodes in current commercial applications are indium-oxide (IO) based. This research project focused on studying series of undoped and doped IO thin films in both amorphous and crystalline form.

Figure 1: Because amorphous TCOs are not rigid, they can be deposited on plastic substrates, which can flex without breaking, further expanding their applications.

The samples were grown by pulsed laser deposition (PLD) at Northwestern University. They were deposited on silica substrates. The substrate temperature was varied from – 50 °C to 600 ° C.

X-Ray Scattering Experiments

X-ray pair-distribution function (PDF) measurements were conducted at the Advanced Photon Source at Argonne National Laboratory. High energy x-rays were used to scatter from the samples at very shallow incident angles. The x-ray patterns were recorded on two-dimensional detectors, as shown below. Data were collected up to $Q = 20$ 1/Å.

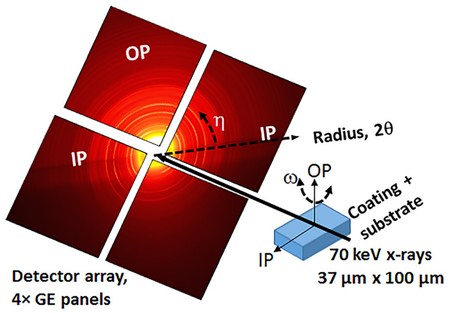


Figure 2: Schematic of the experimental setup, showing the sample and the arrangement of the four-panel array of 2D detectors. Figure taken rom [1]

[1] González G.B., Okasinski J.S., Buchholz D.B., Boesso J., Almer J.D., Bedzyk M.J., and Chang, R.P.H. "Relationship between electrical properties and crystallization of indium oxide thin films using ex-situ grazing-incidence wide-angle x-ray scattering". *J. Appl. Phys* 121, 205306, 2017.

Data Integration

The analysis of the data included integrating the 2D images into 1D data files with GSAS II [2]. Powder standards obtained from the National Institute of Technology (NIST) were used to calibrate the x-ray wavelength, sample-to-detector distance, and the the detector tilt.

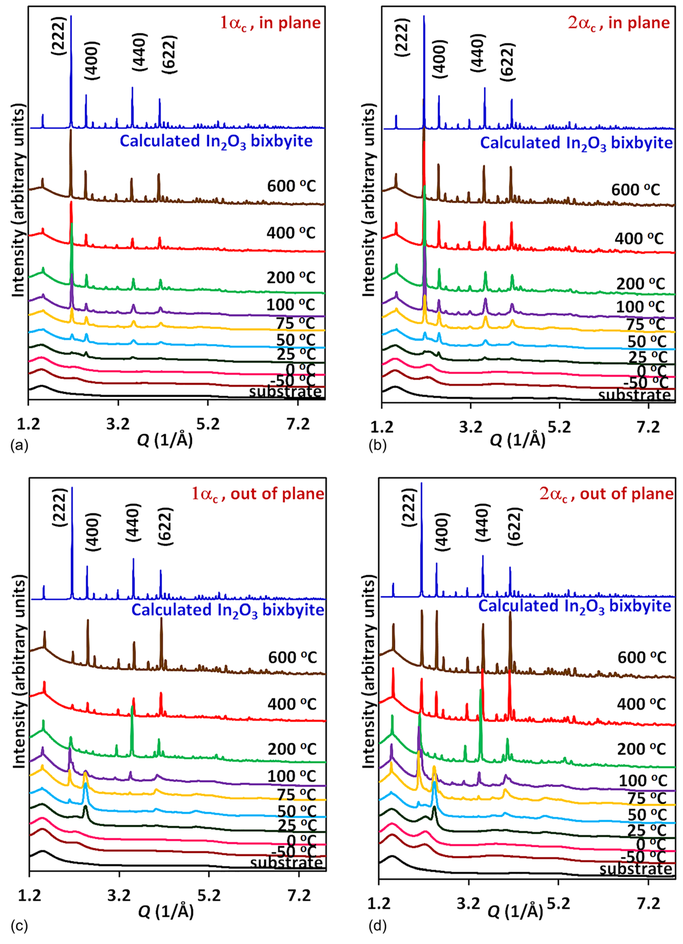


Figure 3: Integrated intensity vs. *Q* for IO thin films. (a) 1α, in plane (b) 2α, in plane (c) 1α, out of plane (d) 2α, out of plane The films at and above room temperature were crystalline. The films crystallize in the indium oxide bixbyite structure. The films have lateral and depth gradients. Figure taken from [1]

[2] Toby B. H. and Von Dreele R. B., "GSAS-II: the genesis of a modern open-source all purpose crystallography software package". *J. Appl. Cryst.* 46(2), 544-549, 2013.

[3] Juhas P., Davis T., Farrow C.L., and Billinge S.J.L. "PDFgetX3: a rapid and highly automatable program for processing powder diffraction data into total scattering pair distribution functions". *App. Cryst.*, 46, 560-566, 2013.

[4] MATLAB Release 2016a, The MathWorks, Inc., Natick, Massachusetts, USA.

PDF Analysis

PDFGEXT3 [3] was used to Fourier transform the data and subtract the substrate signal, leaving only the signal from the thin films.

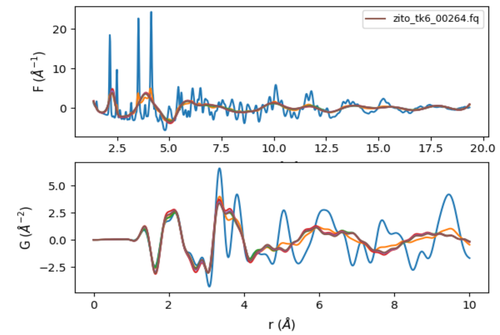


Figure 4: A series of samples after corrections with pdfgetx3. The bottom graph shows the PDF in real space. From this graph, information about the different pairs can be obtained.

Custom MATLAB [4] scripts were then used to fit and obtain the peak area (proportional to the coordination number), peak position (bond length), and peak width (proportional to the disorder) for the first coordination shells (metal-oxygen, and metal-metal pairs). These custom MATLAB scripts transformed the PDFs into RDFs (radial distribution functions). The data from MATLAB revealed the interesting characteristics of the amorphous and crystalline samples.

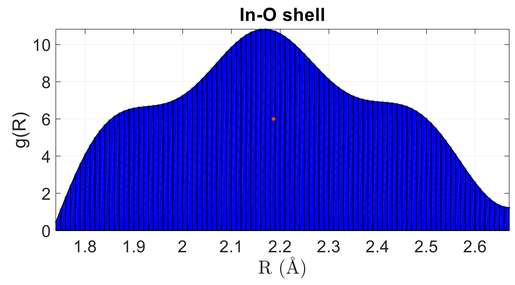


Figure 5: In-O peak showing its "center of mass", which corresponds to the bond length of this pair. The coordination number is proportional to the peak area, and FWHM is related to the disorder in this first coordination shell.

Acknowledgements: This project was funded by the NSF-NU MRSEC grant number DMR-1121262. The x-ray experiments were conducted by John S. Okasinski at Sector 1 of the APS, which is funded by the Office of Science, Office of Basic Energy Sciences in the U.S. Department of Energy. Bruce D. Buchholz from the Chang group prepared the samples and measured their electrical properties at Northwestern University.