Magnetic measurements on ferromagnetic behavior in the bulk II-VI diluted magnetic semiconductor Zn1-xCrxTe

T. M. Pekarek D. J. Arenas B. C. Crooker

I. Miotkowski A. K. Ramdas

Magnetic measurements on ferromagnetic behavior in the bulk II–VI diluted magnetic semiconductor $Zn_{1-x}Cr_xTe$

T. M. Pekarek[a] and D. J. Arenas
Department of Chemistry and Physics, University of North Florida, Jacksonville, Florida 32224

B. C. Crooker
Department of Physics, Fordham University, Bronx, New York 10458

I. Miotkowski and A. K. Ramdas
Department of Physics, Purdue University, West Lafayette, Indiana 47907

(Presented on 8 January 2004)

Magnetic measurements on the ferromagnetic behavior in the bulk II–VI diluted magnetic semiconductor $Zn_{1-x}Cr_xTe$ were made on two $x=0.0033$ single crystals taken from different regions of the same boule. Ferromagnetism was verified in both samples by an Arrott plot analysis with a transition temperature at 365 K (well above room temperature). For both samples at room temperature, the coercive field is ~ 0.0100 T and the remanent magnetization is 23% of the saturated value. The similarity in the observed ferromagnetic behavior between the two samples suggests that a stable Cr_yTe_z or possibly $Zn_xCr_yTe_z$ precipitate phase is responsible, although a Cr-rich region in the bulk $Zn_{1-x}Cr_xTe$ itself cannot currently be conclusively ruled out as the source. © *2004 American Institute of Physics.* [DOI: 10.1063/1.1689431]

I. INTRODUCTION

The II–VI diluted magnetic semiconductors (DMS) have been the topic of many studies over the past two decades.[1] Many of these studies were driven by an interest in combining semiconductors with magnetic materials. The field of spintronics,[2] in particular, would be significantly advanced by the discovery of new ferromagnetic semiconducting materials with transitions significantly above room temperature.

In previous work, we reported on a minority ferromagnetic phase in a bulk $Zn_{1-x}Cr_xTe$ crystal grown by the vertical Bridgman method.[3] We found a transition temperature slightly above 300 K for the ferromagnetic phase. At the time, we ascribed the ferromagnetic signal to a pure CrTe minority phase. For the purpose of this previous work, the observed ferromagnetic signal was a background feature that was characterized sufficiently to allow for its subtraction.

However, recent work by Saito *et al.* on molecular beam epitaxy (MBE) grown epitaxial films of $Zn_{1-x}Cr_xTe$ found that an $x=0.20$ sample was ferromagnetic[4,5] with a similar transition temperature and magnetic properties. This result, combined with other work by Saito *et al.* on the ferromagnetic properties of an $x=0.035$ $Zn_{1-x}Cr_xTe$ epitaxial film with a T_c around 15 K,[6,7] raised the possibility that our minority phase was instead a region of enhanced Cr concentration within our bulk $Zn_{1-x}Cr_xTe$ crystal.

In this paper we report on magnetization measurements on two additional samples removed from different regions of the original boule. An Arrott plot analysis[8] was conducted to confirm the ferromagnetic behavior and determine the Curie temperature. Additional features of the ferromagnetic behavior were characterized for comparisons between different bulk crystals and the reported epitaxial films.

II. EXPERIMENTAL DETAILS

Single-crystalline $Zn_{1-x}Cr_xTe$ samples were grown by the vertical Bridgman method. Three samples (A, B, and C) were taken from the same boule, with a nominal concentration of $x=0.005$. Atomic absorption spectroscopy (AAS) was performed on sample A yielding a value of $x=0.0035 \pm 0.0005$.[3] With this concentration for sample A as a reference, the concentration of samples B and C is determined using the ratio of the magnetization at 5 K and 6 T. This yields values for the concentration for samples B and C of $x=0.0033$ and $x=0.0033$, respectively.

Magnetization measurements on sample A were reported previously at temperatures between 2 and 300 K and in fields up to 6 T.[3] Samples B and C were measured in a new Quantum Design MPMS XL7 superconducting quantum interference device (SQUID) magnetometer at temperatures between 2 and 400 K and in fields up to 7 T. The diamagnetic signal due to the host ZnTe has been subtracted.

It is known that $Zn_{1-x}Cr_xTe$ crystals preferentially cleave along the (110) direction. Samples A, B, and C were all mounted along this cleaved direction. The (110) direction was verified by the standard Laue diffraction technique for samples A and C.

III. EXPERIMENTAL RESULTS AND DISCUSSION

Hysteresis loops in the magnetization for samples B and C were taken at a series of temperatures from 2 to 400 K with a maximum field of ± 7 T. Representative hysteresis loops for sample B are shown in Fig. 1(a) at 5, 300, and 400

[a]Electronic mail: tpekarek@unf.edu

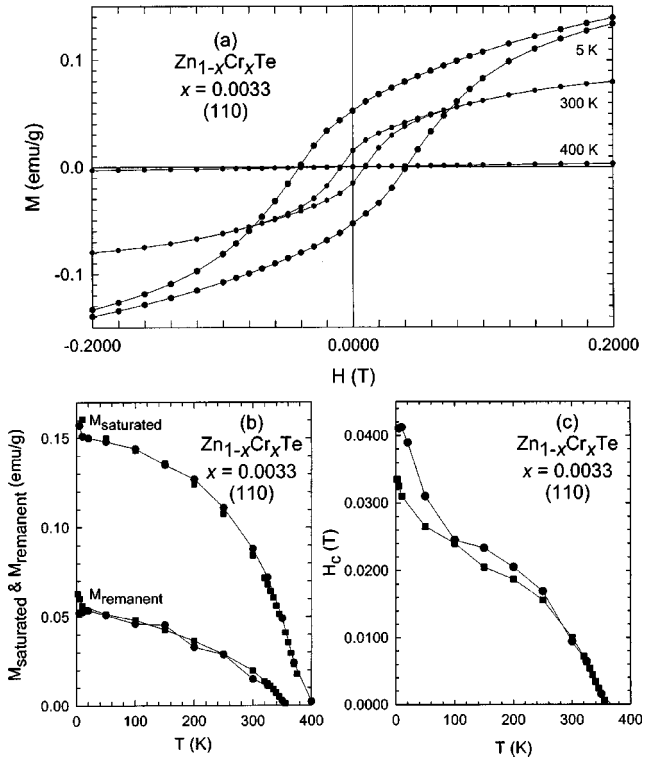

FIG. 1. (a) Magnetization vs field measurements for $Zn_{1-x}Cr_xTe$ ($x=0.0033$) at 5, 300, and 400 K are shown as circles for sample B. The lines are a guide to the eye. Hysteresis is clearly visible at 5 and 300 K but is not at 400 K. (b) The saturated and remanent magnetization vs temperature show the same functional behavior for both samples. The data for sample C have been scaled in Fig. 1(b) by the ratio of the 0 K saturated ferromagnetic signals, a factor of 2.62, to facilitate a comparison with sample B. (c) The coercive field is plotted vs temperature for samples B and C. Samples B and C are shown as circles and squares, respectively, in (b) and (c).

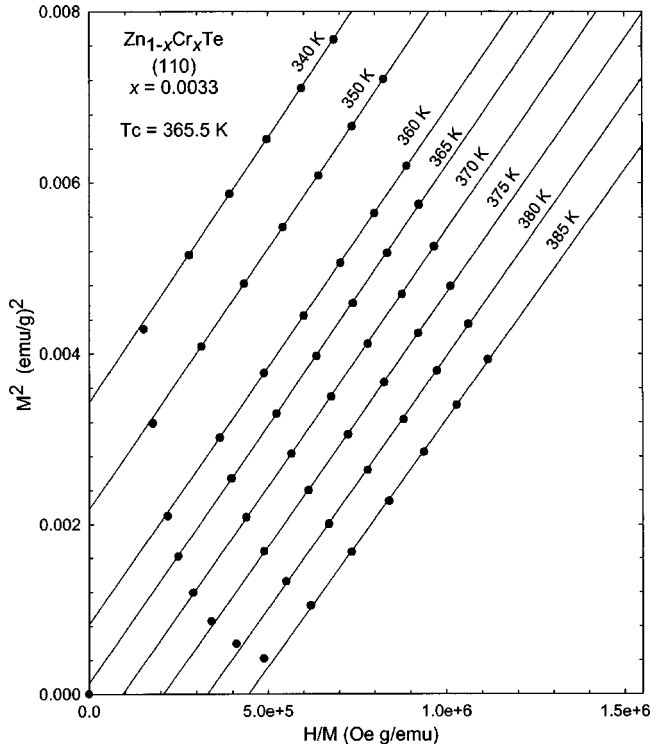

FIG. 2. Magnetization data for sample B taken at 340–385 K in fields between 1 and 7 T are shown as circles and plotted as M^2 vs H/M. At T_c for a ferromagnetic transition, the intercept $1/\chi_0$ on the H/M axis passes through the origin. The lines are linear fits to the high field data at a set temperature. For this sample T_c is 365.5 K.

K. Hysteresis is clearly observed at 300 K and persists up to 350 K. Above ~355 K, any hysteresis cannot be resolved within the uncertainties of the instrument.

Linear fits to the descending branch of the hysteresis loops between 0.8 and 1.5 T were used to determine the saturated ferromagnetic signal of each sample. Extrapolating to 0 K, the saturated ferromagnetic signal is 0.055, 0.152, and 0.058 emu/g for samples A, B, and C, respectively. The saturated ferromagnetic signal as a function of temperature for samples B and C is shown in Fig. 1(b) [note the data for sample C in Fig. 1(b) have been scaled by the ratio of the 0 K saturated ferromagnetic signals, a factor of 2.62, to facilitate a comparison with sample B]. The remanent magnetization for both samples is also shown in Fig. 1(b). After scaling the data for sample C, both samples are seen to exhibit the same temperature dependence for the remanent and saturated ferromagnetic signal.

The coercive field H_c versus temperature for samples B and C is shown in Fig. 1(c). H_c is large at low temperatures and persists up to T_c. The coercive field as a function of temperature follows the same general behavior from T_c down to 100 K. Below 100 K, H_c varies by up to 33% at 10 K where H_c reaches 0.0412 T for sample B.

To confirm that the hysteresis is due to ferromagnetism and to determine the transition temperature T_c, we analyzed magnetization data from 1 to 7 T following the work of Arrott.[8] For temperatures around 365 K, we observed straight lines in a plot of M^2 vs H/M. Data for sample B are shown in Fig. 2. As we approach 365 K from above and below, the intercept $1/\chi_0$ on the H/M axis approaches the origin. Slight curvature toward the origin can be observed in data taken below ~1 T at 340 and 385 K as expected. Extrapolating $1/\chi_0$ vs T (Fig. 3), the value of $T_c=365.5$ K is

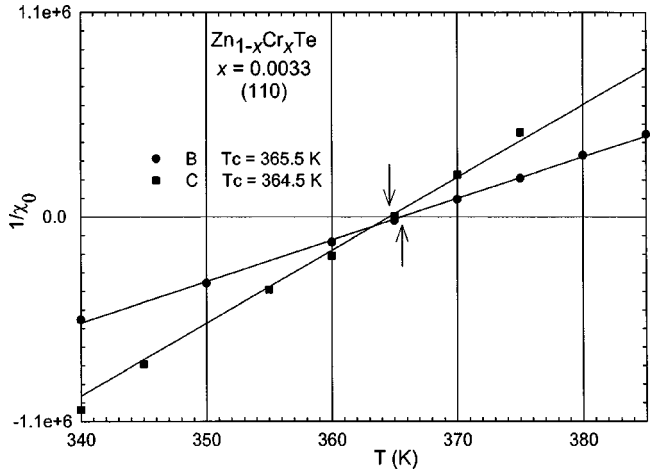

FIG. 3. Intercept $1/\chi_0$ for each set temperature in Fig. 2 is plotted as circles vs set temperature. The temperature where $1/\chi_0$ goes to zero (the x intercept) corresponds to T_c. For sample B, shown by the circles, $T_c=365.5$ K. The corresponding data for sample C are plotted as squares and yield $T_c=364.5$ K.

obtained from the point at which $1/\chi_0$ goes to zero. Similar data were observed for sample C (plotted as squares in Fig. 3) giving $T_c = 364.5$ K.

The magnetization in 0.6 T above T_c was also plotted as H/M vs T (not shown). From a Curie–Weiss fit to the data between 380 and 400 K, we obtain a temperature intercept of 365.1 and 364.7 K for samples B and C, respectively. These values are in excellent agreement to the values of T_c determined from the Arrott plot analysis.

Having established that ferromagnetism exists in these bulk $Zn_{1-x}Cr_xTe$ crystals with a transition temperature at 365 K (well above room temperature), the obvious remaining issue is to determine exactly what within the crystal is responsible for the ferromagnetic signal. Is the ferromagnetism due to regions of enhanced Cr concentration in the bulk II–VI DMS $Zn_{1-x}Cr_xTe$ itself or is a precipitate within the host II–VI DMS responsible? Either way, we have a potentially useful system. If regions within the bulk II–VI DMS $Zn_{1-x}Cr_xTe$ itself are ferromagnetic, then the higher T_c implies local Cr concentrations exceeding the value of $x = 0.20$ obtained by Saito *et al.* for the epitaxial film. Instead, if the observed ferromagnetism is due to precipitates of Cr_yTe_z[9] or possibly $Zn_xCr_yTe_z$ then we have shown it is possible to obtain ferromagnetic precipitates in a II–VI DMS host that was shown by Saito *et al.*[4] to also exhibit ferromagnetism. In this case, one can envision possible spintronic applications based on the interplay between carriers, ferromagnetism in the DMS, and ferromagnetic precipitates.

In comparing the two samples, it is worth emphasizing that the fraction of Cr atoms contributing to the ferromagnetic and singlet magnetization features is significantly different for samples B and C. Sample B has 42% of the Cr contributing to the ferromagnetic signal while sample C has only 16%. And yet, the ferromagnetic behavior exhibited by the two samples is very similar. This suggests that the observed behavior arises from a stable precipitate phase that is compatible with the host $Zn_{1-x}Cr_xTe$ lattice. It is known that such ferromagnetic precipitates can be formed in a host semiconductor. For example, ferromagnetic Fe_3GaAs clusters have been observed in GaAs where Fe_3GaAs grains form well-defined boundaries canted with respect to the GaAs host.[10–12] Such a stable stoichiometry within the semiconductor host could form in regions throughout the boule and exhibit the same T_c and temperature dependence for the saturated and remanent magnetization. Because of different cluster sizes, one might expect the temperature dependence on H_c to vary somewhat throughout the boule.

Ferromagnetism arising from regions of enhanced Cr concentration within the bulk $Zn_{1-x}Cr_xTe$ is unlikely to explain the sharp T_c and similar magnetic properties we observe in these samples. One would expect that concentration gradients throughout the boule would produce different values of T_c in different regions. In the present case, it is difficult to envision finding two samples taken from different locations within a boule with a nominal concentration of $x = 0.005$ containing the same local concentration in excess of $x = 0.20$ that would be required to produce ferromagnetic regions with values for T_c that are so close to the same value.

IV. CONCLUSIONS

Ferromagnetism was verified in two samples of the bulk II–VI diluted magnetic semiconductor $Zn_{1-x}Cr_xTe$ ($x = 0.0033$) by an Arrott plot analysis with a transition temperature at 365 K (well above room temperature). For both samples at room temperature, the coercive field is ~ 0.0100 T and the remanent magnetization is 23% of the saturated value. The observed ferromagnetic behavior is believed to arise from precipitates of $Zn_xCr_yTe_z$ within the II–VI DMS host, although the bulk $Zn_{1-x}Cr_xTe$ itself cannot currently be conclusively ruled out as the source.

ACKNOWLEDGMENTS

This research was supported by the Donors of the American Chemical Society Petroleum Research Fund PRF No. 40209-B5M, a Purdue University Academic Reimbursement Grant, and by the National Science Foundation (NSF) Grant Nos. DMR-03-05653, DMR-01-02699, and ECS-01-29853.

[1] See, for example, *Semiconductors and Semimetals*, edited by J. K. Furdyna and J. Kossut (Academic, Boston, 1988), Vol. 25.
[2] S. A. Wolf, D. D. Awschalom, R. A. Buhrman, J. M. Daughton, S. von Molnar, M. L. Roukes, A. Y. Chtchelkanova, and D. M. Treger, Science **294**, 1488 (2001).
[3] T. M. Pekarek, J. E. Luning, I. Miotkowski, and B. C. Crooker, Phys. Rev. B **50**, 16914 (1994).
[4] H. Saito, V. Zayets, S. Yamagata, and K. Ando, Phys. Rev. Lett. **90**, 207202 (2003).
[5] H. Saito, V. Zayets, S. Yamagata, and K. Ando, J. Appl. Phys. **93**, 6796 (2003).
[6] H. Saito, W. Zaets, S. Yamagata, Y. Suzuki, and K. Ando, J. Appl. Phys. **91**, 8085 (2002).
[7] H. Saito, V. Zayets, S. Yamagata, and K. Ando, Phys. Rev. B **66**, 081201(R) (2002).
[8] A. Arrott, Phys. Rev. **108**, 1394 (1957).
[9] *Solid State Physics Literature Guides*, edited by T. F. Connolly (IFI/Plenum, New York, 1972), Vol. 5.
[10] J. C. P. Chang, N. Otsuka, E. S. Harmon, M. R. Melloch, and J. M. Woodall, Appl. Phys. Lett. **65**, 2801 (1994).
[11] T. M. Pekarek, B. C. Crooker, D. D. Nolte, J. Deak, M. McElfresh, J. C. P. Chang, E. S. Harmon, M. R. Melloch, and J. M. Woodall, J. Magn. Magn. Mater. **169**, 261 (1997).
[12] D. T. McInturff, E. S. Harmon, J. C. P. Chang, T. M. Pekarek, and J. M. Woodall, Appl. Phys. Lett. **69**, 1885 (1996).