Room-temperature ferromagnetism in highly Cr-doped II-Mn-VI magnetic semiconductor $Cd_{1-x-y}Mn_xCr_yTe$

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(Received 16 April 2013; revised manuscript received 26 June 2013; published 14 August 2013)

We describe structural, magnetic, and transport properties of the $Cd_{1-x-y}Mn_xCr_yTe$ ferromagnetic semiconductor alloy. Bulk $Cd_{1-x-y}Mn_xCr_yTe$ crystals containing up to 10% of Cr ($y = 0.10$) were grown by the Bridgman method and were found to be strongly p type and ferromagnetic, with the highest Curie temperature of 362 K observed for $y = 0.10$. Magnetotransport measurements reveal the presence of the anomalous Hall effect up to and above room temperature, providing evidence that the observed ferromagnetism is characteristic of bulk $Cd_{1-x-y}Mn_xCr_yTe$. Our experiment data suggest that Cr ions play a crucial role in establishing ferromagnetism in this quaternary alloy.

DOI: 10.1103/PhysRevB.88.075205 PACS number(s): 75.60.Ej, 81.10.Fq, 75.50.Pp, 75.50.Dd

I. INTRODUCTION

Since the successful demonstration of ferromagnetism in Mn-based III–V semiconductors, ferromagnetic semiconductors have been extensively studied both for their basic physics[1–3] and for the promise they hold for spintronic applications.[4,5] For such applications to be realistic, it is desirable that the semiconductor material remains ferromagnetic up to and above room temperature and that the control of the carrier concentration and mobility in the material is similar to that in typical semiconductors.[6,7]

In III–V-based ferromagnetic semiconductors such as $Ga_{1-x}Mn_xAs$, the highest Curie temperature T_C obtained to date is 191 K,[8] and exploration of various approaches to raise this value has been the focus of intense research during the past decade. One of the obstacles in reaching higher values of T_C has been the difficulty of increasing the incorporation of Mn into the III–V lattice to sufficiently high levels.[8] In this regard, one of the advantages of II–VI-based semiconductors compared to the III–V semiconductors is the high solubility of transition metal ions in II–VI compounds, which has made them particularly attractive for a range of fundamental studies.[9] In addition, ferromagnetism arising from various types of exchange interaction, including the mechanisms of double exchange[10–12] and of ferromagnetic interaction involving bound magnetic polarons,[13–15] were theoretically proposed in discussing the possible reasons for room temperature ferromagnetism in, e.g., ZnO and GaN.[16] This suggests the possibility that ferromagnetism should also occur in II–VI semiconductors such as CdTe and ZnSe containing various transition metals ions, such as Cr. In that context, several earlier studies were carried out on Cr-doped ZnTe.[6,17] So far, however, little attention has been given to II-Mn-VI alloys additionally doped with Cr, in which one can investigate the impact of Cr on the already well-understood II-Mn-VI system.[18] Recently, using magnetization and magnetic circular dichroism measurements,[19] some of us demonstrated that such $Cd_{1-x-y}Mn_xCr_yTe$ alloys with a small fraction of Cr ($y \leqslant 0.03$) are ferromagnetic above 300 K. Irie *et al.*[20] reported that Fe-doped $Cd_{1-x}Mn_xTe$ ($Cd_{1-x-y}Mn_xFe_yTe$) quaternaries also show a high-temperature ferromagnetic behavior in a sample with $x \approx 0.37$ and $y \approx 0.01$, again pointing to possibly important role transition metal ions can play in enhancing the magnetic properties of II-Mn-VI semiconductors.

In this paper we describe a comprehensive study of ferromagnetic properties of quaternary $Cd_{0.63-y}Mn_{0.37}Cr_yTe$ crystals with higher Cr concentration ($y > 0.03$). As will be shown, our results clearly indicate that Cr plays a key role in establishing ferromagnetism in this quaternary alloy.

II. SAMPLE FABRICATION AND EXPERIMENTAL PROCEDURE

The $Cd_{1-x-y}Mn_xCr_yTe$ crystals used in this paper ($x = 0.37$; $y \geqslant 0.03$) were grown by the vertical Bridgman method.[21] The composition and homogeneity of the crystals were determined by electron probe microanalysis. Crystal structure and lattice parameters of the $Cd_{1-x-y}Mn_xCr_yTe$ alloys were studied by x-ray diffraction (XRD) measurements. Magnetic properties of the quaternary systems were investigated by a superconducting quantum interference device (SQUID) magnetometer. The resistivity and carrier concentrations were measured in a physical property measurement system using a standard four-probe configuration, and the carrier type and anomalous Hall effect were studied using a homemade Hall effect measurement apparatus.

III. EXPERIMENT RESULTS

Figure 1 shows a $\theta - 2\theta$ XRD spectrum for the $Cd_{0.63-y}Mn_{0.37}Cr_yTe$ ($y = 0.10$) crystal, indicating that the alloy forms in the zinc-blende structure and providing its lattice constant. The observed spectrum does not reveal the presence of second-phase inclusions, such as NiAs-type CrTe. In addition, XRD spectra for samples with Cr concentrations lower than $y = 0.10$ (not shown) are equally "clean," suggesting that Cr is fully soluble in this II-Mn-V system up to at least $y = 0.10$. The inset in Fig. 1 shows that the lattice constant of $Cd_{0.63-y}Mn_{0.37}Cr_yTe$ obtained for a range of y studied in this paper decreases linearly with Cr concentration up to

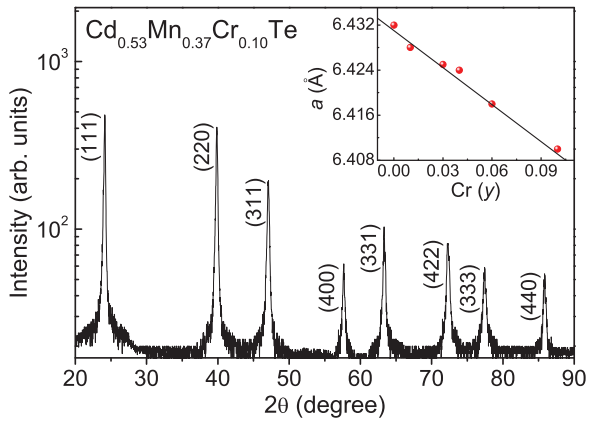


FIG. 1. (Color online) $\theta - 2\theta$ XRD spectrum for a $Cd_{0.53}Mn_{0.37}Cr_{0.10}Te$ sample, indicating that the crystal structure is zinc-blende. The inset shows the lattice constants as a function of Cr mole fraction y.

$y = 0.10$, consistent with the values of the covalent radii of Mn and Cr (1.326 and 1.280 Å, respectively) that are smaller than that of Cd (1.405 Å).[22] As an example, the sample with $y = 0.10$ has a lattice constant of 6.410 Å, 0.34% smaller than that of $Cd_{0.63}Mn_{0.37}Te$ (6.432 Å).[21] The solid line in the inset shows a least squares fit $a = (6.432 - 0.219y)$ Å. This linear dependence of the lattice parameter on y indicates that Cr ions are substitutionally incorporated at the Cd sites in the host CdTe lattice at least up to $y = 0.10$.

Figure 2 shows the temperature dependence of the magnetization of $Cd_{0.63-y}Mn_{0.37}Cr_yTe$ samples, with $y = 0.03$, 0.06, and 0.10 measured in a field of 1000 Oe. The shapes of the magnetization vs temperature curves clearly show ferromagnetic Brillouin function behavior. By fitting the empirical Brillouin function[23–25] $M_S(T) = M_0[1 - (T/T_C)^2]^{1/2}$ to the data (dashed curves), we estimate the Curie point T_C to be 352, 356, and 362 K, respectively, for these three concentrations.

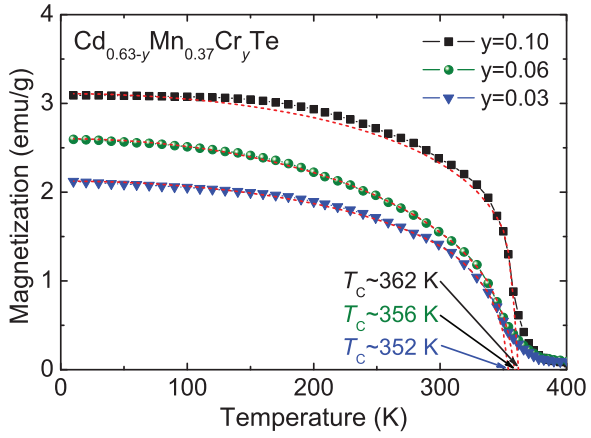
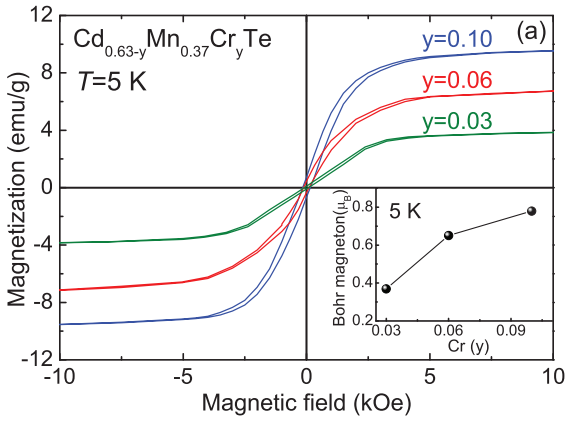


FIG. 2. (Color online) Temperature dependence of magnetization of $Cd_{0.63-y}Mn_{0.37}Cr_yTe$ samples, with $y = 0.03$, 0.06, and 0.10 measured in a field of 1000 Oe. The shapes of the magnetization vs temperature curves clearly show ferromagnetic Brillouin function behavior. By fitting the empirical Brillouin function $M_S(T) = M_0[1 - (T/T_C)^2]^{1/2}$ to the data, we estimate the Curie points T_C for these values of y to be 352, 356, and 362 K, respectively.



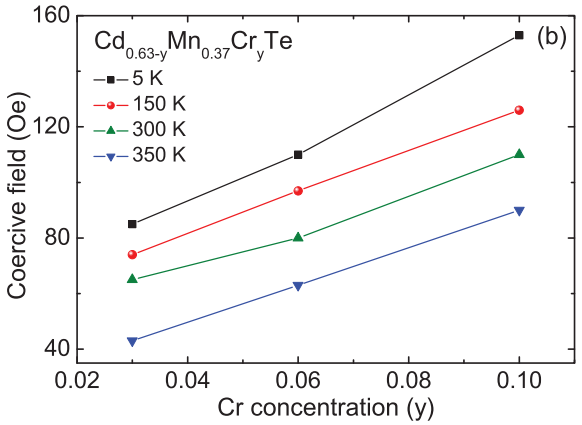


FIG. 3. (Color online) (a) Comparison of magnetic field dependence of the magnetization for $Cd_{0.63-y}Mn_{0.37}Cr_yTe$ samples, with $y = 0.03$, 0.06, and 0.10 at 5 K. The inset shows the average magnetic moment for the Mn and Cr ions in the $Cd_{0.63-y}Mn_{0.37}Cr_yTe$ samples as a function of y at 5 K. (b) Coercive fields of $Cd_{0.63-y}Mn_{0.37}Cr_yTe$ as a function of Cr content for several temperatures.

The data also clearly show that the magnetization increases with y, consistent with our conclusion that the presence of Cr is responsible for the observed ferromagnetic behavior.

Figure 3(a) shows the hysteresis loops of the $Cd_{0.63-y}Mn_{0.37}Cr_yTe$ samples, with $y = 0.03$, 0.06, and 0.10 measured at 5 K. The values of the corresponding coercive fields are shown in Fig. 3(b) for several temperatures, showing that the coercivity increases with decreasing temperature for all Cr concentrations. We also find that, similar to saturation magnetization, coercive fields increase with increasing Cr content, again indicating that Cr ions play a key role in establishing ferromagnetic order observed in these alloys. For completeness, in Fig. 4 we show magnetization M as a function of magnetic field H for $Cd_{0.63-y}Mn_{0.37}Cr_yTe$ ($y = 0.03$, 0.06, and 0.10) at various temperatures obtained from SQUID measurements. The observed hysteresis loops and the magnetization profiles clearly suggest that all samples are ferromagnetic up to and above room temperature.

Using the values of saturation magnetizations measured on the $Cd_{0.63-y}Mn_{0.37}Cr_yTe$ samples at 5 K, we calculated the average magnetic moments per magnetic ion in the system (i.e., for both Cr and Mn) for $y = 0.03$,0.06, and 0.10 to be, respectively, $0.37\mu_B$, $0.65\mu_B$, and $0.78\mu_B$, as shown in the inset in Fig. 3(a). Although these values are smaller

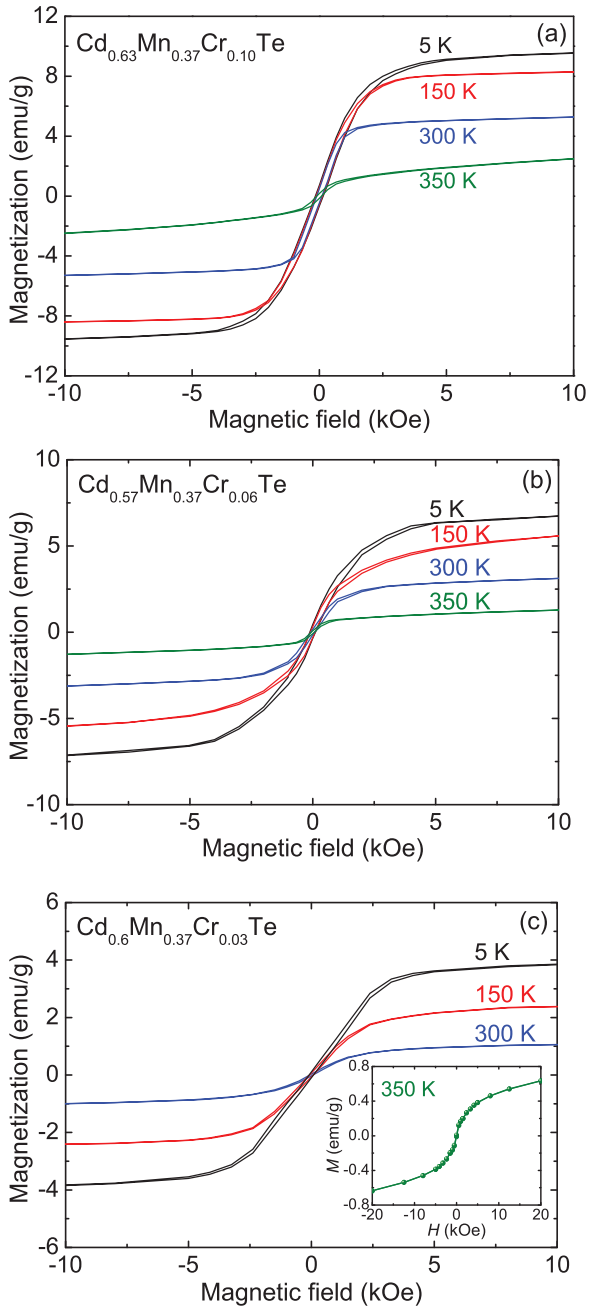


FIG. 4. (Color online) Magnetic field dependence of magnetization of $Cd_{0.63-y}Mn_{0.37}Cr_yTe$ samples with (a) $y = 0.03$, (b) $y = 0.06$, and (c) $y = 0.10$ at 5, 150, 300, and 350 K. The inset in (c) shows the M vs H data for 350 K for this sample, indicating that ferromagnetism persists to that temperature even at the relatively low value of y.

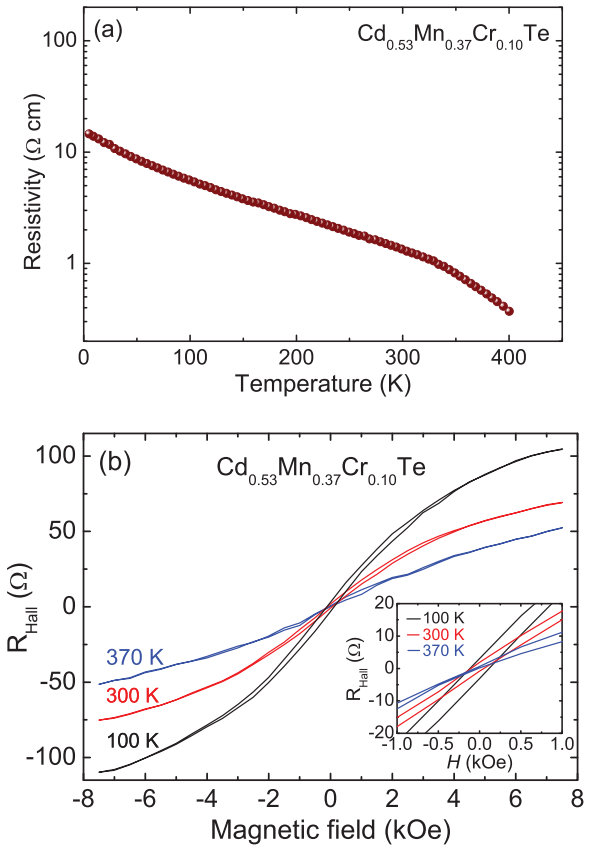


FIG. 5. (Color online) (a) Temperature dependence of resistivity ρ for the $Cd_{0.63-y}Mn_{0.37}Cr_yTe$ ($y = 0.1$) sample. (b) Anomalous Hall resistance of the $Cd_{0.63-y}Mn_{0.37}Cr_yTe$ ($y = 0.1$) sample at several temperatures. The figure also shows a zoomed-in, low-field region to clearly show the magnetic hysteresis in the Hall data.

than magnetic moments reported for other ferromagnetic semiconductors, e.g., $Zn_{1-x}Cr_xTe$ ($2.6\mu_B$),[17] $Ga_{1-x}Mn_xAs$ ($2.3\mu_B$),[26] and $Zn_{1-x}Co_xO$ ($2.0\mu_B$),[27] they indicate that Cr couples with Mn to form magnetic moments either in the form of ferromagnetic (Cr + Mn) clusters or in the form of magnetic polarons that contain both Cr and Mn ions and are bound by the holes contributed by Cr, as discussed below. It is interesting that Irie *et al.*[28] reported the coexistence of similar behavior in the $Cd_{1-x-y}Mn_xFe_yTe$ quaternaries.

Finally, in order to investigate the correlation between magnetization and carrier transport in $Cd_{1-x-y}Mn_xCr_yTe$ alloys, we performed electrical resistivity and Hall effect measurements on our samples. Figure 5(a) shows the temperature dependence of the electrical resistivity of $Cd_{0.63-y}Mn_{0.37}Cr_yTe$ ($y = 0.10$) at a zero magnetic field, showing a clearly semiconductorlike behavior. We can see a distinct slope change in resistance ~350 K, approximately corresponding to the Curie temperature of the material. It is interesting that a similar change in the slope of resistivity is reported near the Curie temperature in $Ga_{1-x}Mn_xAs$.[29]

Figure 5(b) shows the magnetic field dependence of the Hall resistance of the $Cd_{0.63-y}Mn_{0.37}Cr_yTe$ ($y = 0.10$) taken at 100, 300, and 370 K. The Hall resistance, R_{Hall}, is given by the sum of the ordinary and the anomalous Hall terms, $R_{Hall} = R_0H + R_SM$ (where R_0 and R_S are the ordinary and anomalous Hall coefficients, H is the magnetic induction, and M is magnetization).[30] While the ordinary Hall effect term contains information on the carrier density in the sample, the presence of the anomalous Hall effect term is responsible for the deviation of the curve in Fig. 5(b) from linear dependence on H, which provides valuable information on magnetic properties of the ferromagnetic material (as discussed in the following paragraph). The sign of the observed Hall coefficient indicates the sample is of the p type, with a carrier

density of $\sim 3.9 \times 10^{19}$ cm^{-3}, as measured at 370 K. The magnetic field dependence of the Hall resistance observed in $Cd_{0.63-y}Mn_{0.37}Cr_yTe$ ($y = 0.10$) shown in Fig. 5(b) (and in other samples that are not shown) becomes increasingly linear with increasing temperature, consistent with the decrease of the anomalous contribution to R_{Hall} as the transition from the ferromagnetic to the paramagnetic state is approached. Thus, although it is well known that the anomalous Hall resistance tends to give a carrier density lower than the actual value, we can assume from the nearly linear behavior of R_{Hall} in Fig. 5(b) that the hole concentrations measured at 370 K provide a reasonable estimate for p. This value is about two orders of magnitude below the Cr concentration, suggesting that the majority of the Cr ions are in the divalent Cr^{++} state and only a small fraction ($\sim$3% or less) of the Cr ions are in the monovalent Cr^{+} state acting as acceptors.

The $R_S M$ term in the expression for the anomalous Hall resistance seen in Fig. 5(b) reflects the magnetization of the free carrier encountered along its path and thus reflects the spin polarization of the ferromagnetic environment as the bulk material as a whole. A zoomed-in graph of the hysteresis for the low-field region is shown in the inset in the figure. It gratifying that the hysteresis observed in R_{Hall} is close to that seen in the hysteresis of magnetization measured by SQUID, confirming that the effect arises from the ferromagnetic bulk properties of the material.

As a final note regarding experimental observations, additional results point to the ferromagnetism of the $Cd_{1-x-y}Mn_xCr_yTe$ quaternary semiconductor alloys. Verma *et al.*[31] recently investigated x-ray absorption spectroscopy (XAS) and x-ray magnetic circular dichroism (XMCD) on the Cr-doped $Cd_{1-x}Mn_xTe$ for Cr concentrations up to 4%. The XAS and XMCD results suggest that (in contrast to $Cd_{1-x}Mn_xTe$) the behavior of Cr-doped $Cd_{1-x}Mn_xTe$ is clearly ferromagnetic and that this observed magnetization involves the majority Mn ions in the material, consistent with the conclusion that the presence of Cr ions is responsible for the formation of the ferromagnetic order in the system.

IV. DISCUSSION AND CONCLUDING REMARKS

In order to meaningfully discuss the ferromagnetic properties of $Cd_{1-x-y}Mn_xCr_yTe$ alloys, we must first resolve whether the observed ferromagnetic effects could arise from unwanted ferromagnetic precipitates in these alloys, e.g., CrTe. The XRD spectrum for $Cd_{0.63-y}Mn_{0.37}Cr_yTe$ obtained for $y = 0.10$ in Fig. 1 (as well as XRD data for samples with lower y that are not shown) shows no traces of phases other than the zinc-blende structure of the alloy. We also note from the inset in Fig. 1 that the lattice parameters for the series of samples for the range $0 < y < 0.10$ show a systematic progression, indicating that Cr is incorporated at the Cd sites of the $Cd_{0.63-y}Mn_{0.37}Cr_yTe$ lattice, forming a homogeneous quaternary system.

In considering the possibility of formation of second-phase inclusions in a $Cd_{1-x-y}Mn_xCr_yTe$ alloy, CrTe is the only phase that is ferromagnetic, with a relatively high Curie temperature of 340 K. However, the value of T_C obtained for our $Cd_{0.63-y}Mn_{0.37}Cr_yTe$ ($y = 0.10$) sample is clearly above 350 K; and the ferromagnetic behavior of $Cd_{0.63-y}Mn_{0.37}Cr_yTe$ samples observed for the other Cr concentrations ($y = 0.03$ and $y = 0.06$) persists to above 350 K (see Fig. 2), indicating that the observed T_C does not arise from the CrTe impurity phase.

Two additional results indicate that the ferromagnetism that we report for the $Cd_{0.63-y}Mn_{0.37}Cr_yTe$ system comes from the bulk of the material rather than unwanted second-phase ferromagnetic inclusions. First, the presence of anomalous Hall effect, which results from the effect of the ferromagnetic environment as the free carrier traverses the lattice, provides an indication that the bulk of the sample is ferromagnetic. Second, if a ferromagnetic phase was present, from the XRD data we must infer that it would be only in trace amounts. Such amounts would clearly be insufficient to result in saturation magnetizations obtained in our SQUID measurements, as indicated by the number of Bohr magnetons per magnetic ion in the inset of Fig. 3(a).

Based on all of the above arguments, we are convinced that the ferromagnetism observed in our $Cd_{1-x-y}Mn_xCr_yTe$ quaternaries is a bulk property of these materials. As proposed theoretically, ferromagnetism in II–VI materials containing transition metal ions (such as $Cd_{1-x-y}Mn_xCr_yTe$) can occur through two mechanisms. The first of these involves mediation of ferromagnetic interactions between the magnetic ions induced by charge carriers (in our case, holes).[12,32] From Hall measurements, we know that our samples are strongly p type, suggesting that the mechanism for ferromagnetism is in principle available. However, the observed carrier concentration in our $Cd_{1-x-y}Mn_xCr_yTe$ samples ($\sim 4.0 \times 10^{19}$ cm^{-3}) is an order of magnitude lower than that reported for materials in which this interaction is known to lead to ferromagnetism (e.g., in GaMnAs, where hole densities of 10^{20} to 10^{21} cm^{-3} are present).[14,16,33] Recognizing, furthermore, that the requirements of this Zener-like model of ferromagnetism on the carrier concentration become increasingly stringent with increasing lattice constant[34] (the lattice parameter of our material is more than 10% larger than that of GaAs), we consider it unlikely that this form of mediation is responsible for the ferromagnetic behavior reported in this paper.

The second proposed mechanism suggests that magnetic polarons (which form when both charge carriers and magnetic ions are present) can lead to ferromagnetic order in the host material.[13–16,19] In our case, such polarons contain both Cr and Mn ions and are bound by the holes contributed by Cr^{+}. In this case, the hole (whose spin interacts with the magnetic moments of both the Mn and the Cr) acts to ferromagnetically align the moments within its orbit, overcoming the antiferromagnetic ordering that is characteristic of Mn ions. As the model suggests, such polarons then interact among themselves in the collective, resulting in a net magnetization of the medium as a whole.

In summary, we have investigated magnetic and transport properties of quaternary $Cd_{1-x-y}Mn_xCr_yTe$ crystals grown by the vertical Bridgman method, extending the study of this material significantly beyond the Cr concentrations investigated earlier.[19,31] We find alloys to be homogeneous, with Cr ions occupying the Cd sites up to at least $y = 0.10$. Samples with $y > 0.03$ are ferromagnetic to temperatures significantly above room temperature and are strongly p type. We ascribe the p-type doping of the system to the presence a

minority population of Cr^{+} ions that act as acceptors, based on our data, we propose that the ferromagnetic behavior arises from the formation of magnetic polarons between the holes and the magnetic ions (both Mn and Cr) comprising the system.

We must emphasize that we arrive at this conclusion primarily by elimination of other possible mechanisms of ferromagnetic order that, based on our data, appear unlikely. However, this argument cannot conclusively eliminate the possibility that there exists some yet-unidentified form of interaction (other than the polaron mechanism) between the Cr ions and the system as a whole that could also result in long-range ferromagnetic order. Independent of the specific model of ferromagnetism, our results have shown that (1) Cr ions can be incorporated into the $Cd_{1-x}Mn_xTe$ matrix up to 10% (significantly beyond the concentrations reported earlier) alloys and (2) the magnetization of the resulting ferromagnetic system clearly increases with the increase in the Cr content. The high-quality and homogeneity of the obtained quaternary suggests that the Cr concentration in such systems can therefore be extended considerably further, thus leading to further enhancement of their ferromagnetic parameters. While this is promising, clearly more research is needed in this area, as well as in definitively establishing the mechanism of ferromagnetic ordering in this quaternary system.

ACKNOWLEDGMENTS

This paper was supported by Nuclear Research and Development Program (No. 2010-0026181) and the Priority Research Centers Program (No. 2009-0093818) through the National Research Foundation of Korea; funded by the Ministry of Education, Science, and Technology; and funded by National Science Foundation Grant No. DMR10-05851.

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