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Magnetic and structural properties of MBE-grown $Zn_{1-x}Cr_xTe$ films

N Ozaki, N Nishizawa, S Kuroda and K Takita

Institute of Materials Science, University of Tsukuba, 1-1-1 Tennoudai, Tsukuba, Ibaraki, 305-8573, Japan

E-mail: ozaki@ims.tsukuba.ac.jp

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Abstract
The magnetic and structural properties of MBE-grown films of a novel diluted magnetic semiconductor (DMS), $Zn_{1-x}Cr_xTe$, were investigated. The magnetization versus magnetic field (M–H) measurement exhibited a clear hysteresis loop at low temperatures. The ferromagnetic transition temperature (T_C) estimated from the Arrott-plot analysis increased almost linearly with the Cr concentration (x) up to 275 K at $x = 0.17$. In the magnetization versus temperature (M–T) measurement, irreversibility between the zero-field-cooled (ZFC) and field-cooled (FC) processes was observed. This behaviour, which is typically observed in a magnetic random system such as the spin-glass or superparamagnetic phase, is considered to be related to the local structural inhomogeneity observed by high resolution transmission electron microscopy.

1. Introduction

Cr doped II–VI DMSs have been attracting a great deal of attention since the ferromagnetic transition was reported by several groups [1–3]. In particular, Saito *et al* claimed that $Zn_{1-x}Cr_xTe$ with $x = 0.20$ exhibited the ferromagnetic transition around room temperature [1]. However, the mechanism of the ferromagnetism of this novel DMS is still under discussion. These Cr-doped II–VIs are highly resistive, suggesting that the origin of the ferromagnetism is not carrier-mediated interaction as in Mn-doped III–V DMSs [4].

In the present work, we have investigated the magnetic and structural properties of $Zn_{1-x}Cr_xTe$ films grown by MBE. The detailed magnetization measurements revealed that the grown films exhibited ferromagnetic properties in general, but having the features of the magnetic random system at the same time. The origin of the magnetic randomness is considered in relation to the structural defects observed by high resolution transmission electron microscopy (HRTEM).

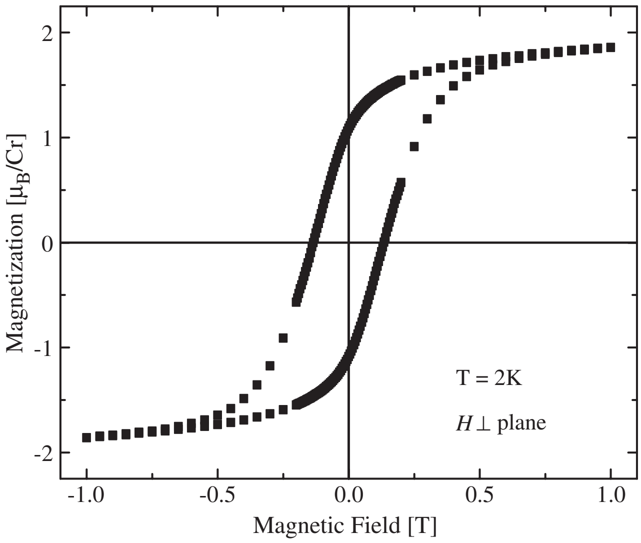

Figure 1. The M–H curve of the $Zn_{1-x}Cr_xTe$ film with $x = 0.17$ at 2 K. The magnetic field was applied perpendicular to the film plane.

2. Experimental details

$Zn_{1-x}Cr_xTe$ films were grown by MBE. A ZnTe buffer layer of approximately 0.72 μm in thickness was grown on a semi-insulating GaAs(100) substrate and a $Zn_{1-x}Cr_xTe$ layer was grown successively on it. In the growth procedure of both layers, the flux ratio of Te to Zn was kept at about 2:1 and the growth temperature was kept at 300 °C. The Cr concentration was measured using an electron probe micro-analyser (EPMA). The magnetization of the grown films was measured by a superconducting quantum interference device (SQUID) magnetometer with magnetic fields perpendicular to the film plane. The local crystal structure was examined using a high resolution transmission electron microscope.

3. Results and discussion

The magnetization measurement revealed ferromagnetic behaviours of the $Zn_{1-x}Cr_xTe$ films in the range of Cr concentrations $x = 0.01$–0.17 at low temperatures. A typical example of the magnetization against magnetic field (M–H) curve is shown in figure 1 for the sample with $x = 0.17$ at 2 K. Diamagnetic response from the GaAs substrate is subtracted. The clear hysteresis loop observed indicates the ferromagnetic interactions between Cr ions in the film. The ferromagnetic transition temperature (T_C) estimated from the Arrott-plot analysis increased almost linearly with the Cr concentration with the maximum $T_C = 275$ K at $x = 0.17$. This linear relation between T_C and x, the extrapolation of which gives $T_C = 300$ K at $x = 0.20$, is consistent with that reported by Saito *et al* [1]. Figure 2(a) shows the magnetization versus temperature (M–T) curve of the same $Zn_{1-x}Cr_xTe$ film with $x = 0.17$ at 500 Oe. As shown in the figure, the irreversibility between the zero-field-cooled (ZFC) and field-cooled (FC) processes and a cusp-like behaviour of the ZFC curve were observed. These behaviours are typically seen in a magnetic random system such as superparamagnetism or spin glass. In addition, the paramagnetic Curie temperature Θ derived from the temperature dependence of the susceptibility was much higher than T_C ($\Theta \approx 310$ K in figure 2(b)). Similar

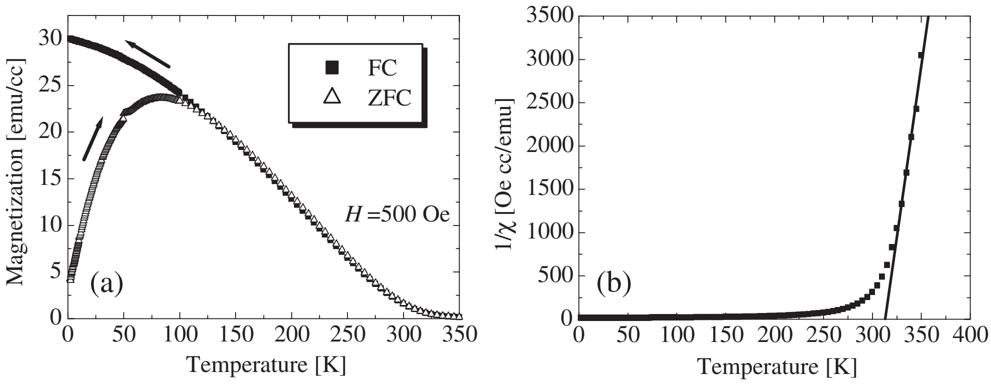

Figure 2. (a) ZFC and FC M–T curves of the $Zn_{1-x}Cr_xTe$ film with $x = 0.17$. Arrows represent the directions of the temperature sweeps. (b) $1/\chi$ versus temperature curve of the $Zn_{1-x}Cr_xTe$ film with $x = 0.17$. The linear fitting to the data at high temperatures gives the Curie–Weiss temperature $\Theta \approx 310$ K.

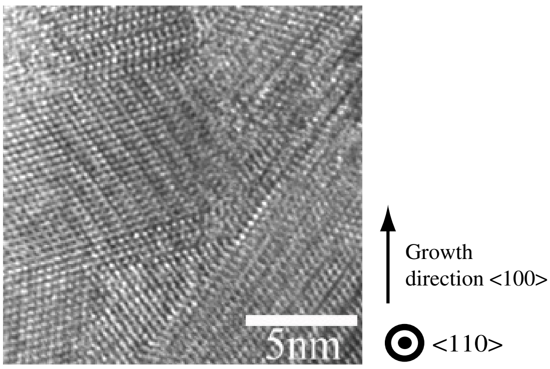

Figure 3. The cross-sectional HRTEM image of the $Zn_{1-x}Cr_xTe$ film with $x = 0.17$.

behaviours were observed in all the $Zn_{1-x}Cr_xTe$ films in the range of $x = 0.02$–0.17. All these observations strongly suggest that $Zn_{1-x}Cr_xTe$ has the character of superparamagnetism. In order to investigate the origin of the superparamagnetic behaviours, we have performed cross-sectional HRTEM observation of the grown films. Figure 3 shows the TEM image of the film with $x = 0.17$. As seen in the image, the crystal is not a perfectly epitaxial layer, but there exist nano-scale domains of different crystal orientations, whereas no apparent precipitates of the second phase are seen. In addition, many stacking faults are observed in the respective domains. A stacking fault of the same type was observed even at a lower Cr concentration of $x = 0.08$ [3]. Therefore the stacking fault is created due to the Cr doping and its formation in a high density causes the creation of domains of different crystal orientations. It is considered that the polycrystalline-like structure of the grown films at high Cr concentrations is related to the observed magnetic behaviours which are characteristic of the magnetic random system.

4. Summary

We have grown $Zn_{1-x}Cr_xTe$ films by MBE and investigated their magnetic and structural properties. The magnetic properties of the grown films were ferromagnetic in general,

but exhibited features which were typical of a magnetic random system at the same time. HRTEM observations of the films revealed the polycrystalline-like structure in $Zn_{1-x}Cr_xTe$ with $x = 0.17$. This structural inhomogeneity could be related to the observed magnetic behaviours.

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