R06

Optical and Magnetic Properties of Polycrystalline $Zn_{1-x}Co_xO$ Ceramics

T. L. Phan*, P. Zhang, Y. D. Zhang, K. Tarigan and S. C. Yu
Department of Physics, Chungbuk National University, Cheongju 361-763, South Korea

In recent years, wide band-gap semiconductors doped with transition metals, called dilute magnetic semiconductors (DMSs), have attracted intensive interest[1-3]. These materials are applicable to spin-based electronic and optoelectronic devices, such as spin-LEDs and spin-based transistors[2]. Among DMSs, two systems of Mn- and Co-doped ZnO are promising candidates, which were predicted to be ferromagnets above room temperature[1,3]. Experimentally, it has been revealed that their magnetic properties are strongly dependent on fabrication conditions, sample processing, concentration of dopants, and structure of materials (i.e., thin films, nanostructures or polycrystalline ceramics). Depending on the variation of these factors, Mn- and Co-doped ZnO compounds can be ferromagnetic, antiferromagnetic, spin-glass, or paramagnetic. The nature of these phenomena has not been clarified yet. To gain more insight into this problem, we have prepared polycrystalline Zn1-xCoxO ceramics, and studied their magnetic properties. The explanation of obtained results is based on structural and Raman-scattering investigations.

Polycrystalline samples of $Zn_{1-x}Co_xO$ with x = 0.02, 0.05, 0.1, 0.2, and 0.3 were prepared by conventional solid reaction. High-purity powders of ZnO and Co_3O_4 combined stoichiometrically (in atomic percentage) were mixed well by using an agate mortar and a pestle. These mixtures were then calcined at 800℃ for 12hrs in air. After the calcining process, they were ground and re-annealed at 900℃ with the conditions similar to the above. Finally, the mixtures were re-ground andpressed into pellets, and then annealed at 1000℃ for 24 hrs. A spinel sample of $ZnCo_2O_4$ was also prepared with the same route, but annealed at 1200℃. The quality of the obtained products was checked by x-ray diffraction (XRD, Philips X'Pert).Raman scattering measurements were performed on Renishaw spectrometers using excitation wavelengths of 488 and 325 nm.Magnetic measurements were performed on a superconducting quantum interference device (SQUID).

XRD patterns reveal that prepared $Zn_{1-x}Co_xO$ samples are not single phase. Besides XRD peaks coming from the main phase of thewurtzite structure, there is the presence of a secondary phase of $ZnCo_2O_4$ spinel, which develops with increasing Co content. Studies of Raman scattering spectra with an excitation wavelength of 488 nm have indicated the appearance of six additional modes apart from conventional modes from the wurtzite structure of ZnO. The analysis of these additional modes indicates that three modes (denoted as SP1-3) are from the $ZnCo_2O_4$ spinel structurewhile the others (denoted as AM1-3) are due to an incorporation of Co dopants into the ZnO host lattice, see Fig. 1. Interestingly, under an excitation of 325 nm, we have observed the longitudinal optical (LO) phononand its overtones up to the sixth order, indicating a large deformation potential existing in our samples.

Magnetic measurements at room temperature exhibit the magnetic properties of $Zn_{1-x}Co_xO$ depending strongly on Co content, and there is the magnetic-phase separation. With Co concentrations of x 0.1, the compounds reveal the paramagnetic behavior. The increase in Co concentration higher than this value ($x > 0.1$) makes $Zn_{1-x}Co_xO$ samples exhibited the ferromagnetic feature, see Fig. 2. Comparing with the magnetic data of reference samples

of $ZnCo_2O_4$ and Co_3O_4 (the inset of Fig. 2), we believe that the ferromagnetism in $Zn_{1-x}Co_xO$ with x > 0.1 is generated from unreacted Co_3O_4 particles, and both phases of the Zn-Co-O wurtzite and ZnCoO4 spinel do not exhibit the ferromagnetic order. The fabrication of $Zn_{1-x}Co_xO$-based spintronic materials is thus impossible in our case.

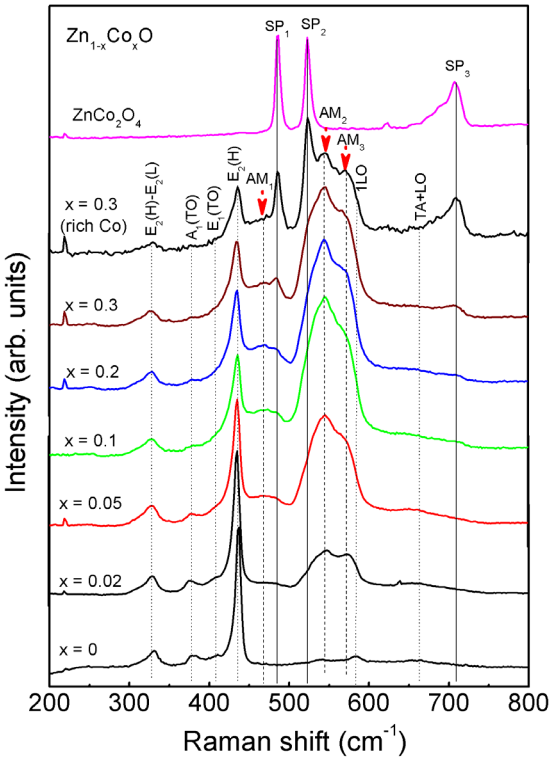

Fig. 1. RS spectra of $Zn_{1-x}Co_xO$ samples and $ZnCo_2O_4$ excited by 488 nm. Dotted lines are conventional modes from the ZnO wurtzitestructure. Solid lines (SP_{1-3}) are from $ZnCo_2O_4$ spinel while dashed lines (AM_{1-3}) are additional modes caused by an incorporation of Co dopants into the ZnO host lattice.

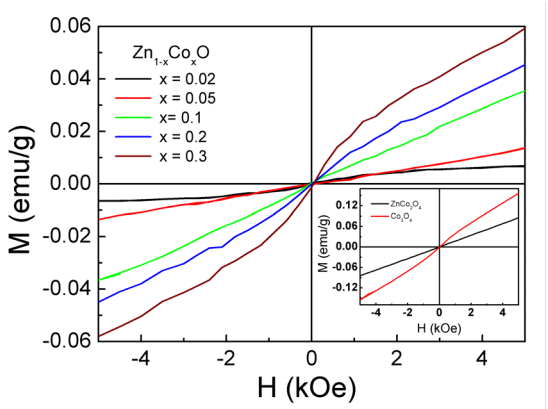

Fig. 2. M-H curves of polycrstalline $Zn_{1-x}Co_xO$ ceramics measured at 300 K. The inset shows M-H curves of Co_3O_4 and $ZnCo_2O_4$.

References

[1] I. Dietl, H. Ohno, F. Matsukura, J. Cibert and D. Ferrand, "*Zener model description of ferromagnetism in zinc-blende magnetic semiconductors*," Science, vol. 287, pp. 1019-1022, Feb. 2000.

[2] T. Bland, K. Lee, S. Steinmuller, "*The spintronics challenges*," Phys. World, pp. 24-28, Jan. 2008.

[3] X. F. Wang, J. B. Xu, B. Zhang, H. G. Yu, J. Wang, X. X. Zhang, J. G. Yu, and Q. Li, "*Signature of intrinsic high-temperature ferromagnetism in cobalt-doped zinc oxide nanocrystals*," Adv. Mater., vol. 18, pp. 2476-2480, Aug. 2006.