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Anisotropic magnetic entropy change in $\text{Cr}_2\text{X}_2\text{Te}_6$ ($\text{X} = \text{Si}$ and Ge)

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Intrinsic, two-dimensional (2D) ferromagnetic semiconductors are an important class of materials for spintronics applications. $\text{Cr}_2\text{X}_2\text{Te}_6$ ($\text{X} = \text{Si}$ and Ge) semiconductors show 2D Ising-like ferromagnetism, which is preserved in few-layer devices. The maximum magnetic entropy change associated with the critical properties around the ferromagnetic transition for $\text{Cr}_2\text{Si}_2\text{Te}_6$ $-\Delta S_M^{max} \sim 5.05 \text{ J kg}^{-1} \text{ K}^{-1}$ is much larger than $-\Delta S_M^{max} \sim 2.64 \text{ J kg}^{-1} \text{ K}^{-1}$ for $\text{Cr}_2\text{Ge}_2\text{Te}_6$ with an out-of-plane field change of 5 T. The rescaled $-\Delta S_M(T, H)$ curves collapse onto a universal curve independent of temperature and field for both materials. This indicates similar critical behavior and 2D Ising magnetism, confirming the magnetocrystalline anisotropy that could preserve the long-range ferromagnetism in few-layers of $\text{Cr}_2\text{X}_2\text{Te}_6$.

I. INTRODUCTION

Layered ferromagnets such as $\text{Cr}_2\text{Ge}_2\text{Te}_6$, CrI_3 , and Fe_3GeTe_2 have recently attracted considerable attention since long-range ferromagnetism (FM) persists in atomically thin devices.¹⁻⁶ Intrinsic magnetic order is not allowed at finite temperature in the two-dimensional (2D) isotropic Heisenberg model by the Mermin-Wagner theorem⁷, however large magnetocrystalline anisotropy in van der Waals (vdW) magnets would lift this restriction.

Bulk $\text{Cr}_2\text{X}_2\text{Te}_6$ ($\text{X} = \text{Si}$ and Ge) exhibit FM below the Curie temperature (T_c) of 32 K for $\text{Cr}_2\text{Si}_2\text{Te}_6$ and 61 K for $\text{Cr}_2\text{Ge}_2\text{Te}_6$, respectively, and show large magnetocrystalline anisotropy as a result of strong spin-orbit coupling (SOC).⁸⁻¹² Neutron scattering measurements showed that bulk $\text{Cr}_2\text{Si}_2\text{Te}_6$ is a strongly anisotropic 2D Ising-like ferromagnet with a critical exponent $\beta = 0.17$ and a spin gap of $\sim 6 \text{ meV}$.¹³ On the other hand, recently observed $\beta = 0.151$ and a much smaller spin gap of $\sim 0.075 \text{ meV}$ argue that the spins in $\text{Cr}_2\text{Si}_2\text{Te}_6$ are Heisenberg-like.¹⁴ $\text{Cr}_2\text{Ge}_2\text{Te}_6$ is proposed to be a 2D Heisenberg ferromagnet based on spin wave theory,⁴ but was also found to follow the tricritical mean-field model,¹⁵ calling for further studies. The magnetocaloric effect (MCE) in the FM vdW materials is also of interest since it can give insight into the magnetic properties. $\text{Fe}_{3-x}\text{GeTe}_2$ with $T_c = 225 \text{ K}$ shows the maximum value of magnetic entropy change $-\Delta S_M^{max}$ about $1.1 \text{ J kg}^{-1} \text{ K}^{-1}$ at 5 T.¹⁶ CrI_3 exhibits anisotropic $-\Delta S_M^{max}$ with values of 4.24 and $2.68 \text{ J kg}^{-1} \text{ K}^{-1}$ at 5 T for $\mathbf{H} // \mathbf{c}$ and $\mathbf{H} // \mathbf{ab}$, respectively.¹⁷

In this work we studied the anisotropic magnetocaloric effect associated with the critical behavior of $\text{Cr}_2\text{X}_2\text{Te}_6$ ($\text{X} = \text{Si}$ and Ge) single crystals. The magnetocrystalline anisotropy constant K_u is temperature-dependent, and is evidently larger for $\text{Cr}_2\text{Si}_2\text{Te}_6$ when compared to $\text{Cr}_2\text{Ge}_2\text{Te}_6$. The maximum magnetic entropy change in out-of-plane field up to 5 T $-\Delta S_M^{max} \sim 5.05 \text{ J kg}^{-1} \text{ K}^{-1}$ for $\text{Cr}_2\text{Si}_2\text{Te}_6$ is nearly double of $-\Delta S_M^{max} \sim 2.64 \text{ J kg}^{-1} \text{ K}^{-1}$ for $\text{Cr}_2\text{Ge}_2\text{Te}_6$. Critical exponents β , γ , and δ and critical isotherm analysis suggest 2D Ising-like spins.

This is further confirmed by the scaling analysis of magnetic entropy change $-\Delta S_M(T, H)$, in which the rescaled $-\Delta S_M(T, H)$ collapse on a universal curve. Our work provides evidence for magnetocrystalline anisotropy that drives the 2D Ising ferromagnetic state in few layers of $\text{Cr}_2\text{X}_2\text{Te}_6$ ($\text{X} = \text{Si}$ and Ge).

II. EXPERIMENTAL DETAILS

Single crystals of $\text{Cr}_2\text{X}_2\text{Te}_6$ ($\text{X} = \text{Si}$ and Ge) were fabricated by the self-flux technique starting from an intimate mixture of pure elements Cr (3N, Alfa Aesar) powder, Si or Ge (5N, Alfa Aesar) pieces and Te (5N, Alfa Aesar) pieces with a molar ratio of 1 : 2 : 6. The starting materials were vacuum-sealed in a quartz tube, heated to $1100 \text{ }^\circ\text{C}$ over 20 h, held at $1100 \text{ }^\circ\text{C}$ for 3 h, and then cooled to $680 \text{ }^\circ\text{C}$ at a rate of $1 \text{ }^\circ\text{C/h}$. The x-ray diffraction (XRD) data were taken with $\text{Cu } K_\alpha$ ($\lambda = 0.15418 \text{ nm}$) radiation of a Rigaku Miniflex powder diffractometer. The dc magnetization was collected in Quantum Design MPMS-XL5 system. The magnetic entropy change $-\Delta S_M(T, H)$ from the dc magnetization data was estimated using the Maxwell relation.

III. RESULTS AND DISCUSSION

A. Structural and basic magnetization data

Bulk $\text{Cr}_2\text{X}_2\text{Te}_6$ ($\text{X} = \text{Si}$ and Ge) were first synthesized by Carreaux *et al.*^{9,10} They crystalize in a layered structure [Fig. 1(a)]. The Cr ions are located at the centers of slightly distorted octahedra of Te atoms. The short X-X bonds result in X-X dimers forming an ethane-like X_2Te_6 groups, similar to P-P dimers in CdPS_3 .¹⁸ Figure 1(c) presents the single crystal x-ray diffraction (XRD) data. The observed (001) peaks distinctly shift to higher angles in $\text{Cr}_2\text{Ge}_2\text{Te}_6$ when compared to $\text{Cr}_2\text{Si}_2\text{Te}_6$ indicating a smaller vdW gap in $\text{Cr}_2\text{Ge}_2\text{Te}_6$. The powder XRD data can be indexed in the $R\bar{3}h$ space group [Fig. 1(d)]. The determined lattice parameters are $a = 6.772(2) \text{ \AA}$

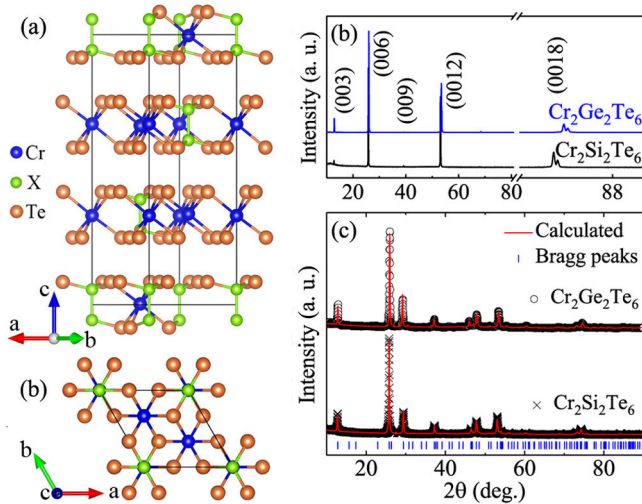


FIG. 1. (Color online) Crystal structure of $\text{Cr}_2\text{X}_2\text{Te}_6$ ($\text{X} = \text{Si}$ and Ge) from (a) side and (b) top views. (c) Single crystal x-ray diffraction (XRD) and (d) powder XRD patterns of $\text{Cr}_2\text{X}_2\text{Te}_6$ ($\text{X} = \text{Si}$ and Ge). The vertical tick marks represent Bragg reflections of the $R\bar{3}h$ space group.

TABLE I. The parameters obtained from fits of the $1/M$ vs T data for $\text{Cr}_2\text{X}_2\text{Te}_6$ ($\text{X} = \text{Si}$ and Ge) single crystals.

	Fit T range (K)	C (emu K/mol)	θ_p (K)	μ_{eff} (μ_B/Cr)
$\text{X} = \text{Si}$				
$\mathbf{H} // \mathbf{c}$	$130 \leq T \leq 300$	1.70(1)	63(1)	3.68(2)
$\mathbf{H} // \mathbf{ab}$	$130 \leq T \leq 300$	1.60(1)	63(1)	3.57(1)
$\text{X} = \text{Ge}$				
$\mathbf{H} // \mathbf{c}$	$150 \leq T \leq 300$	1.22(1)	114(2)	3.12(1)
$\mathbf{H} // \mathbf{ab}$	$150 \leq T \leq 300$	1.54(2)	101(1)	3.51(2)

and $c = 20.671(2)$ Å for $\text{Cr}_2\text{Si}_2\text{Te}_6$ [$a = 6.826(2)$ Å and $c = 20.531(2)$ Å for $\text{Cr}_2\text{Ge}_2\text{Te}_6$], in agreement with the reported values.^{9,10}

Figure 2(a) presents the temperature dependence of the zero field cooling (ZFC) magnetization $M(T)$ measured in $H = 10$ kOe applied in the ab plane and parallel to the c axis, respectively. The FM transition stems from the near-90° Cr-Te-Cr superexchange interaction and is observed in both materials. An apparent bifurcation at low temperature is observed in $\text{Cr}_2\text{Si}_2\text{Te}_6$. The absence of bifurcation in $\text{Cr}_2\text{Ge}_2\text{Te}_6$ indicates smaller magnetic anisotropy. The smaller vdW gap and larger in-plane Cr-Cr distance in $\text{Cr}_2\text{Ge}_2\text{Te}_6$ contribute to the enhancement of the T_c from 32 K for $\text{Cr}_2\text{Si}_2\text{Te}_6$ to 63 K for $\text{Cr}_2\text{Si}_2\text{Te}_6$. The $1/M$ vs T curves at high temperature follow the Curie-Weiss law, $\chi(T) = M/H = C/(T - \theta_p)$, where χ is magnetic susceptibility, M is magnetization, C is the Curie constant, and θ_p is the Weiss temperature. The obtained parameters C and θ_p are listed in Table I. The positive Weiss temperatures θ_p , nearly twice the values of T_c , for both directions suggest strong short-range FM

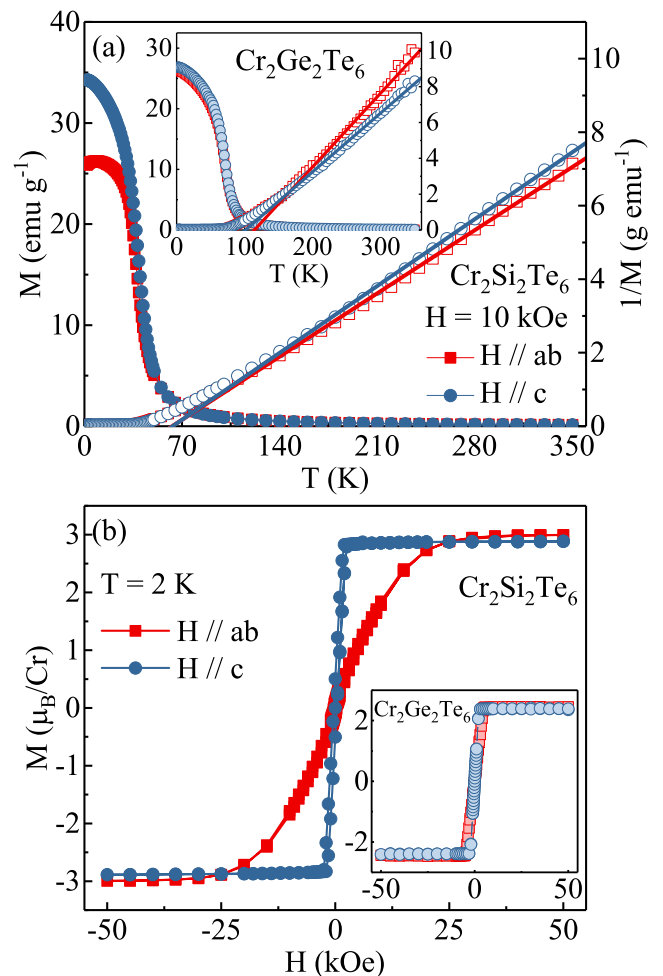


FIG. 2. (Color online) (a) Temperature dependence of zero field cooling (ZFC) magnetic susceptibility χ (left axis) and corresponding $1/\chi$ (right axis) for $\text{Cr}_2\text{X}_2\text{Te}_6$ ($\text{X} = \text{Si}$ and Ge) measured in in-plane and out-of-plane field of $H = 10$ kOe. (b) Field dependence of magnetization measured at $T = 2$ K.

correlation in $\text{Cr}_2\text{X}_2\text{Te}_6$ ($\text{X} = \text{Si}$ and Ge) above T_c . The effective magnetic moment $\mu_{eff} \approx \sqrt{8C}$ is also listed in Table I. The values are close to the theoretical value expected for Cr^{3+} of $3.87\mu_B$. The isothermal magnetization at $T = 2$ K is shown in Fig. 2(b). We estimate the saturation magnetization M_s from the intercept of a linear fit of $M(H)$ at high field and the saturation field H_s as the point of deviation from the linear behavior. The derived $M_s \approx 2.86(1) \mu_B/\text{Cr}$ with out-of-plane field for $\text{Cr}_2\text{Si}_2\text{Te}_6$ is larger than that of $2.40(2) \mu_B/\text{Cr}$ for $\text{Cr}_2\text{Ge}_2\text{Te}_6$. The saturation field $H_s \approx 3$ kOe with out-of-plane field is smaller than $H_s \approx 5$ kOe with in-plane field and is much smaller than that of 25 kOe for $\text{Cr}_2\text{Si}_2\text{Te}_6$. These results confirm the easy c -axis and smaller magnetic anisotropy in $\text{Cr}_2\text{Ge}_2\text{Te}_6$, in agreement with previous reports.⁹⁻¹²

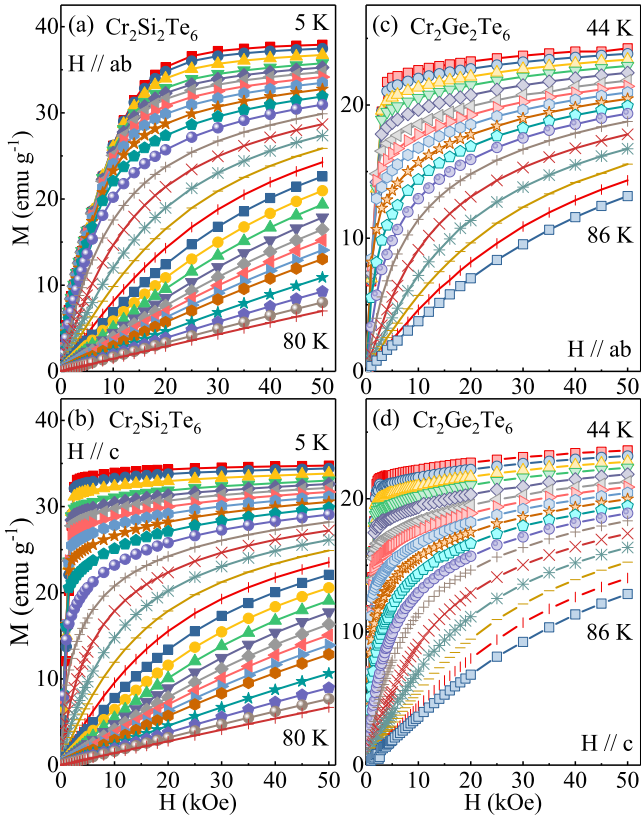


FIG. 3. (Color online) Typical initial isothermal magnetization curves measured in (a,c) $\mathbf{H} // \mathbf{ab}$ and (b,d) $\mathbf{H} // \mathbf{c}$ around T_c for $\text{Cr}_2\text{X}_2\text{Te}_6$ ($X = \text{Si}$ and Ge).

B. Magnetocrystalline anisotropy

Figure 3 shows the magnetization isotherms with field up to 50 kOe applied for both $\mathbf{H} // \mathbf{ab}$ and $\mathbf{H} // \mathbf{c}$ around T_c for $\text{Cr}_2\text{X}_2\text{Te}_6$ ($X = \text{Si}$ and Ge). When $\mathbf{H} // \mathbf{ab}$, the saturation field H_s is associated with the uniaxial magnetocrystalline anisotropy parameter K_u and the saturation magnetization M_s , i.e., $2K_u/M_s = \mu_0 H_s$, where μ_0 is the vacuum permeability.¹⁹ The temperature dependence of K_u as well as M_s and H_s for $\text{Cr}_2\text{X}_2\text{Te}_6$ ($X = \text{Si}$ and Ge) are depicted in Fig. 4. The calculated K_u for $\text{Cr}_2\text{Si}_2\text{Te}_6$ is about 61 kJ/m^3 at 5 K. It gradually decreases to 38 kJ/m^3 at $T_c = 32 \text{ K}$, comparable with the K_u values in CrBr_3 .²⁰ The anisotropy parameter K_u is much lower for $\text{Cr}_2\text{Ge}_2\text{Te}_6$: about 12 kJ/m^3 at 44 K and 5.6 kJ/m^3 at $T_c = 63 \text{ K}$. For clarity, only the K_u values from T_c to 44 K are presented for $\text{Cr}_2\text{Ge}_2\text{Te}_6$. The K_u of 20 kJ/m^3 at 2 K for $\text{Cr}_2\text{Ge}_2\text{Te}_6$ estimated from Fig. 2(b) is much smaller than that of 65 kJ/m^3 for $\text{Cr}_2\text{Si}_2\text{Te}_6$, in line with the magnetization data. The observed decrease of K_u with increasing temperature arises solely from a large number of local spin clusters fluctuating randomly around the macroscopic magnetization vector and activated by a nonzero thermal energy, whereas the anisotropy constants are temperature-independent.^{21,22} This provides

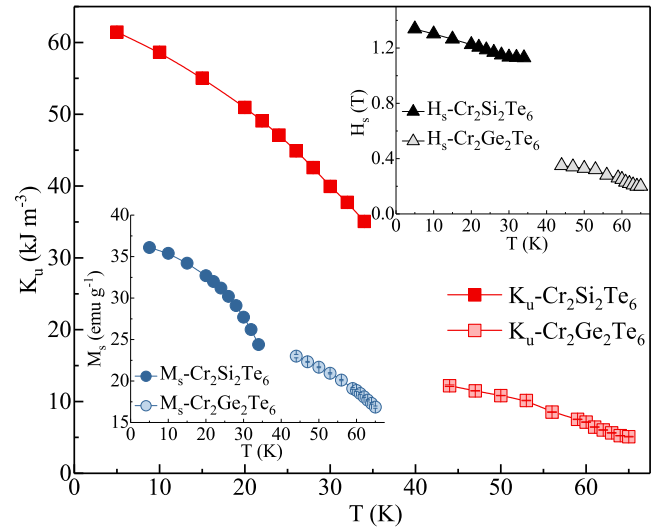


FIG. 4. (Color online) Temperature dependence of the calculated anisotropy constant K_u , the estimated saturation field H_s and the saturation magnetization M_s (insets) below T_c for $\text{Cr}_2\text{X}_2\text{Te}_6$ ($X = \text{Si}$ and Ge).

insight into the understanding of FM in few-layers of $\text{Cr}_2\text{X}_2\text{Te}_6$ ($X = \text{Si}$ and Ge). While in pure 2D system no long-range magnetic order is expected,⁷ mechanical corrugations and magnetic anisotropy are possible pathways to establish magnetism in few-layers samples.

C. Magnetic entropy change

We estimate the magnetic entropy change

$$\Delta S_M(T, H) = \int_0^H \left(\frac{\partial S}{\partial H} \right)_T dH = \int_0^H \left(\frac{\partial M}{\partial T} \right)_H dH, \quad (1)$$

where $\left(\frac{\partial S}{\partial H} \right)_T = \left(\frac{\partial M}{\partial T} \right)_H$ is based on Maxwell's relation. In the case of magnetization measured at small discrete field and temperature intervals [Fig. 3], ΔS_M can be approximated:

$$\Delta S_M(T_i, H) = \frac{\int_0^H M(T_i, H) dH - \int_0^H M(T_{i+1}, H) dH}{T_i - T_{i+1}}. \quad (2)$$

Figures 5(a) and 5(b) present the calculated $-\Delta S_M(T, H)$ as a function of temperature with in-plane and out-of-plane fields. All the $-\Delta S_M(T, H)$ curves show a pronounced peak at T_c , and the peak broadens asymmetrically on both sides with increasing field. The maximum value of $-\Delta S_M$ is $4.9 \text{ J kg}^{-1} \text{ K}^{-1}$ for $\text{Cr}_2\text{Si}_2\text{Te}_6$ and $2.6 \text{ J kg}^{-1} \text{ K}^{-1}$ for $\text{Cr}_2\text{Ge}_2\text{Te}_6$ with in-plane field change of 5 T. These slightly increase to 5.05 and $2.64 \text{ J kg}^{-1} \text{ K}^{-1}$, respectively, with out-of-plane field change of 5 T. The obtained $-\Delta S_M$ values are comparable and larger than that of Fe_3GeTe_2 and CrI_3 .^{16,17} The rotational magnetic entropy change ΔS_M^R is calculated as $\Delta S_M^R(T, H) = \Delta S_M(T, H_c) - \Delta S_M(T, H_{ab})$.

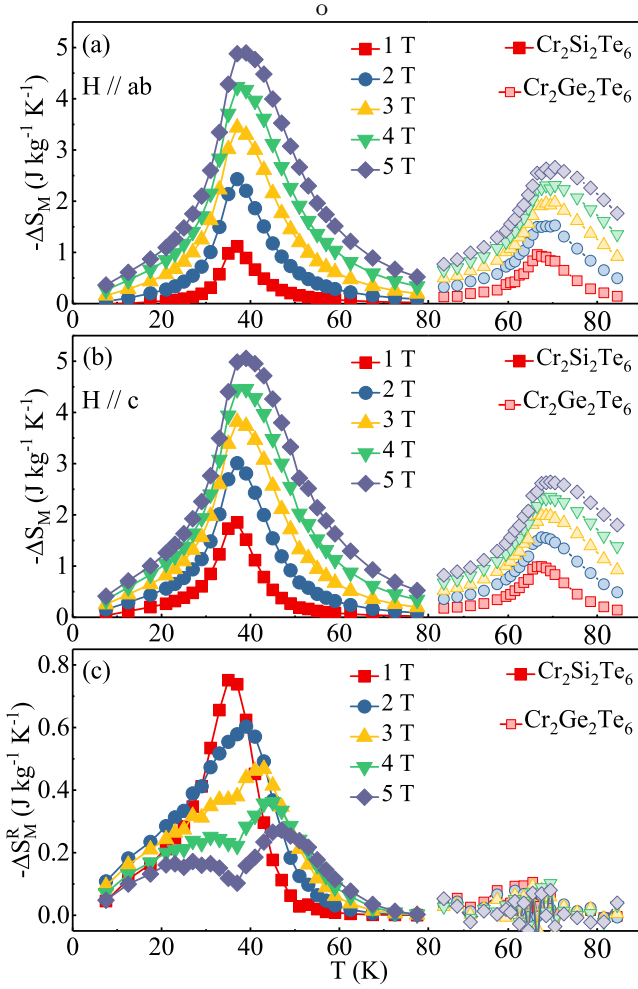


FIG. 5. (Color online) Temperature dependence of isothermal magnetic entropy change $-\Delta S_M$ obtained from magnetization at various magnetic fields change (a) in the ab plane and (b) along the c axis, respectively, for $\text{Cr}_2\text{X}_2\text{Te}_6$ ($X = \text{Si}$ and Ge). (c) Temperature dependence of $-\Delta S_M^R$ obtained by rotating from the ab plane to the c axis in various fields.

As shown in Fig. 5(c), the value of ΔS_M^R for $\text{Cr}_2\text{Si}_2\text{Te}_6$ is larger than that for $\text{Cr}_2\text{Ge}_2\text{Te}_6$, in line with the calculated K_u [Fig. 4]. The anisotropy is gradually suppressed in higher field, and interestingly, it splits into two peaks on both sides of T_c with field above 3 T for $\text{Cr}_2\text{Si}_2\text{Te}_6$.

D. Critical behavior

According to the scaling hypothesis, the second-order phase transition around T_c can be characterized by a set of interrelated critical exponents and magnetic equation of state.²⁴ The exponents β and γ can be obtained from spontaneous magnetization M_{sp} and inverse initial susceptibility χ_0^{-1} , below and above T_c , respectively, while δ is a critical isotherm exponent at T_c . The mathematical

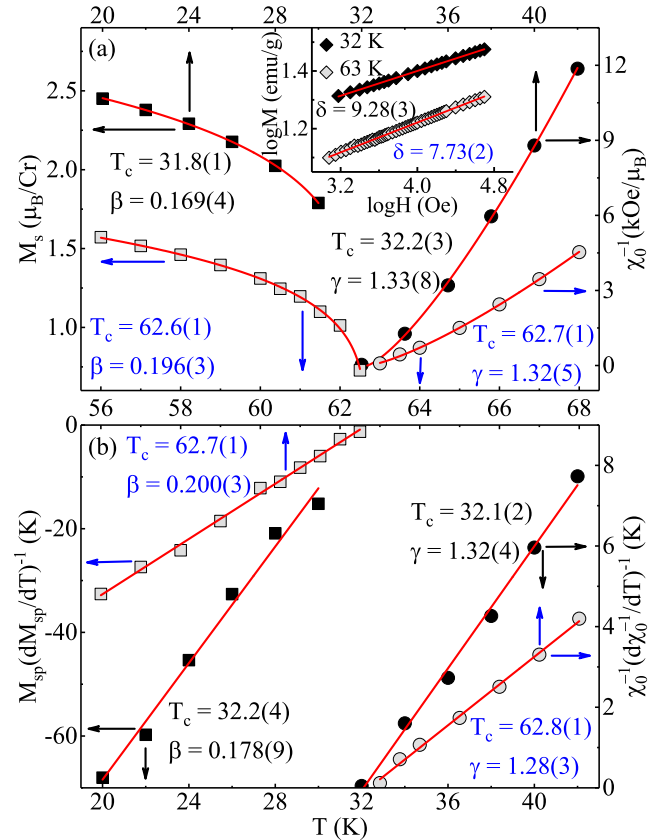


FIG. 6. (Color online) (a) Temperature dependence of the spontaneous magnetization M_{sp} (left axis) and the inverse initial susceptibility χ_0^{-1} (right axis) in out-of-plane field with solid fitting curves for $\text{Cr}_2\text{X}_2\text{Te}_6$ ($X = \text{Si}$ and Ge). Inset shows $\log M$ vs $\log H$ collected at T_c with linear fitting curves. (b) Kouvel-Fisher plots of $M_{sp}(dM_{sp}/dT)^{-1}$ (left axis) and $\chi_0^{-1}(d\chi_0^{-1}/dT)^{-1}$ (right axis) with solid fitting curves.

definitions of the exponents from magnetization measurement are given below:

$$M_{sp}(T) = M_0(-\varepsilon)^\beta, \varepsilon < 0, T < T_c, \quad (3)$$

$$\chi_0^{-1}(T) = (h_0/m_0)\varepsilon^\gamma, \varepsilon > 0, T > T_c, \quad (4)$$

$$M = DH^{1/\delta}, T = T_c, \quad (5)$$

where $\varepsilon = (T - T_c)/T_c$ is the reduced temperature, and M_0 , h_0/m_0 and D are the critical amplitudes.²⁵

The critical exponents β , γ , and δ , as well as the precise T_c can be obtained by the modified Arrott plot of $M^{1/\beta}$ vs $(H/M)^{1/\gamma}$ in the vicinity of T_c with a self-consistent method.^{26,27} This gives $\chi_0^{-1}(T)$ and $M_{sp}(T)$ as the intercepts on the H/M axis and the positive M^2 axis, respectively. Figure 6(a) presents the final $M_{sp}(T)$ and $\chi_0^{-1}(T)$ as a function of temperature. According to Eqs. (3) and (4), the critical exponents $\beta = 0.169(4)$ with $T_c = 31.8(1)$ K [$\beta = 0.196(3)$ with $T_c = 62.6(1)$ K], and $\gamma = 1.33(8)$

TABLE II. Critical exponents of $\text{Cr}_2\text{X}_2\text{Te}_6$ ($\text{X} = \text{Si}$ and Ge). The MAP, KFP and CI represent the modified Arrott plot, the Kouvel-Fisher plot and the critical isotherm, respectively.

	β	γ	δ	n	m
$\text{X} = \text{Si}$					
$-\Delta S_M^{max}$				0.52(2)	
RCP					1.09(1)
MAP	0.169(4)	1.33(8)	8.9(3)	0.45(3)	1.112(4)
KFP	0.178(9)	1.32(4)	8.4(2)	0.45(3)	1.119(3)
CI			9.28(3)		1.108(1)
$\text{X} = \text{Ge}$					
$-\Delta S_M^{max}$				0.51(1)	
RCP					1.13(1)
MAP	0.196(3)	1.32(5)	7.7(2)	0.47(2)	1.130(3)
KFP	0.200(3)	1.28(3)	7.4(1)	0.46(1)	1.135(2)
CI			7.73(2)		1.129(1)

with $T_c = 32.2(3)$ K [$\gamma = 1.32(5)$ with $T_c = 62.7(1)$ K], are obtained for $\text{Cr}_2\text{Si}_2\text{Te}_6$ [$\text{Cr}_2\text{Ge}_2\text{Te}_6$].

Based on the Kouvel-Fisher (KF) relation:²⁸

$$M_{sp}(T)[dM_{sp}(T)/dT]^{-1} = (T - T_c)/\beta, \quad (6)$$

$$\chi_0^{-1}(T)[d\chi_0^{-1}(T)/dT]^{-1} = (T - T_c)/\gamma. \quad (7)$$

Linear fitting to the plots of $M_{sp}(T)[dM_{sp}(T)/dT]^{-1}$ vs T and $\chi_0^{-1}(T)[d\chi_0^{-1}(T)/dT]^{-1}$ vs T , as shown in Fig. 6(b), yield $\beta = 0.178(9)$ with $T_c = 32.2(4)$ K [$\beta = 0.200(3)$ with $T_c = 62.7(1)$ K], and $\gamma = 1.32(4)$ with $T_c = 32.1(2)$ K [$\gamma = 1.28(3)$ with $T_c = 62.8(1)$ K]. The third exponent δ can be calculated from the Widom scaling relation $\delta = 1 + \gamma/\beta$. From β and γ obtained with the modified Arrott plot and the Kouvel-Fisher plot, $\delta = 8.9(3)$ and $8.4(2)$ [7.7(2) and 7.4(1)] for $\text{Cr}_2\text{Si}_2\text{Te}_6$ [$\text{Cr}_2\text{Ge}_2\text{Te}_6$], which are close to the direct fits of δ taking into account that $M = DH^{1/\delta}$ at T_c [$\delta = 9.28(3)$ at 32 K and 7.73(2) at 63 K, inset in Fig. 6(a)]. The critical exponents of $\text{Cr}_2\text{X}_2\text{Te}_6$ ($\text{X} = \text{Si}$ and Ge) are summarized in Table II. They are close to but not identical to values expected for the 2D-Ising model ($\beta = 0.125$, $\gamma = 1.75$ and $\delta = 15$). This deviation is most likely associated with non-negligible interlayer coupling and spin-lattice coupling in this system.^{11,13}

E. Scaling analysis of the ΔS_M data

For a material displaying a second-order transition,²⁹ the field dependence of the maximum magnetic entropy change shows a power law $-\Delta S_M^{max} = aH^n$,³⁰ where a is a constant and the exponent n at T_c is related to the critical exponents as $n(T_c) = 1 + (\beta - 1)/(\beta + \gamma)$. Another important parameter is the relative cooling power (RCP): $RCP = -\Delta S_M^{max} \times \delta T_{FWHM}$ where $-\Delta S_M^{max}$ is

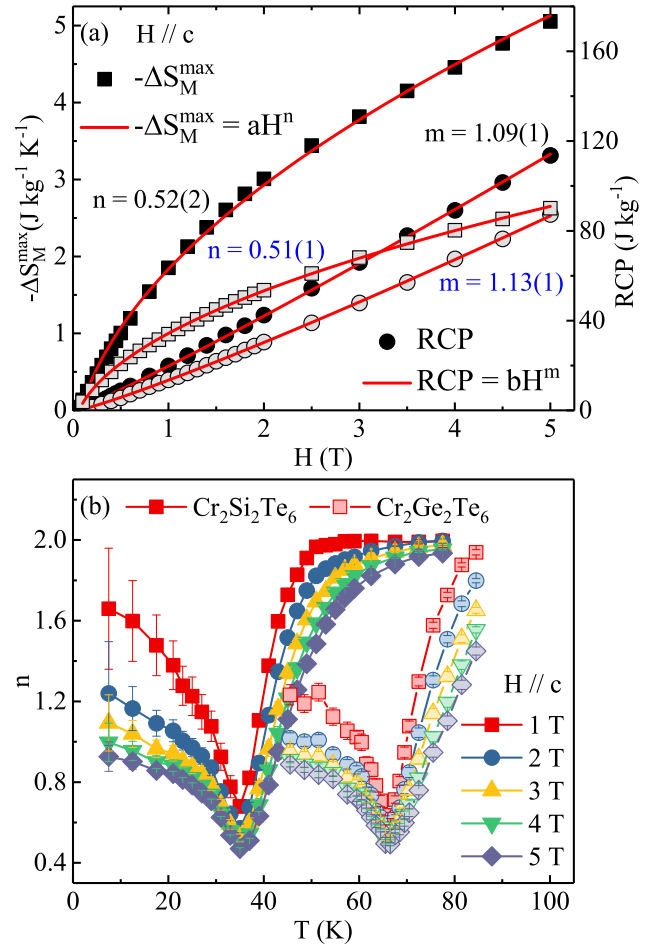


FIG. 7. (Color online) (a) Field dependence of the maximum magnetic entropy change $-\Delta S_M^{max}$ and the relative cooling power RCP with power law fitting in red solid lines for $\text{Cr}_2\text{X}_2\text{Te}_6$ ($\text{X} = \text{Si}$ and Ge). (b) Temperature dependence of n in various fields.

the maximum entropy change near T_c and δT_{FWHM} is the full-width at half maximum.³¹ The RCP also depends on the magnetic field with $RCP = bH^m$, where b is a constant and m is associated with the critical exponent δ , $m = 1 + 1/\delta$.

Figure 7(a) presents the summary of the out-of-plane field dependence of $-\Delta S_M^{max}$ and RCP. The calculated values of RCP are about 114 and 87 J kg^{-1} for $\text{Cr}_2\text{Si}_2\text{Te}_6$ and $\text{Cr}_2\text{Ge}_2\text{Te}_6$, respectively, with out-of-plane field change of 5 T. Fitting of the $-\Delta S_M^{max}$ gives that $n = 0.52(2)$ and $0.51(1)$ for $\text{Cr}_2\text{Si}_2\text{Te}_6$ and $\text{Cr}_2\text{Ge}_2\text{Te}_6$, respectively [Fig. 7(a)], which is quite close to that of $n = 0.53$ for the 2D-Ising model ($\beta = 0.125$, $\gamma = 1.75$). Fitting of the RCP generates $m = 1.09(1)$ and $1.13(1)$ [Fig. 7(a)], which is also close to the expected value of 1.07 for 2D-Ising model ($\delta = 15$). Figure 7(b) displays the temperature dependence of $n(T)$ in various fields. All $n(T)$ curves follow a universal behavior.³² At low temperatures, well below T_c , n has a value around 1. On the other side, well above T_c , n is close to 2 as a consequence

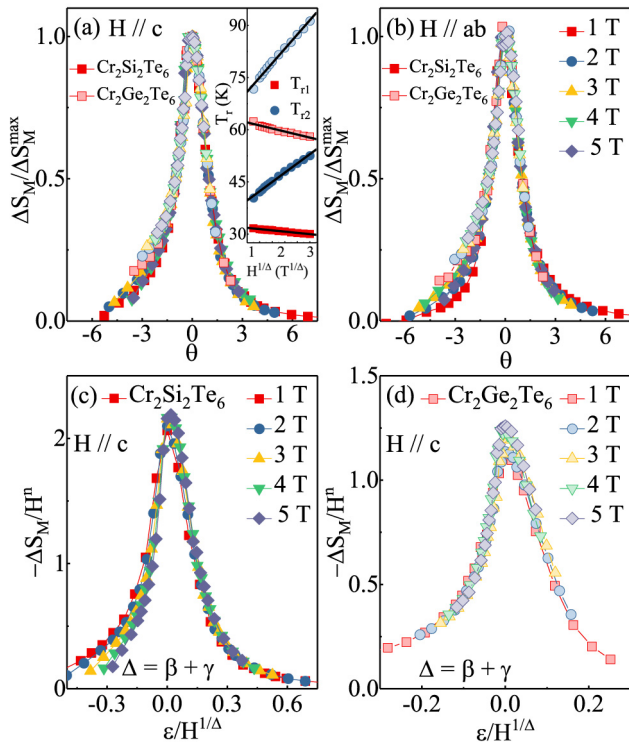


FIG. 8. (Color online) The normalized ΔS_M as a function of the reduced temperature θ with (a) out-of-plane and (b) in-plane field for $\text{Cr}_2\text{X}_2\text{Te}_6$ ($X = \text{Si}$ and Ge). Scaling plot for (c) $\text{Cr}_2\text{Si}_2\text{Te}_6$ and (d) $\text{Cr}_2\text{Ge}_2\text{Te}_6$ based on the critical exponents β and γ obtained in out-of-plane field.

of the Curie-Weiss law. At $T = T_c$, $n(T)$ has a minimum.

Scaling analysis of $-\Delta S_M$ can be built by normalizing all the $-\Delta S_M$ curves against the respective maximum $-\Delta S_M^{max}$, namely, $\Delta S_M/\Delta S_M^{max}$ by rescaling the reduced temperature θ_{\pm} as defined in the following equations,³³

$$\theta_- = (T_{peak} - T)/(T_{r1} - T_{peak}), T < T_{peak}, \quad (8)$$

$$\theta_+ = (T - T_{peak})/(T_{r2} - T_{peak}), T > T_{peak}, \quad (9)$$

where T_{r1} and T_{r2} are the temperatures of two reference points that corresponds to $\Delta S_M(T_{r1}, T_{r2}) = \frac{1}{2}\Delta S_M^{max}$. Following this method, all the $-\Delta S_M(T, H)$ curves in

various fields collapse into a single curve in the vicinity of T_c for $\text{Cr}_2\text{X}_2\text{Te}_6$ ($X = \text{Si}$ and Ge), as shown in Figs. 8(a) and 8(b). The values of T_{r1} and T_{r2} depend on $H^{1/\Delta}$ with $\Delta = \beta + \gamma$ [inset in Fig. 8(a)].

In the phase transition region, the scaling analysis of $-\Delta S_M$ can also be expressed as

$$\frac{-\Delta S_M}{a_M} = H^n f\left(\frac{\varepsilon}{H^{1/\Delta}}\right), \quad (10)$$

where $a_M = T_c^{-1}A^{\delta+1}B$ with A and B representing the critical amplitudes as in $M_{sp}(T) = A(-\varepsilon)^\beta$ and $H = BM^\delta$, respectively, and $f(x)$ is the scaling function.³⁴ If the critical exponents are appropriately chosen, the $-\Delta S_M$ vs T curves should be rescaled into a single curve, consistent with normalizing the $-\Delta S_M$ curves with two reference temperatures (T_{r1} and T_{r2}). As shown in Figs. 8(c) and 8(d), the rescaled $-\Delta S_M$ for $\text{Cr}_2\text{X}_2\text{Te}_6$ ($X = \text{Si}$ and Ge) with out-of-plane field collapse onto a single curve, confirming the reliable critical exponents and 2D Ising behavior for $\text{Cr}_2\text{X}_2\text{Te}_6$ ($X = \text{Si}$ and Ge).

IV. CONCLUSIONS

In summary, we have studied the critical behavior and magnetocaloric effect around the FM-PM transition in $\text{Cr}_2\text{X}_2\text{Te}_6$ ($X = \text{Si}$ and Ge) single crystals. The critical exponents β , γ , and δ estimated from various techniques match reasonably well and the scaling analysis of magnetic entropy change confirms that they are 2D Ising ferromagnets with non-negligible interlayer coupling. The uniaxial magnetocrystalline anisotropy confirmed here could be the possible origin of existence of long-range FM in few-layers of $\text{Cr}_2\text{X}_2\text{Te}_6$ ($X = \text{Si}$ and Ge).

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