Structural, magnetic, and critical behavior of CrTe$_{1-x}$Sb$_x$ alloys

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Abstract We investigate the structural and critical properties of CrTe$_{1-x}$Sb$_x$ with 0.0 $\leq$ x $\leq$ 0.2. The XRD patterns revealed that Sb substitution resulted in a pure NiAs-hexagonal structure with P6$_3$/mmc (194) space group. Lattice refinement of the structure revealed little changes in the $a$-lattice parameter, along with a more pronounced reduction in the $c$-axis. The critical behavior in CrTe$_{1-x}$Sb$_x$ has been investigated using the magnetization isotherms near the ferromagnetic transition. The obtained critical exponents ($\beta$, $\gamma$, and $\delta$) revealed that all samples (with 0.0 $\leq$ x $\leq$ 0.2) closely follow a mean field-like behavior with ferromagnetic Curie temperature near room temperature. The results from Widom scaling relation indicate self-consistency of the acquired values. Moreover, the magnetization isotherms near the Curie temperature follow a universal scaling behavior, giving further support for the obtained critical exponents.

1 Introduction

Cr-based pnictogens and chalcogens continue to attract the attention of scientists due to their versatile physical properties [1–5]. These properties range from superconducting to semiconducting and half-metallic [6, 7], ferromagnetic van der Waals to antiferromagnetic (AFM) behavior [8–14]. Some of the emerging possible applications are in magnetic cooling and spintronic devices [14]. The binary compounds of Cr-pnictogens (N, As, Sb) and the Cr-chalcogens (S, Se, Te) crystallize in the hexagonal NiAs crystal structure [1–5]; however, the chemical bonding may lead to diverse magnetic and physical properties. Ionic bonding dominates the chalcogenides, while the bonding in pnictide is nonionic. For instance, Cr$_7$Te is a ferromagnetic (FM) material with a Curie temperature covering wide range (320–360 K) depending on the value of x [6–15]. It is well established that Cr$_7$Te is nonstoichiometric and does not occur in pure hexagonal phase [16, 17]. While substitution at the Cr sites helps
to stabilize the hexagonal NiAs structure, deficiency and defects may also stabilize other structures like trigonal Cr$_2$Te$_3$ and monoclinic Cr$_3$Te$_4$ structure. Chromium antimony CrSb is antiferromagnetic with $T_N \sim 700$ K, and it crystalizes in a NiAs-type structure [18–22] with $\mu_{\text{eff}} = 3.89 \mu_B$ [23, 24]. In CrSb, the Cr spins are aligned ferromagnetically within the basal plane; while in the adjacent planes the spins are aligned antiferromagnetically [22]. Substitution in the solid solutions of CrTe$_{1-x}$Sbx affects the spins configurations and the magnetic phase in this compound [21, 25]. Increasing the Sb concentrations in CrTe$_{1-x}$Sbx leads to a gradual reduction in FM ordering temperature and the saturated magnetization along with moderate reduction in unit cell volume. At high concentration of Sb, a more complex magnetic phases may occur such as canted spin structures and noncollinear spin configurations and even spin-glass like behavior have been observed [14, 26, 27]. Some of these magnetic changes qualitatively agree with de Gennes model of the double exchange interaction in AFM–FM system [28].

Preliminary critical behavior analysis in CrTe$_{1-x}$Sbx has been published in an earlier work [29] where we investigate the effect of Sb substitution over relatively wide range. The modified Arrott plots and Kouvel–Fisher critical exponents’ analyses revealed that upon increasing the Sb, the critical exponents deviate significantly from the mean field values and gradually shift toward 3D Heisenberg models. More recently, we found that repeated grinding and annealing during sample preparation improve the phase purity in CrTe$_{1-x}$Sbx, especially at low Sb concentration [14]. For these samples, we investigated the magnetocaloric effect near room-temperature CrTe$_{1-x}$Sbx and found that the relative cooling power is comparable to pure Gd—the prototype magnetocaloric material.

In this work, we extend our investigation of the magnetic and the structural properties of CrTe$_{1-x}$Sbx alloys at low Sb concentrations. The samples used in this study are the same samples we used in Ref. 14. We also investigate the detailed properties of the critical behavior near the FM-PM transition in these newly prepared samples and compare them with our earlier works [29]. We also include evidences on the changes in the bond length and angle with Sb substitution that ultimately may affect the magnetic and critical behavior. This may lead to a better understanding of the observed differences with earlier work on the critical behavior [29] in this material.

2 Experimental techniques

Polycrystalline CrTe$_{1-x}$Sbx ($x \leq 0.2$) was prepared using high-temperature solid-state reaction [30, 31]. High-purity (4–5 N) powders of Cr, Te, and Sb are used. The detail of the preparation method is reported in our previous work [29]; however, for the samples in this study we used several repetition of intermediate grinding and annealing to improve the phase purity and quality of the samples. Bruker X-ray diffractometer D2-Phaser with Cu Kα ($\lambda = 1.54056$ Å) has been used to obtain the powdered XRD patterns over 20°–80° [14, 32]. We used Rietveld refinements available in FULLPROF software to analyze the XRD patterns. Quantum design 3-T VersaLab has been used to collect the magnetic data in the temperature range of 50–400 K.

3 Analysis and results

Figure 1 shows the room-temperature XRD patterns of the studied CrTe$_{1-x}$Sbx samples. Rietveld refinement of the data was carried out to determine the lattice parameters, phase
Fig. 1 XRD patterns for CrTe$_{1-x}$Sb$_x$. The stars indicate the position of the impurity phase.

purity, and structure [33, 34]. The results and the lattice parameters for all investigated samples are discussed in detail elsewhere [14]. Those observations are consistent with the noticed shifts of the positions of the diffractions peaks with the increasing the Sb substitution ratio in the samples (Fig. 2); the color map at Fig. 2 shows a noticeable shift in the $(00l)$ peaks to higher angles indicating a shortening in the c-lattice parameter in direct space.

A relatively smaller shift to the left in the $(h\,k\,0)$ peaks indicates a relaxation along the basal plane $a$-lattice. Moreover, the figure (for $x = 0.0$) also reveals the presence of secondary-phase Cr$_2$O$_3$, which has a trigonal structure with R-3c space group, leading to the nonstoichiometric Cr$_x$Te sample in line with earlier results [8]. Street et al. concluded that chromium telluride Cr$_x$Te exists over a range of Te concentration with the hexagonal structure [8, 35]. The Wyckoff positions for the hexagonal structure are given in Table 1. Sb substitution causes relaxation of the atomic bond along $(a-b)$ axes; this probably will reflect residual stress along the $c$-axis (Fig. 3) leading to the observed variations in the lattice parameters and the overall increase in the volume of the unit cell for $x = 0–0.10$ [see 14].

The magnetization dependence on temperature in the range of 50–400 K at $H = 0.03$ T is published elsewhere [14]. The thermomagnetic curves for the selected samples ($x = 0.00$, 0.05, 0.10, 0.15, 0.20) are shown in Fig. 4.
Table 1 Wyckoff position for CrTe$_{1-x}$Sb$_x$ hexagonal structure space group P6$_3$/mmc (194)

| Atom  | Wyckoff | Site  | $x/a$ | $y/b$ | $z/c$ |
|-------|---------|-------|-------|-------|-------|
| Cr    | 2a      | −3 m  | 0     | 0     | 0     |
| Te/Sb | 2c      | −6m2  | 1/3   | 2/3   | 1/4   |

Fig. 3 Unit cell structure of the CrTe$_{1-x}$Sb$_x$ hexagonal phase. The red balls (big) represent Te atoms, while the blue ones (small) represent Cr atoms. The arrows at a indicate the changes in the unit cell dimensions by increasing the Sb substitution ratio; b shows the CrTe octahedron

0.10, and 0.20) reveal a clear FM–PM phase transition at an inflection points ~ 325.0, 296.0, and 287.0 K, respectively [14]. The results also reveal a slight increase in the magnetization along with sharper transition increasing the concentration of antimony. This probably indicates an increase in the effective magnetic moment per Cr-ion resulting from the exchange interaction with increasing Sb concentration. Figure 4 presents the variations of the inverse magnetic susceptibility ($\chi^{-1}$) with temperature. In the paramagnetic region (i.e., above Tc), the inverse magnetic susceptibility obeys Curie–Weiss law $\chi = C (T - \theta_p)^{-1}$, where $C$ and $\theta_p$ are the Curie constant and the Curie temperature, respectively, following a linear behavior down to $\theta_p$ in all samples. The linear extrapolation gives the following ($\theta_p$) values 344, 309, and 302 K for $x = 0.0, 0.1$ and 0.2, respectively [14]. The values of $\theta_p$ are higher than the Tc values obtained from the inflection points of the $M$ versus $T$ curves. This probably suggests more complex spin configuration than normal FM spin configuration. Recently, Kong et al. observed similar results in CrSbSe$_3$ [36].

The linear portion of the $\chi^{-1}$ versus $T$ curves can be used to evaluate the effective magnetic moment $\mu_{\text{eff}}$ using $\mu_{\text{eff}} = \sqrt{3k_B C/N} = \sqrt{8C} \mu_B$, where $C$ is the inverse of the slope and $\mu_B$ is the Bohr magneton. The obtained values are 3.92 $\mu_B$, 4.40 and 4.56 for $x = 0.0, 0.1$ and 0.2, respectively. The expected value for Cr$^{3+}$ is 3.87 $\mu_B$ [23, 24]. These values indicate a gradual increase in the Cr effective moment with increasing Sb concentration. These values are in line with the noticed increase in the observed magnetization seen in [14]. The obtained values of $\mu_{\text{eff}}$ along with $\theta_p$ are listed in Table 2.

The magnetic isotherms $M(H, T)$ of CrTe sample are shown in Fig. 5 in the temperature range of 300–376 K. The magnetic behavior in Fig. 5 is deeply discussed elsewhere [14]. The magnetization data in Fig. 5 are presented in modified Arrott plots (MAP) in Fig. 6 in order
Fig. 4 Temperature-dependent inverse magnetization for different CrTe$_{1-x}$Sb$_x$ samples measured at $H = 0.03$ T

Table 2 Effective magnetic moments and $\theta_p$ values for the CrTe$_{1-x}$Sb$_x$

| $x$ | $\theta_p$ (K) | $\mu_{eff}$ ($\mu_B$) |
|-----|----------------|----------------------|
| 0.00 | 342.9          | 3.9(2)               |
| 0.10 | 308.3          | 4.4(0)               |
| 0.20 | 301.0          | 4.5(6)               |

Fig. 5 Variations of the magnetization with field with $\Delta T = 2$ K intervals for CrTe sample

to investigate the critical behavior and other magnetic properties in the following sections [13, 37, 38].

4 Critical behaviors

The modified Arrott plots (MAP) of the magnetization along with Kouvel–Fisher (K–F) plots greatly refine the critical exponents near the phase transition [39, 40]. The K–F analysis is based on obtaining a linear behavior of the inverse the magnetic susceptibility ($\chi_0^{-1}$) and the spontaneous magnetization (Ms). Moreover, Ms and $\chi_0^{-1}$ linear graphs follow a universal
Fig. 6 Modified Arrott plots for CrTe: a mean field, b 3D-Ising, c tricritical mean field and d 3D-Heisenberg model

behavior and their slopes determine the critical exponents [41]. $M_s$ and $\chi_0^{-1}$ can be obtained from:

$$M_s(T) = M_0(-\epsilon)^\beta, \text{ below } T_c$$  \hspace{1cm} (1)

$$\chi_0^{-1} = (h_0/m_0)\epsilon^\gamma, \text{ above } T_c$$  \hspace{1cm} (2)

$$M = DH^{1/\delta}, \text{ at } T_c$$  \hspace{1cm} (3)

where $\beta$, $\gamma$, and $\delta$ are the critical exponents; $\epsilon$ is the reduced temperature; $D$, $M_0$ and $h_0/m_0$ are the critical amplitudes and $M$ is the critical isotherm $M(H)$ at $T_c$ [40]. The critical exponents $\beta$, $\gamma$ take different values in different magnetic models.

According to the MAP analysis, the magnetic isotherms at high fields are expected to be parallel lines for the correct set of critical exponents. The analysis is based on Arrott–Noakes equation of state, namely:

$$\left(\frac{H}{M}\right)^{1/\gamma} = \left(\frac{T - T_c}{A + (M/B)^{1/\beta}}\right)$$  \hspace{1cm} (4)

where $A$ and $B$ are constants [42].

The commonly used magnetic models are the tricritical mean field ($\beta = 0.25; \gamma = 1$), 3D-Heisenberg ($\beta = 0.365; \gamma = 1.336$) and the 3D-Ising ($\beta = 0.325; \gamma = 1.24$) [42].

In Fig. 6, we present the variation of $M^{1/\beta}$ with $(H/M)^{1/\gamma}$ for CrTe sample using various magnetic models. The initial slopes of the curves in Fig. 6 are positive above and below $T_c$; according to Banerjee, the positive slopes indicate a second-order phase transition [43]. The slopes at high fields in Fig. 6 can be used to obtain the relative slope (RS) analysis.
that allows us to determine the most suitable magnetic model that closely represents the magnetic isotherms. The RS values obtained from Fig. 6 are represented in Fig. 7. The RS curve obtained from mean field model is the closest to the horizontal line at RS = 1 above and below Tc indicating that the mean field model closely represents CrTe magnetic state [44]. Similar analysis has been applied for CrTe$_{1-x}$Sb$_x$ samples. We found that the mean field model closely represents the magnetic isotherms in the modified Arrott plot representation. The RS curves for CrTe$_{1-x}$Sb$_x$ samples are presented in Fig. 7, and the modified Arrott plots using the mean field theory (MFT) (for $x = 0.10$ and 0.20) are shown in Fig. 8. It is worth mentioning that, for $x = 0.20$, there are noticeable differences between the previously published data and results obtained in this work (see [29] for comparison). Such differences may be caused by the better quality of the samples used in the current study and the observed relaxation of the atomic bond in the (a-b)-plane especially at high Sb concentration.

The mean field representation of the modified Arrott plots (Figs. 6, 8) has been used to extract the spontaneous magnetization $M_s(T)$ and the inverse of the magnetic susceptibility $\chi^{-1}_0(T)$. At each temperature, the curves at high fields are linearly fitted to Eq. 4 and extended to the $M^{1/\beta}$ and $(H/M)^{1/\gamma}$ axes. The intercept points represent the $M_s(T)$ and $\chi^{-1}_0$. The variations of $M_s(T)$ and $\chi^{-1}_0(T)$ with temperature for CrTe$_{1-x}$Sb$_x$ samples with $x = 0.0, 0.1$ and 0.2 are presented in Figs. 9a, 10a and 11a, respectively, and then fitted to Eqs. 1 and 2 in order to evaluate the critical exponents $\beta$ and $\gamma$ along with the transition temperature $T_c$. The obtained values for CrTe sample are: $\beta = 0.49 \pm 0.10$, and $\gamma = 1.04 \pm 0.02$ and
Fig. 8 Modified Arrott plots using MFT for CrTe$_{1-x}$Sb$_x$ ($x = 0.1$ and $0.2$)

Fig. 9 a Variations of $M_s$ and $\chi_0^{-1}$ with temperature, b Kouvel–Fisher plots for $M_s(T)$ and $\chi_0^{-1}(T)$ plot for CrTe

$T_c = 334.0 \pm 0.1$. Similarly, for CrTe$_{1-x}$Sb$_x$ with $x = 0.1$ and $0.2$, the values are: ($\beta = 0.50 \pm 0.01$, $\gamma = 0.99 \pm 0.01$ and $T_c = 303.3 \pm 0.1$) and ($\beta = 0.51 \pm 0.01$, $\gamma = 1.00 \pm 0.01$ and $T_c = 295.4 \pm 0.1$), respectively.

The Kouvel–Fisher (K–F) plots representations of the spontaneous magnetization and the susceptibility greatly improve the accuracy of modified Arrott analysis for the critical exponents. According to the (K–F) model [40],

\[
\frac{dM_s(T)}{dT} = \frac{T - T_c}{\beta} \\
\frac{d\chi_0^{-1}(T)}{dT} = \frac{T - T_c}{\gamma}
\]

Both equations are linear in temperature ($T$-$T_c$) with slopes equal to $1/\beta$ and $1/\gamma$. Figures 9b, 10b and 11b illustrate the K–F plot for CrTe$_{1-x}$Sb$_x$ samples with $x = 0.0$, 0.1 and 0.2.
0.2, respectively. The obtained critical exponents for \( x = 0.0 \) are: \( \beta = 0.49 \pm 0.01 \) and \( \gamma = 0.94 \pm 0.02 \) with \( T_c = 334.4 \pm 0.3 \) K. We notice that \( \beta \) is very close to the values obtained using the modified Arrott plot while \( \gamma \)-value has been slightly reduced but still near the theoretical mean field value. Similar results have been obtained for \( x = 0.10 \) and \( x = 0.20 \), which are shown in Figs. 10b and 11b. All values of the critical exponents extracted from MAP as well as K–F are given in Table 3. Moreover, to obtain more accurate critical exponents (\( \beta \), and \( \gamma \)) and the critical temperature \( T_c \), we have adopted the iterative method for the unsubstituted sample (CrTe) based on K–F method (Eqs. 5 and 6). The results shows, after four rounds, that the values of the critical exponents with \( T_c \) are stable and close to the standard mean field model, namely \( \beta = 0.5 \), and \( \gamma = 1 \), within the uncertainty (see Table 4).

The values of third critical exponent (\( \delta \)) can be calculated using the Widom scaling relation:

\[
\delta = 1 + \frac{\beta}{\gamma},
\]

and \( \beta \) and \( \gamma \) values obtained from MAP and K–F method [45]. The values are listed in Table 3.
Table 3 Critical exponents along with $T_c$ for CrTe$_{1-x}$Sb$_x$ (0.0 $\leq$ $x$ $\leq$ 0.2) samples

| Sample      | Model | $T_c$ (K) | $\beta$       | $\gamma$       | $\delta$ Theory | $\delta$ expt |
|-------------|-------|-----------|---------------|---------------|-----------------|---------------|
| CrTe        | MAP   | 334.0±0.1 | 0.49±0.01     | 1.04±0.02     | 3.12±0.12       | 2.95±0.01     |
|             | K–F   | 334.4±0.3 | 0.49±0.01     | 0.94±0.02     | 2.92±0.12       |               |
| CrTe$_{0.90}$Sb$_{0.10}$ | MAP | 303.3±0.1 | 0.50±0.01     | 0.99±0.01     | 2.98±0.09       | 3.11±0.01     |
|             | K–F   | 303.2±0.3 | 0.48±0.01     | 1.00±0.03     | 3.08±0.16       |               |
| CrTe$_{0.80}$Sb$_{0.20}$ | MAP | 295.4±0.1 | 0.51±0.01     | 1.00±0.01     | 2.96±0.09       | 2.90±0.01     |
|             | K–F   | 295.5±0.2 | 0.46±0.01     | 0.98±0.02     | 3.13±0.13       |               |

Table 4 Critical exponents along with $T_c$ for CrTe sample using the iterative method

| Iteration # | Initials | $T_c$ (K) | $\beta$ | $\gamma$ |
|-------------|----------|-----------|---------|----------|
| 0           | 0.5      | 334.37±0.26 | 0.49±0.01 | 0.94±0.02 |
| 1           | 0.49     | 334.63±0.52 | 0.48±0.01 | 0.96±0.03 |
| 2           | 0.48     | 333.95±0.53 | 0.48±0.01 | 0.97±0.03 |
| 3           | 0.48     | 333.75±0.54 | 0.48±0.01 | 0.98±0.03 |

Fig. 12 Modified Arrott plot of $M^{1/\beta}$ versus $(H/M)^{1/\gamma}$ at high field values with the obtained critical exponents of $\beta$ and $\gamma$ with a red linear dashed curve passed through the origin for a CrTe, b CrTe$_{0.90}$Sb$_{0.10}$, and c CrTe$_{0.80}$Sb$_{0.20}$

To illustrate the validity of the K–F analysis and the confidence in the obtained critical exponents and critical behavior, we used these critical exponents to replot the modified Arrott plots in Fig. 12. The figure shows a set of nearly perfect parallel straight lines at high field ($H$ > 2 T), indicating that the critical exponents $\beta$ and $\gamma$ values obtained using K–F method for the mean field model are quite accurate choice. Moreover, the dashed lines in Fig. 12 are a linear fitting of isothermal magnetization extrapolated near to the origin point (0,0). The obtained $T_c$ values are 334 K, 302 K and 294 K for $x$ = 0, 0.1 and 0.2, respectively. These values are in agreement with the $T_c$ values obtained from K–F analysis. This further indicates that the critical exponents are close agreement with the MFT critical exponents.

Equation 3 should yield a linear graph of the magnetic isotherms at ($T = T_c$) in a log–log presentation (Fig. 13) with a slope = $1/\delta$. The linear fit yields $\delta$ = 2.95±0.01. The result is consistent with the one obtained from Widom’s relation as shown in Table 3.
Close to the transition temperature $T_c$, the normalized equation of state can be expressed as:

$$m = f_{\pm}(h)$$

where $f_{\pm}$ are functions above ($+$) and below ($-$) $T_c$; $m \equiv |\epsilon|^{-\beta} M(H, \epsilon)$ is the normalized magnetization and $h \equiv H|\epsilon|^{-(\beta+\gamma)}$ is the normalized field.

For the correct values of the critical exponents, Eq. (7) implies that for the reliable values of $\beta$, $\gamma$ and $\delta$; two universal branches above and below $T_c$ presented as $M/|\epsilon|^{\gamma+\beta}$ vs. $H/|\epsilon|^{-(\beta+\gamma)}$ [44]. Both curves merge asymptotically at $T = T_c$. The critical exponents ($\beta$, $\gamma$ and $\delta$) obtained from Kouvel–Fisher analysis are used to represent the normalized magnetization isotherm ($m$) versus the normalized field ($h$) for all investigated samples. The scaled data for CrTe sample are presented in Fig. 14 as two branches of the magnetization above and below $T_c$. At high fields, both branches are converging toward each other at $T_c$. Figure 14b shows the data in log–log scale, which vividly reveals the merge of the data giving further evidence for the reliability of the critical exponents’ values within the mean field model. The scaling analysis has been applied for CrTe$_{1-x}$Sb$_x$ for $x = 0.10$ and $x = 0.20$. The results are shown in
Fig. 15  
(a) Universal scaling behavior above and below $T_c$ for $\text{CrTe}_{0.90}\text{Sb}_{0.10}$.  
(b) log–log scale representation.

Fig. 16  
(a) Universal scaling behavior above and below $T_c$ for $\text{CrTe}_{0.80}\text{Sb}_{0.20}$.  
(b) log–log scale representation.

Figs. 15 and 16, respectively. The well-scaled data for $x = 0.20$ in the current work comparing to the scaled data published in [29] further confirm the differences and improvements in the experiment conditions.

4.1 Conclusion

We investigated the structural and critical behavior of $\text{CrTe}_{1-x}\text{Sb}_x$ with varying Sb contents. Antimony substitution resulted in pure NiAs-hexagonal structure with P63/mmc (194) space group. The critical exponents ($\beta$, $\gamma$, and $\delta$) follow mean field model for all samples with different Sb concentrations. The magnetization isotherms near the ferromagnetic transition follow a universal scaling behavior, giving further support for the mean field behavior in $\text{CrTe}_{1-x}\text{Sb}_x$ and confirming the obtained values of the critical exponents.
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