

## Photoemission study of the diluted magnetic semiconductor $\text{Zn}_{1-x}\text{Co}_x\text{O}$

M. Kobayashi<sup>1</sup>, Y. Ishida<sup>1</sup>, J. I. Hwang<sup>1</sup>, T. Mizokawa<sup>1</sup>, A. Fujimori<sup>1</sup>, K. Mamiya<sup>2</sup>,  
H. Saeki<sup>3</sup>, H. Tabata<sup>3</sup>, T. Kawai<sup>3</sup>

<sup>1</sup>Department of Physics and Complexity Science and Engineering, Univ. of Tokyo, Kashiwa, Chiba 277-8561, Japan

<sup>2</sup>Synchrotron Radiation Research Center, JAERI, SPring-8, Mikazuki, Hyogo 679-5148, Japan

<sup>3</sup>Institute of Science and Industrial Research, Osaka Univ., Ibaraki, Osaka 567-0047, Japan

### Introduction

Diluted magnetic semiconductors (DMSs) are key materials in spintronics (spin plus electronics), which is intended to manipulate both the spin and charge degrees of freedom. ZnO-based DMS have been predicted to be a candidate for a room temperature ferromagnet by theoretical studies [1]. In n-type Co-doped ZnO thin films, Ueda *et al.* succeeded to observe room temperature ferromagnetism [2]. In DMS, Photoemission spectroscopy (PES) and configuration-interaction (CI) analysis using the cluster model are powerful tools to investigate the electronic structure of the substituted transition metal ion [3].

### Experimental

A  $\text{Zn}_{1-x}\text{Co}_x\text{O}$  ( $x=0.05$ ) thin film was prepared on a  $\text{Al}_2\text{O}_3$  (0001) substrate by the pulsed laser deposition technique using an ArF excimer laser at the substrate temperature of  $\sim 300^\circ\text{C}$  in an ambient oxygen pressure of  $1.0 \times 10^{-5}$  mbar. The total thickness of the  $\text{Zn}_{1-x}\text{Co}_x\text{O}$  layer was  $\sim 200\text{nm}$  on a 50nm ZnO buffer layer.

Ultraviolet photoemission (UPS) measurements were performed at BL-18A of Photon Factory. Spectra were taken in an ultrahigh vacuum below  $7.5 \times 10^{-10}$  Torr. Photoelectrons were collected in the angle integrated mode at room temperature. The total resolution of the spectrometer with a VG CLAM hemispherical analyzer including temperature broadening was about 200 meV. Sample surface was cleaned by  $\text{Ar}^+$ -ion sputtering at 1.5 kV and annealing at  $250^\circ\text{C}$ . Cleanliness was checked by low energy electron diffraction (LEED) and the absence of a high binding-energy ( $E_B$ ) shoulder in the O 1s spectrum and of C 1s contamination by x-ray photoemission spectroscopy (XPS).

### Results

Absorption spectrum measured in the total yield mode shows that Co 3p  $\rightarrow$  3d absorption occurs at  $h\nu \sim 61$  eV. Figure 1 (a) shows the Co 3d partial density of states (PDOS) of  $\text{Zn}_{0.95}\text{Co}_{0.05}\text{O}$ , which has obtained by subtracting the off-resonance spectrum ( $h\nu = 60.0$  eV) from the on-resonance one ( $h\nu = 61.5$  eV) that have been normalized to the integrated valence band (VB) intensity of ZnO (integration range:  $0 < E_B < 9$  eV). The Co 3d PDOS shows a peak at  $E_B \sim 3.0$  eV, and a satellite at  $E_B \sim 7.0$  eV.

By applying the CI cluster-model analysis to the Co 3d PDOS in the VB, electronic structure parameters are obtained as the ligand to 3d charge transfer energy

$\Delta = 5.0 \pm 0.5$  eV, the d-d Coulomb interaction energy  $U = 6.0 \pm 0.5$  eV and the Slater-Koster parameter ( $pd\sigma$ ) =  $-1.6 \pm 0.1$  eV (a transfer integral between O 2p and Co 3d). These parameters are consistent with the chemical trend in II-VI DMSs [4]. Figure 1(b) shows that the main structure of the spectrum dominantly consists of the charge-transferred electronic states, i.e.  $d^7\bar{L}$ , where  $\bar{L}$  denotes a ligand 2p hole. These results mean that it is important to consider the strong correlation effects for the electronic structure of Co.

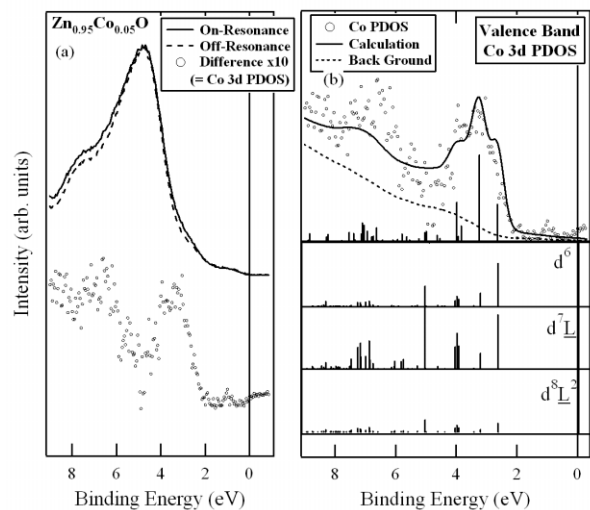


Fig. 1. (a) On-resonance ( $h\nu = 61.5$  eV) and off-resonance ( $h\nu = 60.0$  eV) spectra normalized to the integrated valence band intensity of the ZnO. Difference between these spectra represents the Co 3d PDOS. (b) CI cluster model analysis for the Co 3d PDOS.

### References

- [1] K. Sato and H. Katayama-Yoshida, Jpn. J. Appl. Phys. 40, 790 (2001).
- [2] K. Ueda, H. Tabata and T. Kawai, Appl. Phys. Lett. 79, 988 (2001)
- [3] T. Mizokawa, T. Nambu, A. Fujimori, T. Fukumura and M. Kawasaki, Phys. Rev. B 65, 085209 (2002)
- [4] T. Mizokawa and A. Fujimori, Phys. Rev. B 56, 6669 (2002)

\*masaki@wyvern.phys.s.u-tokyo.ac.jp