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Ferromagnetism in II–VI diluted magnetic semiconductor \( \text{Zn}_{1-x}\text{Cr}_x\text{Te} \)

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Magnetic and transport properties of an epitaxial film of ferromagnetic II–VI diluted magnetic semiconductor (DMS) \( \text{Zn}_{1-x}\text{Cr}_x\text{Te} (x = 0.035) \) were investigated. The Curie temperature \( T_C \) of the film was about 15 K, which is the highest among the reported ferromagnetic II–IV DMS. Hall effect measurements at room temperature showed a hole concentration \( p \) of about \( 1 \times 10^{15} \text{ cm}^{-3} \), which is several orders lower than that reported for carrier-induced ferromagnetic \( \text{Zn}_{1-x}\text{Mn}_x\text{Te} \). These results suggest that a ferromagnetic superexchange interaction between Cr ions is responsible for the observed ferromagnetism in \( \text{Zn}_{1-x}\text{Cr}_x\text{Te} \). \( \text{DOI: } 10.1063/1.1452649 \)

I. INTRODUCTION

Ferromagnetic diluted magnetic semiconductors (DMS) are important materials for use in spin electronics devices. Carrier-induced ferromagnetism has been reported in Mn-doped DMS such as \( \text{Ga}_{1-x}\text{Mn}_x\text{As} \)\(^2\) and \( p \)-doped \( \text{Zn}_{1-x}\text{Mn}_x\text{Te} \).\(^3\) To achieve the ferromagnetism in Mn-doped DMS, a large quantity of carriers (holes) is necessary to overcome an antiferromagnetic superexchange interaction between Mn ions. However, it is difficult to control the conductive and magnetic properties independently in those materials. Therefore, a ferromagnetic DMS with few carriers is desirable. Cr-doped II–VI DMS is expected to meet such a demand, because a ferromagnetic superexchange interaction between Cr ions has been theoretically shown.\(^4\) Moreover, the band calculation results for Cr-doped II–VI DMS predicted that the ferromagnetic state would be stable compared to the nonmagnetic state.\(^5,6\) From the magneto-optical studies, \( \text{Zn}_{1-x}\text{Cr}_x\text{M} \) (\( \text{M} = \text{S}, \text{Se}, \text{and Te} \)) were confirmed as being DMS,\(^7\)–\(^9\) but no ferromagnetism has been reported.\(^7\)–\(^11\)

Very recently, we successfully grew epitaxial films of \( \text{Zn}_{1-x}\text{Cr}_x\text{Te} \) having a relatively high Cr concentration \( x \) up to 0.035 and confirmed its ferromagnetism.\(^12\) In this study, we report the detailed magnetic and transport properties of a ferromagnetic \( \text{Zn}_{1-x}\text{Cr}_x\text{Te} (x = 0.035) \) film.

II. EXPERIMENT

Epitaxial films of \( \text{Zn}_{1-x}\text{Cr}_x\text{Te} \) having a zinc-blende type crystal structure were grown by a molecular beam epitaxy (MBE) method. A 100 nm thick \( \text{ZnTe} \) buffer layer was first grown on the GaAs substrate at \( T_S = 300 \)°C. Then a 200 nm thick \( \text{Zn}_{1-x}\text{Cr}_x\text{Te} \) layer was grown at \( T_S = 250–300 \)°C under Te-rich conditions. A \( (2 \times 1) \) reflection high-energy electron diffraction (RHEED) pattern was observed during the growth of the \( \text{Zn}_{1-x}\text{Cr}_x\text{Te} \) films having a Cr concentration \( x \) up to 0.035. The Cr concentration \( x \) was calculated by electron probe micro-analysis (EPMA) and secondary ion mass spectroscopy (SIMS). Magnetization measurements were carried out using a superconducting quantum interference device (SQUID) magnetometer under the magnetic field \( H \) applied parallel to the film plane. The magnetization of the \( \text{Zn}_{1-x}\text{Cr}_x\text{Te} \) films was derived by subtracting the contribution from the substrate, and the magnetization of the substrate was also measured by a SQUID. The errors in the magnetization of the \( \text{Zn}_{1-x}\text{Cr}_x\text{Te} \) films are of the order of a few percent. Resistivity and Hall resistivity were measured in the van der Pauw configuration.

III. RESULTS AND DISCUSSION

Figure 1 shows the magnetization curves for a \( \text{Zn}_{1-x}\text{Cr}_x\text{Te} (x = 0.035) \) film at various temperatures \( T \). A ferromagnetic hysteresis loop is clearly observed in the curve at \( T = 5 \) K. By magnetic circular dichroism (MCD) measurements, it was confirmed that the ferromagnetic hysteresis comes from \( \text{Zn}_{1-x}\text{Cr}_x\text{Te} \), and that the film is free from magnetic precipitates.\(^12\) The hysteresis in the magnetization curve almost disappears at \( T = 10 \) K, as shown in the inset of Fig. 1, suggesting that the Curie temperature \( T_C \) of the film is about 10 K. Above \( T = 50 \) K, the curves show paramagnetic behavior. To determine \( T_C \), the Arrott-plots of the film with \( x = 0.035 \) at various \( T \) are given in Fig. 2. The plots show a linear relation in relatively high magnetic field ranges. The intercept of a linear extrapolation of \( M^2 \) at \( H = 0 \) gives a square of the spontaneous magnetization \( M_S^2 \). With increasing \( T \), \( M_S^2 \) decreases and becomes zero at around \( T = 15 \) K, which corresponds to \( T_C \). It should be noted that the obtained \( T_C \) of the film is about one order higher than that of the reported carrier-induced ferromagnetic II–VI DMS. The
$T_C$ of a Cd$_{1-x}$Mn$_x$Te quantum well,\textsuperscript{13} $p$-doped Zn$_{1-x}$Mn$_x$Te,\textsuperscript{2} and Be$_{1-x}$Mn$_x$Te (Ref. 14) are 1.8, 2.4, and 2.5 K, respectively.

Figure 3 shows the temperature dependence of susceptibility $\chi$ and the inverse susceptibility $\chi^{-1}$ for the film with $x=0.035$ in $H=10$ kOe. It has been reported that the magnetic precipitate in Zn$_{1-x}$Cr$_x$Te is a mainly NiAs-type CrTe compound whose $T_C$ is slightly above 300 K.\textsuperscript{10} No anomaly in the $\chi-T$ curve is apparent in the vicinity of the $T_C$ of CrTe, meaning that the film is free from the CrTe compound. The $\chi-T$ curve shows a large $\chi$, even at a much higher temperature than the $T_C$ estimated from the Arrott-plots, as shown in Fig. 2. Fitting the $\chi^{-1}-T$ curve using the Curie–Weiss law, the paramagnetic Curie temperature obtained is about 150 K, which is significantly higher than a ferromagnetic $T_C$. These results indicate that the magnetic short-range order remains even far above $T_C$.

The temperature dependence of resistivity $\rho$ for the film with $x=0.035$ is shown in Fig. 4. Down to $T=50$ K, the curve shows semiconductive behavior. Resistivity becomes too high for a reliable measurement below about $T=50$ K. The value of $\rho$ is several orders higher than that of carrier-induced ferromagnetic DMS such as Ga$_{1-x}$Mn$_x$As,\textsuperscript{2} and Zn$_{1-x}$Mn$_x$Te.\textsuperscript{3} From the Hall effect measurement at room temperature, $p$-type conduction is clearly observed as shown in the inset of Fig. 4. The carrier concentration $p$ is estimated to be about $1\times10^{15}$ cm$^{-3}$, which is 3–5 orders lower than that of the reported ferromagnetic DMS ($p = 10^{18}$–$10^{20}$ cm$^{-3}$). Therefore, an even higher $T_C$ in Zn$_{1-x}$Cr$_x$Te is expected by means of hole-doping.

In DMS, there are two possible magnetic interactions between localized spins. One is the carrier (hole)-induced interaction, and the other is the superexchange interaction.\textsuperscript{4,15,16} The former interaction is ferromagnetic, whereas a sign of the latter interaction depends on the magnetic transition metals used.\textsuperscript{17,18} In the case of an Mn-doped DMS, the superexchange interaction between Mn ions is antiferromagnetic.\textsuperscript{15} It should be noted that the $T_C$ of Zn$_{1-x}$Cr$_x$Te is higher than that of the carrier-induced ferromagnetic Zn$_{1-x}$Mn$_x$Te, although the $p$ of Zn$_{1-x}$Cr$_x$Te is several orders lower than that of Zn$_{1-x}$Mn$_x$Te, as mentioned.
above. This suggests that a superexchange interaction is responsible for the ferromagnetism in $\text{Zn}_{1-x}\text{Cr}_x\text{Te}$, which is in accordance with the theoretical results.\(^4\)

**IV. CONCLUSION**

Magnetic and transport properties of a ferromagnetic II–VI DMS, $\text{Zn}_{1-x}\text{Cr}_x\text{Te}$, have been investigated. The Curie temperature $T_C$ of 15 K for the film of $x = 0.035$ is higher than that of the other carrier-induced ferromagnetic II–VI DMS, yet the hole concentration is quite low. This suggests that a superexchange interaction is responsible for the ferromagnetism in $\text{Zn}_{1-x}\text{Cr}_x\text{Te}$.

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