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Nondestructive Characterization of Atomic Density Profile in CdS/Zn_2SnO_4 Junctions by X-ray Fluorescence

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ABSTRACT

Atomic density profile of selected atomic species in a series of CdS/Zn_2SnO_4 junctions has been investigated by using the method of Angular Dependence of X-ray Fluorescence (ADXRF). Samples of CdS, Zn_2SnO_4 (ZTO), and CdS/ZTO junctions were grown on glass, followed by different annealing processes. Special attention is directed to a comparison of samples heat-treated in argon and in $CdCl_2$ at different temperatures. It has been found that different heat treatment conditions can result in drastic variations in the density distribution of the constituents in the system.

It has been discovered recently that the performance of CdS/CdTe solar cells can be greatly enhanced by integrating a thin layer of Zn_2SnO_4 (ZTO) into the interface between CdS and the transparent conducting oxide films. This ZTO layer plays an important role for improving the CdTe cell efficiency to a new world record. The mechanism responsible for this improvement is believed to be mainly associated with interdiffusion of CdS and ZTO across the interface. More quantitative understanding of the compositional depth distribution of CdS and ZTO would seem highly desirable.

To examine the compositional depth profile without perturbing the material structure under study, the *angular dependence of x-ray fluorescence* (ADXRF) technique is particularly useful. This nondestructive method is also element-specific, thus it is well suited for probing the depth distribution of selected atomic species in complex systems. This method can be very useful for solving the problem of ubiquitous intermixing of constituent elements across the heterointerfaces, as found in many advanced thin film PV materials.

The CdS film (~800 Å) was deposited on 7059 Corning glass substrates by a chemical-bath deposition (CBD) technique using cadmium acetate $Cd(C_2H_3O_2)_2$, ammonium acetate ($NH_4C_2H_3O_2$), ammonia hydroxide (NH_4OH), and thiourea ($CS(NH_2)_2$). The ZTO film (~1400 Å) was deposited on the CdS film by rf magnetron sputtering. A commercial hot-pressed oxide target with a composition of 33 mol% SnO_2 and 67 mol% ZnO was used. Deposition was made in pure oxygen at room temperature, and the samples were then annealed in Ar, He, or $CdCl_2$ environment at different temperatures.

The fluorescence intensity from specific atoms can generally be expressed as

$$I_{FY} \propto \int dz\left(-\frac{dS_z(z)}{dz}\right)\Phi(z), \qquad (1)$$

where S_z is the z-component (perpendicular to the interface) of the Poynting vector, and $\Phi(z)$ is the density profile of the specific atomic species in the z-direction. The data can be analyzed from the field distribution assuming a model of the depth profile of the selected atoms in the system. By a comparison of the experimental results with theoretical calculations, information of the depth distribution of a specific constituent atomic species can be obtained. Some details of this technique can be found in previous publications: J. Appl. Phys. **83**, 4173 (1998); Appl. Phys. Lett. **74**, 218 (1999); J. Appl. Phys. **86**, 6052 (1999); and J. Vac. Sci. Technol **A17**, 2685 (1999).

The x-ray experiments were performed at beamline X3B1 of the National Synchrotron Light Source in Brookhaven National Laboratory. The energy of incident photons was selected by using a Si(111) double-crystal monochromator. A two-axis goniometer with angular resolution of 0.001° was used to control the grazing incidence angle θ between the incident x-ray beam and the sample surface. The x-ray fluorescent photons from the irradiated samples were collected by using a solid state Si (Li) detector and the intensity of Zn or Cd Kα fluorescence yield (FY) was measured as a function of θ using a single channel pulse-height analyzer.

Some typical ADXRF data obtained with CdS/ZTO junctions prepared under different conditions are shown in Fig. 1. The FY output from Cd atoms in the system is plotted against the grazing incidence angle (or the penetration depth) of x-rays impinging from the ZTO side. The Cd FY is low at small angles depending on the amount of Cd present in the ZTO film, it rises rapidly when the x-rays penetrate into the CdS layer at the critical angle near 0.1^{o}. This also indicates the approximate position of the

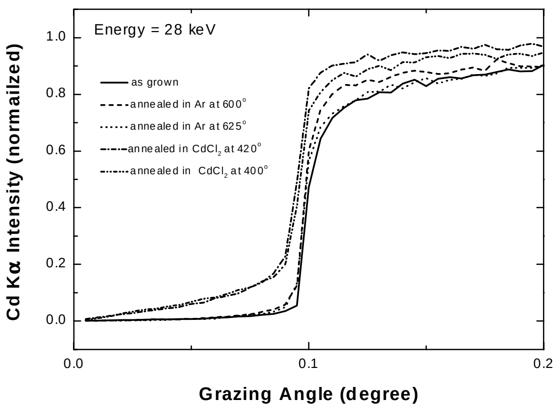

Fig. 1 – Cd Kα FY intensity vs. incident angle

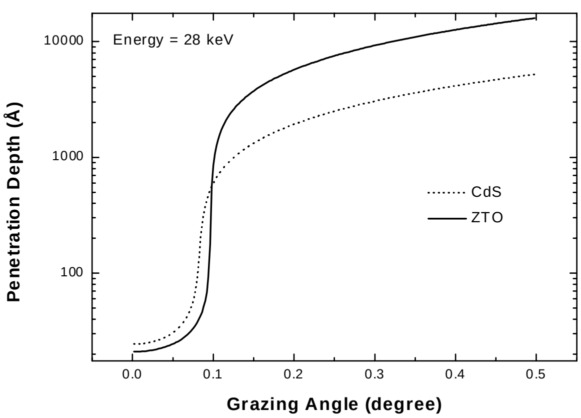

Fig. 2 – Conversion between grazing incidence angle and x-ray penetration depth.

CdS/ZTO interface. It can be seen that annealing in Ar at relatively high temperatures results in only very small changes in the Cd depth distribution as compared to the as-made sample. However, annealing in $CdCl_2$ can lead to a substantial increase of Cd atoms in the ZTO film, as revealed by a marked increase of the Cd FY in ZTO (below 0.1^o) and also in the vicinity of the interface.

The conversion from grazing incidence angle to penetration depth of x-rays into ZTO or CdS is illustrated schematically in Fig. 2. In the present case, the incident x-ray enters the sample from the top ZTO layer, hence the photon flux increases rapidly as the grazing angle reaches approximately 0.1^o, where the x-ray penetration depth exceeds the ZTO layer thickness.

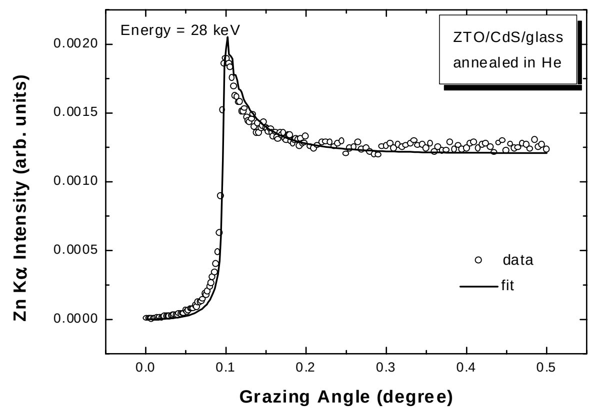

Fig. 3(a) – Comparison between data and theoretical calculation for Zn Kα FY output.

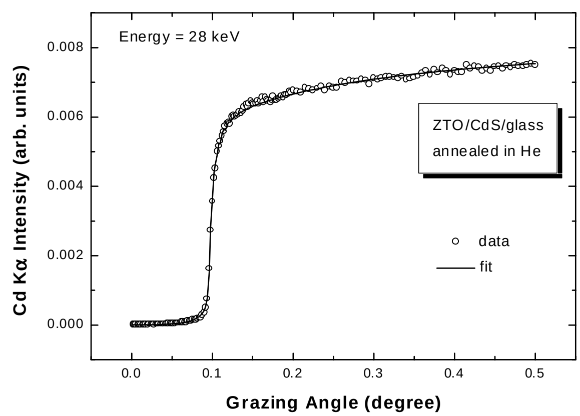

Fig. 3(b) – Comparison between data and theoretical calculation for Cd Kα FY output.

Figures 3(a) and 3(b) show examples of a comparison between data and theoretical calculations for ADXRF of the Zn Kα and Cd Kα, respectively, the agreement is quite satisfactory. Further quantitative analysis is in progress.

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