Evidence of Oxygen Vacancy Enhanced Room-temperature Ferromagnetism in Co Doped ZnO

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Transition metal (TM)-doped oxides have been investigated as a promising class of diluted magnetic semiconductors (DMSs) to bring out advanced spintronic devices since the discovery of room temperature (RT) ferromagnetism in these systems. However, a number of studies[1,2] indicate the RT ferromagnetism in TM-doped oxides may come from precipitation of magnetic clusters or from secondary magnetic phases. On the other hand, there are reports suggest the absence of magnetic clusters or secondary phases and support intrinsic ferromagnetic origin.[3,4] Even so, the origin of ferromagnetism in oxide DMSs remains a very controversial topic. In this letter, we study the correlation of electronic structure, local structure and magnetic properties of Co-doped ZnO thin films. **The XAS experiments were carried out in the wiggler-C (17C1) beam line.**

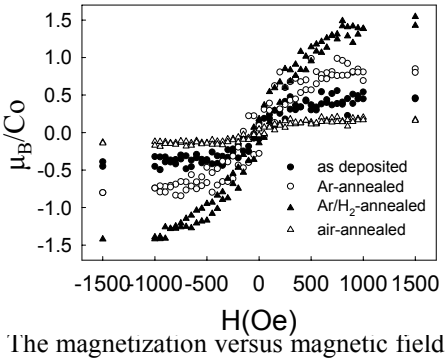

Figure 1. The magnetization versus magnetic field curves for the as deposited and annealed Co:ZnO films at room temperature.

The annealing effects on structure and magnetism for Co-doped ZnO films under air, Ar and Ar/H_2 atmosphere at 250°C have been systematically investigated. Room temperature (RT) ferromagnetism has been observed for the as-deposited and annealed films. However, the saturation magnetization (M_s) varied drastically for different annealing processes with M_s~0.5, 0.2, 0.9 and 1.5 μ_B/Co for the as-deposited, air-annealed, Ar-annealed and Ar/H_2-annealed films, respectively.

The magnetic and XAS results suggest that the observed RT ferromagnetism for the as-deposited and annealed samples is in accompany with good intrinsic DMS structures.Therefore, the enhancement of ferromagnetic signal by Ar and Ar/H_2 annealing should come from microstructural variation of DMS phase, instead of from extrinsic factor such as precipitation of Co clusters.

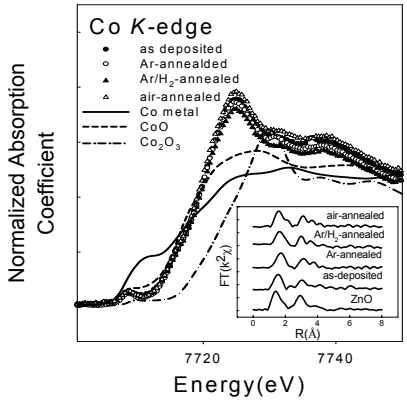

Figure 2. Normalized absorption spectra of the as -deposited sample and annealed films on Co *K*-edge. The spectra of Co foil and Co oxides are also provided for reference. The inset shows the radial distribution function on Co *K*-edge of the as -deposited and annealed films together with the Zn *K*-edge of ZnO (500Å) for reference.

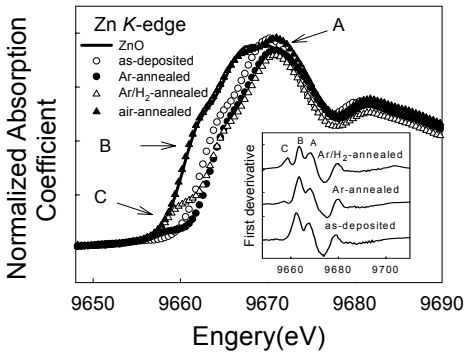

Figure 3. Normalized absorption spectra for the ZnO (500Å), as -deposited and annealed films on Zn *K*-edge. The inset shows the first derivative of normalized absorption spectra for the as-deposited, Ar-annealed and Ar/H_2-annealed films.

We show that XANES on Zn *K*-edge is rather sensitive to the formation of oxygen vacancies in ZnO modulated by the annealing processes. The change in pre-edge feature of XANES spectra is associated with the concentration variation of oxygen vacancy as revealed by the scattering calculation. Evidence of oxygen vacancy enhanced magnetization is provided by comparison of the magnetic and XANES results. Our finding has important implications for the current research on the magnetic origin in TM doped oxide DMS systems.