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SPIN GLASS BEHAVIOR AND ANTIFERROMAGNETIC EXCHANGE COUPLING IN LASER-DEPOSITED $Zn_{1-x}Co_xO$ THIN FILMS

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Recently, we have seen a rapid advance in the evolving field of spin electronics (or spintronics). In spintronics, some feasibilities of new electronic applications utilizing spin degree of freedom have been explored. Diluted magnetic semiconductors (DMSs) are promising materials for spintronics because they have both charge and spin degree of freedom in a single substance. DMSs are refereed to semiconductor alloys in which some atoms are randomly substituted for by magnetic atoms. ZnO-based DMSs are currently attracting much attention due to theoretical predictions of room temperature ferromagnetism. Here, we report on the magnetization measurements of $Zn_{1-x}Co_xO$ films to reveal the nature of magnetism and exchange coupling in this DMS.

Pulsed laser deposition using KrF excimer laser pulses (248 nm, 5 Hz) with a fluence of 1.5 J/cm^2 was employed to grow $Zn_{1-x}Co_xO$ (x = 0.05–0.3) films on sapphire (0001) substrates at 600 °C under an oxygen pressure of 1×10^{-5} Torr. X-ray diffraction measurements confirmed that the films had c-axis oriented wurtzite structure without any second phase. The Co content in the film was determined by electron probe microanalysis. The magnetization was measured using a dc superconducting quantum interference device (SQUID) magnetometer (Quantum Design MPMS) with the magnetic field applied parallel to the film surface.

Figure 1 shows the field-cooled (FC) and zero-field-cooled (ZFC) magnetizations for $Zn_{1-x}Co_xO$ (x = 0.2, 0.25) films in various magnetic fields (H). It was found that, for $x \geq 0.2$, the FC and ZFC magnetizations at each magnetic field start to differ below a certain temperature, depending on the value of H, as shown in Fig. 1. Such a feature for magnetization has frequently been observed in other II-VI DMSs including $Zn_{1-x}Mn_xO$, indicating spin-glass behavior. The temperature at which FC and ZFC magnetizations merge increases with decreasing H. This merging temperature in the limit of $H\to0$, the spin-freezing temperature T_f, is about 8 K and 12 K for x = 0.2 and 0.25, respectively. On the other hand, for $x < 0.2$, no significant difference in FC and ZFC magnetizations was observed, at least, down to 2 K.

The temperature dependence of the FC magnetization for x = 0.05–0.3 displays a Curie-Weiss behavior, $M/H = C_0 x/(T - \Theta_0 x)$, at high temperatures, as shown in Fig. 2. By fitting the data to this formula, the Curie-Weiss temperature $\Theta_0 = -810$ K was obtained. This large negative value of Θ_0 indicates a strong antiferromagnetic exchange coupling in $Zn_{1-x}Co_xO$. The value of J_1/k_B, where J_1 is the effective exchange integral between nearest-neighbor Co^{2+} ions and k_B is the Boltzmann constant, is then estimated to be –27 K from the relation $\Theta_0 = 2S(S+1)zJ_1/3k_B$, where z is the number of the nearest-neighbor cations (z = 12 for the wurtzite structure) and S is the value of the spin for the Co^{2+} ions, using the value of $S = 3/2$ for the Co^{2+} ions.

In conclusion, we examined magnetic properties of the oxide-diluted magnetic semiconductor ZnCoO thin films fabricated by pulsed laser deposition. Contrary to the theoretical predictions by Sato and Katayama-Yoshida, ZnCoO exhibit paramagnetic or spin glass behavior having a large Curie–Weiss temperature corresponding to a strong aniferromagnetic exchange coupling found in other Co-doped II–VI DMSs.

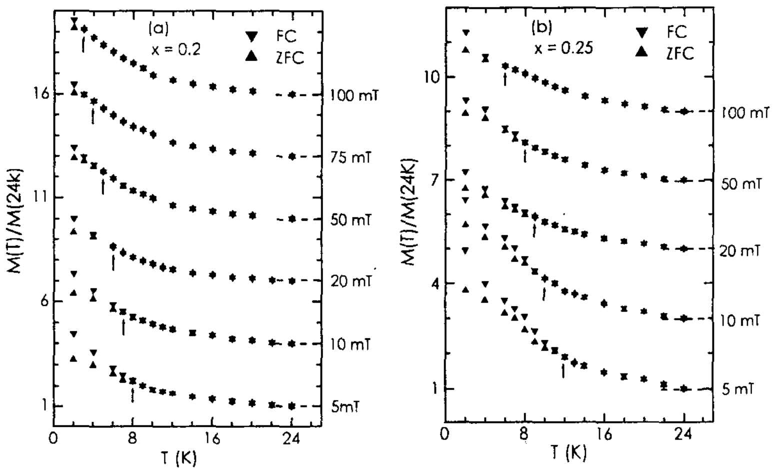

Fig. 1. Field-cooled (FC) and zero-field-cooled (ZFC) magnetizations normalized at 24 K for (a) x = 0.2 and (b) x = 0.25 in various magnetic fields. The curves are shifted vertically as represented by the dotted ticks around 24 K for clarity.

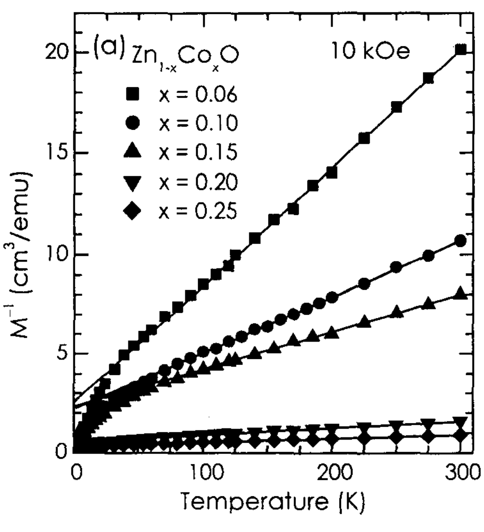

Fig. 2. Inverse magnetization of laser-deposited ZnCoO films as a function of temperature for various Co concentrations. The solid lines represent the limiting high-temperature Curie–Weiss behavior.