

On the origin of ferromagnetism and weak spin interaction in $\text{Zn}_{1-x}\text{Co}_x\text{O}$

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The ZnO-based diluted magnetic semiconductor (DMS) is a promising candidate for realizing a ferromagnetic semiconductor with high Curie temperature. $\text{Zn}_{1-x}\text{Co}_x\text{O}$ with the Curie temperature higher than room temperature has become a model example for diluted magnetic oxides (DMOs)¹. However, the magnetic properties of $\text{Zn}_{1-x}\text{Co}_x\text{O}$ reported so far by different research groups are quite contradictory. Some papers found that $\text{Zn}_{1-x}\text{Co}_x\text{O}$ is ferromagnetic at room temperature but the magnetic moment is much smaller than expected. On the other hand, there are also works that report on the absence of ferromagnetism in these materials. Here we present the results of experimental study of the weak ferromagnetism in $\text{Zn}_{1-x}\text{Co}_x\text{O}$. The temperature dependencies of magnetization and thermal diffusivity have been conducted. Diluted magnetic semiconductor $\text{Zn}_{1-x}\text{Co}_x\text{O}$ ($x = 0.03$) thin films were deposited on Si (100) substrates by using ultrasonic spray pyrolysis. Aqueous solutions of zinc acetate (0.5 mol/l) and cobalt acetate (0.5 mol/l) were used as sources of Zn and Co, respectively. The substrate temperature was set at 400 °C, and the thickness of $\text{Zn}_{0.97}\text{Co}_{0.03}\text{O}$ films were about 200 nm. The magnetization dependencies on the magnetic field, $M(H)$ curve, and on the temperature, $M(T)$ curve, were measured using a superconducting quantum interference device (SQUID) magnetometer. The thermal diffusivity measurements were made using photothermal method. The structural analysis of Co doped ZnO films were carried out by using an X-ray diffractometer (XRD, Rigaku mini flex).

The XRD pattern for $\text{Zn}_{0.97}\text{Co}_{0.03}\text{O}$ films grown on Si substrates show only peaks related to the wurtzite structure without any secondary phase up to the detection limit of the instrument. Magnetization $M(H)$ measurements show a hysteresis loop which indicates an existence of ferromagnetism in $\text{Zn}_{0.97}\text{Co}_{0.03}\text{O}$. However, the magnetic moment per Co ion is much lower than expected. A comparison of $M(T)$ measured at zero-field-cooled (ZFC) and field-cooled (FC) conditions shows a superparamagnetic like behavior and the blocking temperature is about 130K. Temperature dependence of the thermal diffusivity of $\text{Zn}_{0.97}\text{Co}_{0.03}\text{O}$ shows a pronounced lambda-shaped minimum at 130K, which indicates an existence of a second-order phase transition at this temperature. The low value of phase transition entropy shows that only a small part of Co ions is involved in ferromagnetic ordering. The weak ferromagnetism in $\text{Zn}_{0.97}\text{Co}_{0.03}\text{O}$ with a Curie temperature of 130 K is ascribed to an uncompensated magnetic moment of CoO nanoclusters with radii of about 1 nm existing at the surface^{2,3}.

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