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# Magnetic susceptibility and compositional dependence of the energy gap in $\text{Cd}_{1-x}\text{Co}_x\text{Te}$

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## Abstract

Single crystals of the diluted magnetic semiconductor  $\text{Cd}_{1-x}\text{Co}_x\text{Te}$  grown by the vertical Bridgman technique are characterized by wavelength-modulated reflectivity and magnetization measurements. Low-temperature magnetization and high-temperature magnetic susceptibility analysis were used to extract the exchange integrals for up to third-neighbour  $\text{Co}^{2+}$  pairs in the CdTe host:  $J_1/k_B = -25 \pm 7$  K,  $J_2/k_B = -3.0 \pm 1.5$  K, and  $J_3/k_B = -1.3 \pm 0.3$  K. These values correspond well to the values obtained for other Co-based II–VI diluted magnetic semiconductors and are a clear manifestation of the unusually large  $\text{Co}^{2+}$ – $\text{Co}^{2+}$  antiferromagnetic interaction. The excitonic energy gap  $E_g(x)$  of  $\text{Cd}_{1-x}\text{Co}_x\text{Te}$  in the wavelength-modulated reflectivity shows a linear monotonic increase with increasing  $x$  in the composition range studied.

## 1. Introduction

Diluted magnetic semiconductors (DMSs) are an important subgroup of the ternary compound semiconductors, which due to the substitutional incorporation of magnetically active elements (e.g. Mn, Fe, Co) offer a number of magnetic-field-dependent effects [1]. The most unique feature of DMSs is the strong exchange interaction between localized d electrons and the p and s valence and conduction band carriers, respectively. Despite the progress made during the last two decades in understanding the origin of these exchange interactions [2, 3], these properties have not yet been fully explored. Among the II–VI DMSs, the Co-based ternaries are distinguished by a particularly strong d–d interaction among the  $\text{Co}^{2+}$  ions [4, 5], and a strong p–d interaction between the ions and the conduction band carriers. Typically, the values of the nearest-neighbour (NN)  $\text{Co}^{2+}$  exchange constant  $J_1$  are in the range of 30–50 K. These values

are a factor of 4–5 larger than those for  $\text{Mn}^{2+}$  and  $\text{Fe}^{2+}$ . Some of the experimental methods used to determine the  $J$ -values are based on magnetic susceptibility measurements [4], inelastic neutron scattering [5], and magnetization steps [6].

The experimental data for  $\text{Cd}_{1-x}\text{Co}_x\text{Te}$  published to date are very limited due to the difficulties in preparing homogeneous crystals with sufficient amounts of substitutionally incorporated Co [7–9]. Presumably, the Co concentrations of samples used in the earlier studies were too small to carry out susceptibility measurements at higher temperatures, where the Curie–Weiss procedure [4] could be reliably applied. In this context, it is clearly of interest to study the magnetization of specimens with a higher Co content to determine the  $J$ -values. Here, we report the results and analysis of measurements on the magnetization of  $\text{Cd}_{1-x}\text{Co}_x\text{Te}$  samples ( $x \leq 0.022$ ) in magnetic fields up to 5.5 T and in the temperature range of 1.5–300 K. The free exciton signature of the ternary alloy observed in their wavelength-modulated reflectivity (WMR) spectra is also discussed in terms of the alloy composition.

## 2. Experimental details

The single crystals were grown by a modified Bridgman technique. The starting materials used were Cd and Te, highly purified by multiple vacuum distillation, and Co of 99.998% purity. All the elements were taken in stoichiometric proportions to assure a total ingot weight of about 20 g. Details of the growth procedure are explained elsewhere [10].

Sample compositions were determined using a JEOL JXA-6400 scanning electron microscope with a LINK system for energy-dispersive electron microprobe analysis (EDEMPA). The contents were also checked using theoretical fittings of the magnetization and susceptibility data, as described in the following section.

The magnetization measurements were made in a range of 1.5–300 K and in fields up to 5.5 T using a quantum design superconducting quantum interference device (SQUID) magnetometer. The optical characterization of the samples was performed using WMR. The WMR spectra were recorded with a SPEX-1870 0.5 m monochromator.

## 3. Theoretical background

The free  $\text{Co}^{2+}$  ion has a  $^4\text{F}_{9/2}$  ground state. Under the influence of the crystal field of a  $T_d$  site in CdTe, its sevenfold orbital degeneracy splits into a  $^4\Gamma_2$  singlet, a  $^4\Gamma_5$  triplet, and a  $^4\Gamma_4$  triplet in order of increasing energy. The crystal-field-splitting energy between the ground state  $^4\Gamma_2$  and the first excited state  $^4\Gamma_5$  is  $\Delta = 390.5$  meV [11]. Therefore, only the ground state is populated at low temperatures. Spin–orbit coupling does not split the  $^4\Gamma_2$  ground state, retaining its fourfold spin degeneracy [12]. Thus, at low temperatures, the  $\text{Co}^{2+}$  ion behaves paramagnetically with an effective spin  $S = 3/2$ . The excited state orbital triplets are responsible for the value of the gyromagnetic factor,  $g_{\text{Co}^{2+}} = 2.309$  [13], which is significantly different from the  $g = 2$  value for a spin-only state.

The magnetization (per unit mass) of  $\text{Cd}_{1-x}\text{Co}_x\text{Te}$  as a function of the field  $H$  can be expressed as

$$M_m(H) = M_m^*(H) + \chi_{dia}H = -\frac{g_{\text{Co}^{2+}}\mu_B N_A}{W(x)}x\langle\langle S_z \rangle\rangle + \chi_{dia}H, \quad (1)$$

where  $N_A$  is Avogadro's number,  $\mu_B$  is the Bohr magneton, and  $W(x)$  is the molar weight of  $\text{Cd}_{1-x}\text{Co}_x\text{Te}$ . Here  $M_m^*$  is the contribution of the  $\text{Co}^{2+}$  ions to the magnetization, while  $\chi_{dia}$  arises from the diamagnetic susceptibility of the DMS.

In equation (1), the mean spin  $\langle\langle S_z \rangle\rangle$ , denotes the spatial as well as the thermal average of the spin component along the magnetic field  $H$ . Assuming the magnetization of a dilute alloy, in which it is reasonably justified to consider contributions of only two types of complex: *singles* (with no other neighbouring magnetic ion), and *pairs* (when two ions are neighbours), and for spin interactions up to the  $k$ th neighbour,

$$\langle\langle S_z \rangle\rangle_k = P_k^s(x)\langle S_z \rangle_k^s + \sum_{j=1}^k P_j^p(x)\langle S_z \rangle_j^p, \quad (2)$$

where the index  $s$  refers to isolated ions up to the  $k$ th neighbour, and the index  $p$  corresponds to pairs that are first ( $j = 1$ ), ...,  $k$ th ( $j = k$ ) neighbours.  $P_j(x)$  is the probability that a magnetic ion spin belongs to the complex  $j$ . For a random distribution of ions over an fcc sublattice, Kreitman and Barnett [14] and Okada [15] give the probabilities for first-, second-, and third-neighbour interactions.

The contribution of  $k$ th-order single ions to the magnetization can be described by a modified Brillouin function for a spin  $S = 3/2$ :

$$\langle S_z \rangle_k^s = -\frac{3}{2}\mathcal{B}_{3/2}(\eta), \quad (3)$$

where  $\eta = [Sg_{\text{Co}^{2+}}\mu_B H]/[k_B(T + T_0)]$ , and  $\mathcal{B}_{3/2}(\eta)$  is the Brillouin function [16]. Here,  $T_0$  is a phenomenological parameter that accounts for the long-range antiferromagnetic coupling of distant-neighbour magnetic ions (more distant than  $k$ th neighbour) [17].

Besides the modified Brillouin function contribution of single ions to the magnetization, which saturates at high fields, there can also be a significant high-field susceptibility contribution due to strong interactions of second- and more-distant-neighbour pairs of magnetic ions [18, 19]. For a  $k$ th-neighbour pair of Co<sup>2+</sup> ions with exchange integral  $J_k$ , the eigenvalues of their Hamiltonian are given by [6, 18]

$$E_k^p(S_T, m_S) = -J_k[S_T(S_T + 1) - S(S + 1)] + g_{\text{Co}^{2+}}\mu_B m_S H, \quad (4)$$

where  $S_T$  is the total spin of the ion pair ( $0 \leq S_T \leq 3$ ), and  $m_S$  is the component of the total spin along the field  $H$  ( $|m_S| \leq S_T$ ). The thermal average of the spin per ion can then be calculated from

$$\langle S_z \rangle_k^p = \sum_{S_T} \sum_{m_S} m_S \exp\left(-\frac{E_k^p(S_T, m_S)}{k_B T}\right) / \sum_{S_T} \sum_{m_S} \exp\left(-\frac{E_k^p(S_T, m_S)}{k_B T}\right). \quad (5)$$

One of the possible methods used to determine the  $J_1$ -value in a DMS is to extract it from high-temperature susceptibility data, as reported for Cd<sub>1-x</sub>Co<sub>x</sub>Se [4]. For  $T > J_1/k_B$ , the exchange interaction between the magnetic ions in a DMS can be incorporated into its susceptibility  $\chi$  by using the mean-field approximation or the high-temperature expansion for randomly diluted Heisenberg antiferromagnets [20]. Then, the magnetic susceptibility can be expressed in the form of the Curie–Weiss law, which in turn can be used to extract the effective exchange integral  $J_{eff}$  between the magnetic ions. The molar susceptibility is given by [21]

$$\chi(T) = \chi^*(T) + \chi_G + \chi_{dia} = \frac{C(x)}{T - \Theta(x)} + \chi_G + \chi_{dia}, \quad (6)$$

with the Curie constant per mole

$$C(x) = \frac{N_A \mu_B^2}{3k_B} x g_{\text{Co}^{2+}}^2 S(S + 1), \quad (7)$$

and the Curie–Weiss temperature

$$\Theta(x) = \frac{2}{3k_B} z x S(S + 1) J_{eff}. \quad (8)$$

Here  $z$  is the number of cations in the first coordination sphere (12 for the zinc-blende structure), and  $\chi_G$  is the temperature-independent molar susceptibility:

$$\chi_G = 8xN_A\mu_B^2/\Delta. \quad (9)$$

This term results from the admixture of higher orbital states with the ground state of  $\text{Co}^{2+}$  [21].

It should be noted that  $J_{eff}$  is not equal to the NN exchange integral  $J_1$ , but is rather its upper limit. In addition to the dominant interaction between first neighbours, it also includes interactions between higher-order neighbours. If the magnetic ion concentration is small ( $x \sim 0.01$ ), for strong exchange interactions up to the  $k$ th neighbour, it is possible to express  $J_{eff}$  in terms of the  $J_1, J_2, \dots, J_k$  [4]:

$$J_{eff}N_1 = \sum_{i=1}^k N_i J_i, \quad (10)$$

where  $N_i$  is the number of cation sites in the  $i$ th coordination sphere.

## 4. Results and discussion

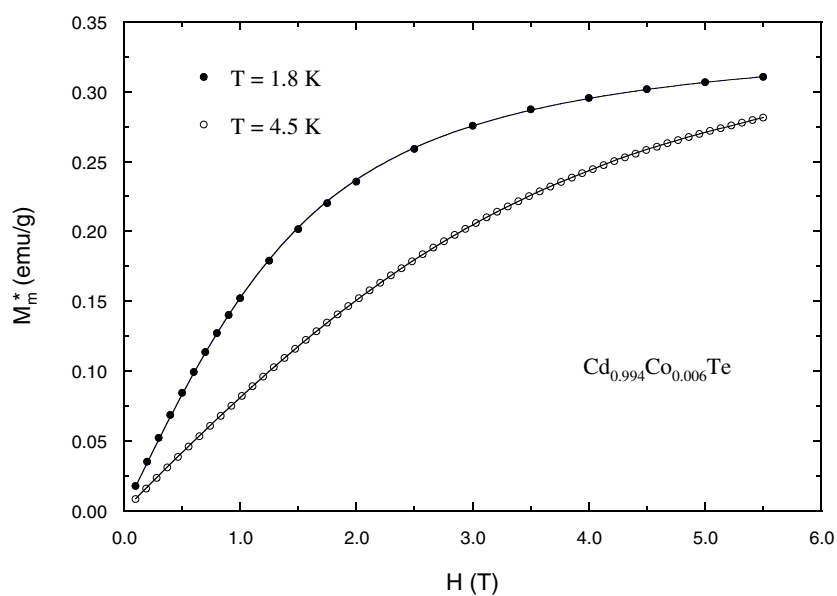
### 4.1. Low-temperature magnetization

The magnetization  $M_m(H)$  for a  $\text{Cd}_{1-x}\text{Co}_x\text{Te}$  specimen with  $x = 0.006$  was measured at low temperatures. After subtracting the diamagnetic contribution,  $M_m^*(H)$  values for  $T = 1.8$  and  $4.5$  K were plotted in figure 1. It is clear from this figure that the data points cannot be fitted simply with a modified Brillouin function which saturates at high fields. This is an indication of the strong interaction between the  $\text{Co}^{2+}$  ions which extends beyond the first neighbours. Indeed, to fit these points, strong interactions at least up to the third neighbour had to be considered. Therefore, the fitting to the data points involved three exchange constants:  $J_1, J_2$ , and  $J_3$ , with the weaker further-neighbour interactions included in the phenomenological parameter  $T_0$ .

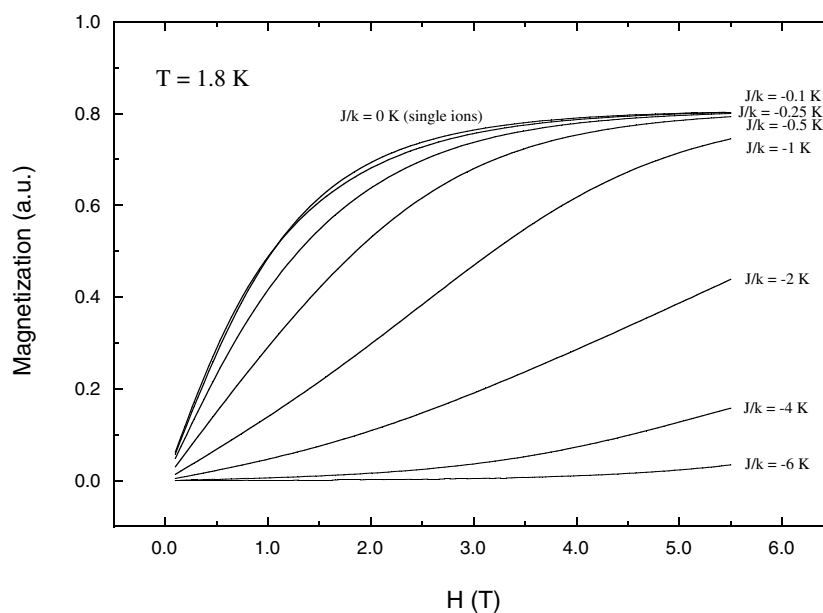
Based on equations (4) and (5), figure 2 shows the theoretical calculation of the magnetization per  $\text{Co}^{2+}$  ion in a pair as a function of magnetic field (up to 5.5 T) for various exchange constants at the fixed temperature of 1.8 K. It is important to notice that in the available magnetic field range,  $\text{Co}^{2+}$  pairs with an exchange integral  $|J|/k_B > 6$  K contribute very little to the magnetization. These pairs are frozen together, and their exchange interaction is too strong to be broken by the external magnetic field.

Since  $|J_1|/k_B = 6.1$  K for  $\text{Mn}^{2+}$  in CdTe, it is reasonable to expect  $|J_1|/k_B > 6$  K for  $\text{Co}^{2+}$ . Therefore, the contribution of  $\text{Co}^{2+}$  NN pairs to the magnetization of the specimen should be negligible for the range of  $H/T$  used in this investigation. Assuming that only second- and further-neighbour pairs contribute to the magnetization (i.e.,  $\langle S_z \rangle_1^p = 0$ ), the data points were fitted with four free parameters:  $x, T_0, J_2$ , and  $J_3$ . The fit of the theoretical model for a  $\text{Cd}_{1-x}\text{Co}_x\text{Te}$  sample with  $x = 0.006$  at  $T = 1.8$  and  $4.5$  K is shown in figure 1 and the results are summarized in table 1. The best estimates for the exchange integrals are:  $J_2/k_B = -3.0 \pm 1.5$  K and  $J_3/k_B = -1.3 \pm 0.3$  K.

Low-temperature magnetization measurements were also made for samples with larger concentration of magnetic ions ( $x \simeq 0.02$ ). However, it was not possible to extract reasonable values of  $J_2$  and  $J_3$  from those data. The uncertainty in fitting was equal to or larger than the value of the exchange integral, presumably an indication of the theoretical model limitations. The probability that a  $\text{Co}^{2+}$  ion is a third-neighbour single ion, or belongs to a pair of up to third neighbours, is equal to 96% for  $x = 0.006$ , while it is only 66% for  $x = 0.02$ . This means that for  $x = 0.02$  almost a third of the magnetic ions are not accounted for by the cluster

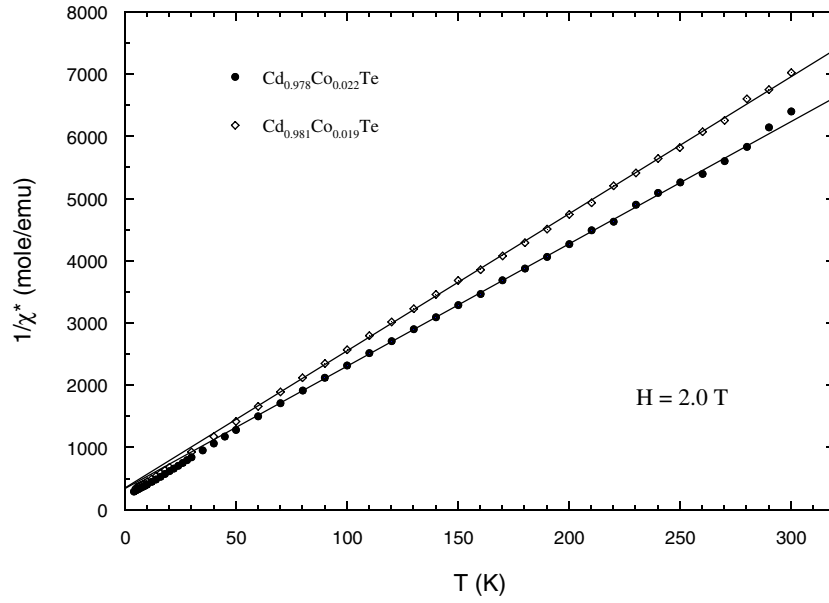


**Figure 1.** Magnetization of a  $\text{Cd}_{0.994}\text{Co}_{0.006}\text{Te}$  sample (corrected for the host diamagnetism) as a function of the magnetic field ( $H$ ) for  $T = 1.8$  and 4.5 K.



**Figure 2.** Theoretical calculation of magnetization per  $\text{Co}^{2+}$  ion in a pair as a function of magnetic field for different values of the exchange constant. The temperature is set to 1.8 K.

model. Those ions still contribute to the parameter  $T_0$ , which does improve the overall fit, but it does not make the values of  $J_2$  and  $J_3$  more accurate.



**Figure 3.** Inverse molar magnetic susceptibility as a function of temperature for  $\text{Cd}_{1-x}\text{Co}_x\text{Te}$  with  $x = 0.022$  and  $0.019$ . The straight lines are least-squares linear fits to the data points for the range of  $T > 90$  K.

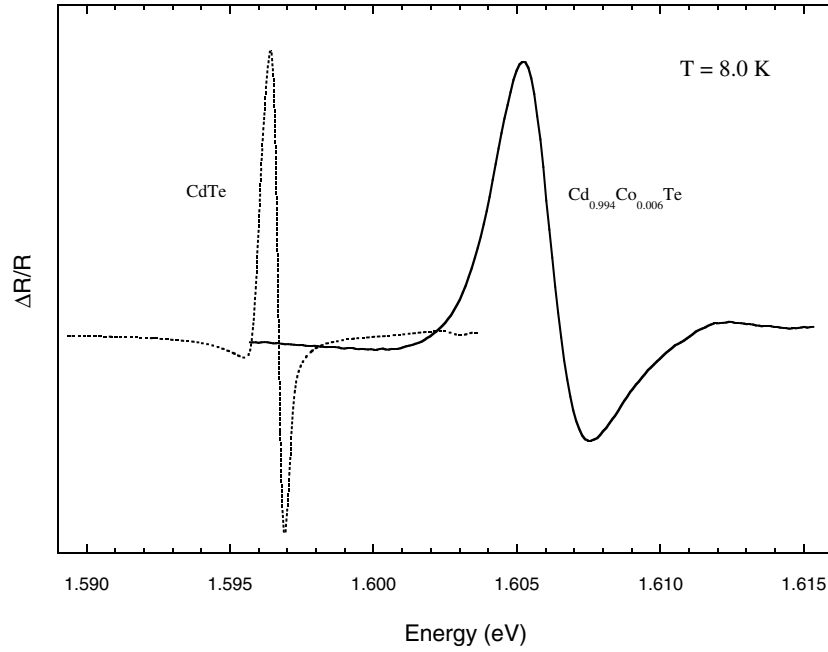
**Table 1.** Least-squares fitting parameters for the low-temperature magnetization data.

$x$ (EMPA)	$T$ (K)	$x$ (magnetization fit)	$T_0$ (K)
$0.006 \pm 0.001$	1.8	$0.0043 \pm 0.0001$	0.42
$0.006 \pm 0.001$	4.5	$0.0045 \pm 0.0001$	0.48
$0.011 \pm 0.001$	1.8	$0.010 \pm 0.007$	2.6

#### 4.2. High-temperature susceptibility

The magnetic susceptibility  $\chi(T)$  for two  $\text{Cd}_{1-x}\text{Co}_x\text{Te}$  samples with  $x = 0.022$  and  $0.019$  were measured for the range  $T = 2\text{--}300$  K at  $H = 2.0$  T. After the temperature-independent parts ( $\chi_G$  and  $\chi_{dia}$ ) were subtracted, equations (7) and (8) were used to extract  $x$  and  $J_{eff}$  from the data. A plot of  $1/\chi^*$  as a function of temperature for the two samples is shown in figure 3. The straight lines represent the least-squares fit of the Curie–Weiss law to the experimental data in the high-temperature range. Below  $\sim 90$  K, the magnetic susceptibility does not follow the Curie–Weiss law. This deviation is similar to that observed in  $\text{Cd}_{1-x}\text{Mn}_x\text{Te}$ , but it occurs at a much higher temperature. This is another indication of the strong exchange interaction between  $\text{Co}^{2+}$  ions compared to that for  $\text{Mn}^{2+}$  ions in the CdTe host. A similar behaviour is observed for other Co-based II–VI DMSs [4].

The results of the least-squares fit to the data are summarized in table 2. The value of  $J_{eff}/k_B = -29 \pm 6$  K obtained from the fits is negative for both samples, indicating an antiferromagnetic interaction between the  $\text{Co}^{2+}$  ions. Using equation (10) and the values obtained above for  $J_2$  and  $J_3$ , it is possible to deduce  $J_1/k_B = -25 \pm 7$  K for the  $\text{Co}^{2+}$  NN exchange integral in CdTe.



**Figure 4.** The WMR spectrum of a  $\text{Cd}_{0.994}\text{Co}_{0.006}\text{Te}$  sample, along with a reference spectrum for CdTe.

**Table 2.** Least-squares fitting parameters for the high-temperature susceptibility data ( $T > 90$  K).

$x$ (EMPA)	$x$ (susceptibility fit)	$J_{eff}/k_B$ (K)
$0.019 \pm 0.001$	$0.018 \pm 0.002$	$-29 \pm 6$
$0.022 \pm 0.001$	$0.020 \pm 0.002$	$-29 \pm 6$

#### 4.3. Compositional dependence of the energy gap

The excitonic energy gaps ( $E_g$ ) of the  $\text{Cd}_{1-x}\text{Co}_x\text{Te}$  specimens at 8 K were determined using WMR. An example of the WMR spectrum for a sample with  $x = 0.006$  is shown in figure 4, along with that for CdTe as a reference. The blue-shift of  $E_g$  by  $\sim 9$  meV is clearly evident in the figure. The broadening of the excitonic feature for the  $\text{Cd}_{1-x}\text{Co}_x\text{Te}$  sample, compared to that of CdTe, can also be seen. This broadening results from the structural disorder introduced in the CdTe lattice by the incorporation of  $\text{Co}^{2+}$  ions. The monotonic increase of this broadening observed for samples with increasing  $x$  is in good agreement with this conclusion.

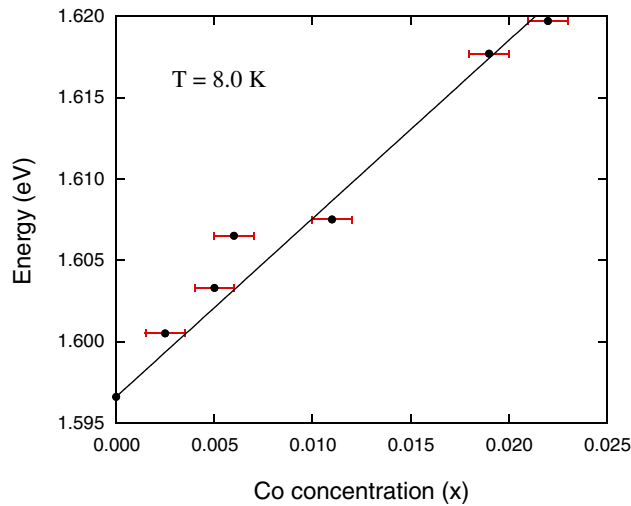
Dependence of the  $E_g$  on the composition  $x$  is shown in figure 5. In the small range of compositions available,  $0 \leq x \leq 0.022$ , the dependence  $E_g(x)$  seems to be linear. A least-squares fit of the data yields

$$E_g(x) = 1.597 + 1.095x \text{ (eV)}. \quad (11)$$

Despite the small range of Co composition considered in this study, it is possible to make a crude estimate of  $E_g$  for CoTe (i.e.,  $x = 1$ ). Assuming that there is no bowing in  $E_g(x)$ , as is also reported for  $\text{Cd}_{1-x}\text{Mn}_x\text{Te}$  [22] and  $\text{Cd}_{1-x}\text{Fe}_x\text{Te}$  [23], extrapolation of the data yields a value of 2.69 eV for the zinc-blende CoTe. This value is much smaller than the values 3.19 and 3.37 eV for MnTe and FeTe, respectively<sup>5</sup>.

<sup>5</sup> Extracted by extrapolation from the data in [22, 23]





**Figure 5.** Dependence of the energy gap of  $\text{Cd}_{1-x}\text{Co}_x\text{Te}$  on the composition of the samples used in this study. The straight line is the least-squares linear fit to the data points.

(This figure is in colour only in the electronic version)

**Table 3.** First- and second-neighbour exchange constants for some II–VI DMSs containing Mn, Fe, and Co.

Material	$-J_1/k_B$ (K)	$-J_2/k_B$ (K)	Reference
$\text{Cd}_{1-x}\text{Mn}_x\text{Te}$	$6.1 \pm 0.2$	$1.1 \pm 0.2$	[29]
$\text{Cd}_{1-x}\text{Fe}_x\text{Te}$	$8.8 \pm 1.2$	1.5	[30]
$\text{Cd}_{1-x}\text{Co}_x\text{Te}$	$25 \pm 7$	$3.0 \pm 1.5$	Present study
$\text{Cd}_{1-x}\text{Co}_x\text{Se}$	$31 \pm 2$		[31]
$\text{Cd}_{1-x}\text{Co}_x\text{S}$	$30.4 \pm 1.1$		[32]
$\text{Zn}_{1-x}\text{Co}_x\text{Te}$	$38 \pm 2$	$5.7 \pm 0.6$	[5]

## 5. Concluding remarks

From the magnetization results obtained in the present investigation, it is found that the antiferromagnetic d–d interaction between  $\text{Co}^{2+}$  ions is much stronger than that between  $\text{Mn}^{2+}$  ions in CdTe. This conclusion is in agreement with the large values of d–d exchange constants  $|J_1|$  and  $|J_2|$  for Co-based compared to Mn- and Fe-based II–VI DMSs [3, 24]. The large d–d interaction in  $\text{Cd}_{1-x}\text{Co}_x\text{Te}$  is also consistent with the large p–d interaction reflected in the value of  $\beta$  [25, 26]. According to the superexchange model of Larson *et al* [27], developed for Mn-based DMSs, one can state roughly that a larger magnitude of  $\beta$  leads to a larger d–d interaction between the magnetic moments of the transition metal ions. The large increase of  $|J_1|$  and  $|J_2|$  from Mn- to Fe- to Co-based II–VI DMSs is indeed in accordance with the corresponding increase of  $|\beta|$  [19, 24, 26].

Table 3 shows a list of  $J$ -values for several II–VI DMSs containing Mn, Fe, and Co. The  $J$ -values obtained in the present study for  $\text{Cd}_{1-x}\text{Co}_x\text{Te}$  fit well into the general trends among these DMSs. For example, the  $J$ -values for a given magnetic ion increase in going from heavier to lighter ions in the host II–VI binary: from Cd to Zn cations, and from Te to Se to S anions. This trend is mainly due to the spatial extension of the exchange interaction, which decreases exponentially with increasing NN distance [28]. For a given II–VI binary host, there

is also a general trend that the  $J_1$ -values increase in going from Mn to Fe to Co. This result is consistent with the theoretical calculation of the exchange integral  $J$  for the superexchange, which is believed to be the dominant exchange mechanism in this case. The calculated  $J$ -value is inversely proportional to the square of the ionic spin ( $J \propto 1/S^2$ ), as reported by Blinowski and Kacman [33].

The value of  $E_g \sim 2.69$  eV deduced in the present study for the zinc-blende CoTe is significantly lower than the value 3.90 eV determined by extrapolating the data for Zn<sub>1-x</sub>Co<sub>x</sub>Te [19]. Part of this discrepancy can be attributed to the relatively small range of Co content in both studies ( $x \leq 0.022$  for Cd<sub>1-x</sub>Co<sub>x</sub>Te and  $x < 0.012$  for Zn<sub>1-x</sub>Co<sub>x</sub>Te), and hence a large uncertainty in the extrapolated values at  $x = 1$ . But another possibility is a deviation of  $E_g$  from a linear dependence on  $x$  and the existence of a bowing in  $E_g(x)$ . This claim can only be checked by growing Cd<sub>1-x</sub>Co<sub>x</sub>Te and Zn<sub>1-x</sub>Co<sub>x</sub>Te specimens with higher Co content, possibly by non-equilibrium techniques like molecular beam epitaxy.

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