18A/2004G002

Co-3*d* partial density of states of heavily Co-doped ZnO

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Introduction

Recently, it has been reported that transparent oxide-based diluted magnetic semiconductors (DMS's), especially ZnO-based DMS's, show ferromagnetism with Curie temperature above the room temperature (RT). These DMS's have attracted much attention as candidates for room temperature ferromagnetic DMS's [1]. Although there are several reports on RT ferromagnetic Co-doped ZnO, the ferromagnetism is still in dispute because of low reproducibility and of the possibility of extrinsic origins of ferromagnetism such as Co-metal clusters.

In this work, we have succeeded in observing the Co-3*d* partial density of states of Co 15%-doped ZnO DMS. The PDOS have been compared with that of the Co 5% one and cluster-model calculation.

Experiment

A $Zn_{1-x}Co_xO$ (x=0.15) thin film was epitaxially grown on an α-Al_2O_3(0001) substrate by the pulsed laser deposition technique using an ArF excimer laser. The total thickness of the $Zn_{1-x}Co_xO$ layer was ~200 nm on a 50 nm ZnO buffer layer.

Co 3*p* → 3*d* resonant photoemission measurements were performed at BL-18A. Spectra were taken in an ultrahigh vacuum below 7.5×10^{-10} Torr. Photoelectrons were collected with a VG CLAM hemispherical analyzer in the angle integrated mode at RT. The total resolution of the spectrometer including temperature broadening was about 200 meV. Sample surface was cleaned by cycles of Ar^+-ion sputtering. Cleanliness of the sample surface was checked by the absence of a high binding-energy (E_B) shoulder in the O 1*s* spectrum and C 1s contamination by x-ray photoemission spectroscopy with Al-$K\alpha$ source ($h\nu$ = 1486.6 eV).

Results and discussion

Ultraviolet absorption spectrum measured in the total electron yield mode shows a clear Co 3*p* → 3*d* absorption at about $h\nu$ ~ 61 eV. Then, we choose an on-resonance spectrum taken at $h\nu$ = 61.5 eV and off-resonance one at $h\nu$ = 60 eV. The top panel of Fig. 1 shows on- and off-resonance spectra normalized to the integrated intensity in ZnO between 0 and 9 eV. The Co 3d PDOS has been obtained by subtracting the off-resonance spectrum from the on-resonance one. The PDOS shows a peak near the valence band maximum and a satellite at E_B ~ 7 eV as shown in the bottom panel of Fig.1.

By applying configuration-interaction (CI) cluster-model analysis for the PDOS, electronic structure parameters, namely, the ligand to 3*d* charge transfer energy Δ, the d-d Coulomb interaction energy U and the Slater-Koster parameter ($pd\sigma$), were obtained to be the same as in a previous report on $Zn_{1-x}Co_xO$ (x = 0.05) [2]. This is consistent with XPS results (not shown). This result implies that one can incorporate Co ions into ZnO at least up to 15% without forming secondary phases and/or Co-metal clusters. Since $Zn_{1-x}Co_xO$ with low Co concentration (<5%) showed RT ferromagnetism, it is expected that heavily Co-doped ZnO shows stronger RT ferromagnetism if it was supplied with enough carriers.

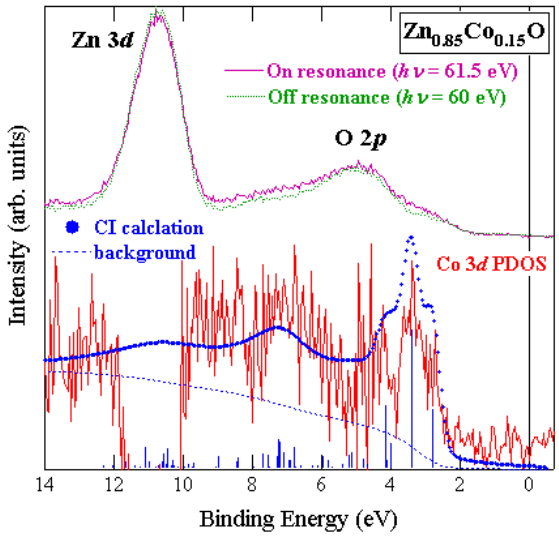

Fig. 1. Valence-band photoemission spectra. Top: On- ($h\nu$ = 61.5 eV) and off-resonance ($h\nu$ = 60 eV) spectra normalized to the integrated valence band intensity of the ZnO. Bottom: CI cluster-model analysis for the Co 3*d* PDOS.

References

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