

Thermodynamic and transport properties of high-quality single crystals of the altermagnet CrSb

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Altermagnetism (AM) is an emerging magnetic order unifying essential characteristics of ferromagnetic and antiferromagnetic states. The CrSb has attracted significant interest owing to its large altermagnetic spin-splitting energy. In this paper, we present the growth details of high-quality single crystals of CrSb using the self-flux method and investigate their physical properties. We obtained large (001) oriented hexagonal crystals, up to $2 \times 2.5 \times 1 \text{ mm}^3$ in size with residual resistivity ratio ~ 11 . A pronounced positive magnetoresistance of up to 80% is observed at 3.5 K. Most strikingly, the room temperature specific heat value exceeds the Dulong-Petit limit, being attributed to a broad magnon contribution from the altermagnetic order of CrSb. The specific heat fit reveals a magnon energy gap $\sim 16 \pm 1 \text{ meV}$. Further, ac susceptibility measurements demonstrate the absence of superconductivity down to 0.1 K. These findings underscore CrSb as a viable altermagnet for room temperature magnonic and spintronic applications.

INTRODUCTION

A new magnetic state termed altermagnetism (AM) has recently been proposed through the reclassification of antiferromagnets (AFMs) within the framework of spin groups [1–4]. Among the wide variety of altermagnets (AMs) predicted theoretically [2], RuO₂ [5–9], and MnTe [10] have been investigated most extensively. Although CrSb shows a remarkably high magnetic ordering temperature near 700 K [11, 12] and substantial spin-splitting energy [2, 13], it has drawn relatively less attention. CrSb adopts a hexagonal NiAs-type crystal structure (space group: $P6_3/mmc$) [11–19] and exhibits a collinear antiferromagnetic ground state [20–24], with Cr moments oriented along the c -axis, ferromagnetically aligned within each hexagonal plane and antiferromagnetically coupled between neighboring planes, as shown in Fig. 1a. The altermagnetic order in CrSb can be well described within the framework of nonrelativistic spin-group symmetries. A spin space group operation is defined as a combined transformation in spin and real space, defined as $[R_i^s || R_j]$, where R_i^s acts exclusively on the spin degrees of freedom (restricted to the identity E and the spin-flip symmetry operation C_2^s) and R_j denotes a spatial symmetry operation [1–4, 25]. The opposite-spin sublattices in altermagnet must be related by a nontrivial spatial symmetry—such as an n -fold rotation (screw) or mirror ($\mathcal{M}$) symmetry (glide) instead of translation ($\vec{t}$) or inversion (P). The antiparallel Cr sublattices in CrSb are connected by the spin group symmetries $[C_2^s || \mathcal{M}_z]$ and $[C_2^s || \vec{C}_{6z}]$, where $\vec{C}_{6z}$ denotes a sixfold screw operation about the z axis accompanied by a half-unit-cell translation ($\vec{t} = (0, 0, \frac{c}{2})$), as shown in Fig. 1b. Breaking of the spin group symmetries $[C_2^s || \vec{t}]$ and space-time reversal symmetry $[\mathcal{T} || P]$ in CrSb leads to momentum-

dependent spin-splitting of the electronic bands [1–4, 25]. This symmetry identifies CrSb as a bulk g -wave altermagnet [2], characterized by spin-split electronic bands that reverse polarity six times in the k_x - k_y plane around k_z axis. Such altermagnetic material retains key advantages of antiferromagnets—such as ultrafast spin dynamics and weak sensitivity to external magnetic fields—making it a promising platform for novel spin-to-charge conversion, spin-torque effects, and future spintronic or magneto-optical applications [2, 8, 26–28].

The electronic and spin-transport properties of altermagnets have attracted significant attention, but their thermodynamic signatures, particularly the specific heat, remain comparatively less explored. Specific heat would

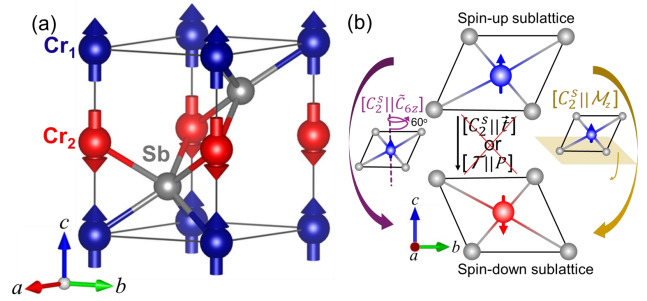


Figure 1. (a) Schematic diagram of crystal and magnetic structures drawn by VESTA [29]. Opposite-spin sublattices are denoted as Cr₁ (blue: spin-up) and Cr₂ (red: spin-down), with Sb atoms shown as grey spheres. (b) Schematic view of symmetry-driven sublattice interchange in CrSb. The two magnetic sublattices are related through either a screw operation accompanied by a half unit cell translation along the c direction or a mirror operation with respect to a plane perpendicular to the c axis.

Table I. Comparison of magnon characteristics in ferromagnet (FM), antiferromagnet (AFM) and altermagnet (AM).

Parameter	Isotropic FM	Anisotropic FM	Isotropic AFM	Anisotropic AFM	Weak-SOC AM	Moderate/strong-SOC AM
Low k dispersion	quadratic ($\omega \propto \Delta + D'k^2$) $\Delta = 0$	quadratic ($\omega \propto \Delta + D'k^2$) $\Delta \neq 0$	linear ($\omega \propto vk$) $\omega_{(\uparrow)}(k) = \omega_{(\downarrow)}(k)$	mixed ($\omega \propto \sqrt{\Delta + v^2k^2}$) $\Delta \neq 0$	linear $\omega_{(\uparrow)}(k) \neq \omega_{(\downarrow)}(k)$ $\omega(k \rightarrow 0) \rightarrow 0$	mixed $\omega_{(\uparrow)}(k) \neq \omega_{(\downarrow)}(k)$ $\omega(k \rightarrow 0) = \Delta_{\uparrow,\downarrow}$
Spin-wave branch	non-degenerate	non-degenerate	degenerate	non-degenerate, split	non-degenerate	non-degenerate, split
spin polarization	Yes	Yes	No	Yes, k -dependent	Yes, k -dependent	Yes, k -dependent
C_{mag}	$\propto T^{3/2}$	$\propto T^{\frac{1}{2}} \exp(-\frac{\Delta}{T})$	$\propto T^3$	$\propto T^{\frac{1}{2}} \exp(-\frac{\Delta}{T})$	power law	$\propto T^{\frac{1}{2}} \exp(-\frac{\Delta}{T})$
Magnon gap (Δ)	gapless	gapped	gapless	gapped	gapless	gapped

provide a direct thermodynamic measure of low-energy electronic and magnetic excitations of the altermagnetic state. In conventional AFMs with linear spin-wave dispersion, the low-temperature specific heat is typically governed by gapless magnons with a characteristic power law dependence ($C_{\text{mag}} \propto T^d$, where $d = 1, 2, 3$ for a 1-D (linear), 2-D (layer), and 3-D (spatial) antiferromagnet, respectively) [30–32], whereas isotropic ferromagnets (FMs) with quadratic spin wave dispersion, the magnetic contribution typically follows a $T^{3/2}$ dependence [32, 33]. In altermagnets, symmetry induces anisotropic and spin-split altermagnon modes without net magnetization [22–24, 32, 34]. A comparative overview of magnon dispersions and the anticipated magnetic specific-heat behaviour in FMs, AFMs, and AMs systems is summarised in Table I. Recent neutron diffraction studies on CrSb demonstrate that altermagnetic order lifts altermagnon spin degeneracy and opens finite gaps in the magnon spectrum [22–24]. Thus, specific heat measurements provide a powerful probe of symmetry-driven magnetic excitations in altermagnets. It has been reported that the specific heat of CrSb from room temperature to 900 K exhibits a pronounced rise near 685 K, exhibiting an antiferromagnetic transition [12]. However, a detailed study of the low-temperature specific heat in CrSb single crystals is still lacking. Moreover, there are no detailed reports on the growth of high-quality CrSb single crystals. Most existing reports use Sn flux and yield needle-like crystals with a cross-section of about 0.3 mm [14–16].

In this work, we report our new approach to the growth of high-quality CrSb single crystals using the self-flux method and investigate their low-temperature thermodynamic and transport properties. We revealed that our Sn free growth technique significantly enhances both the crystal size and quality. Our large crystals enable us to investigate thermodynamic properties using one crystal, allowing more accurate measurements free from complications arising from the use of multiple crystals. Comprehensive measurements of specific heat uncover the magnetic contributions over a wide temperature range, which play a crucial role in the temperature dependence of spe-

cific heat. This provides thermodynamic support for the existence of the gapped magnons from the altermagnetic order. This result shows that CrSb gives rise to robust gapped magnon modes that remain stable far above room temperature, making it a promising candidate for room temperature magnonic and spin-transport applications. In addition, it is important to examine the existence of superconductivity in CrSb, since non-stoichiometric polycrystalline $\text{CrSb}_{1+\delta}$ is reported to show superconductivity below $T_c = 8.8$ K [35]. We report that superconductivity in our CrSb crystal is absent down to 0.1 K.

SINGLE CRYSTAL GROWTH

Growth technique

We employed the self-flux method using high-purity Cr chunks (Thermo Fisher Scientific, 99.995%) and Sb shots (Thermo Fisher Scientific, 99.999%) as starting materials. The materials were loaded into an alumina crucible of dimensions 25 mm (length) $\times$ 10 mm (inner diameter) that is inert to the melt. We put the material with a higher melting temperature at the bottom. As the lower-melting-point material (Sb) liquefy, they flow over the higher-melting-point one (Cr), gradually dissolving them into the melt. A plug of quartz wool was used as a cap to serve as a filter during the flux separation process. The crucible was placed inside a quartz ampoule, slightly raised above the ampoule base using small pieces of quartz wool to prevent cracking due to thermal expansion mismatch between the crucible and the quartz tube. Additional quartz wool was placed above the crucible as shown in Fig. 2. This arrangement helps protect the quartz tube from damage during the high-speed centrifugation step. After assembling the setup, the quartz tube was purged several times with argon and then sealed under high vacuum (10^{-5} mbar). Finally, the sealed tube was placed vertically inside a box furnace for thermal treatment.

Table II. Conditions of the growth of CrSb crystals. Temperature is monitored by the system's thermocouple.

Run number	Cr:Sb ratio	Growth temp. ($^{\circ}\text{C}$)	Centrifuge temp ($^{\circ}\text{C}$)	Cooling rate ($^{\circ}/\text{hrs}$)	Remarks	Typical crystal size (mm^3)
1	30:70	1110	670	2	CrSb ₂ crystals	$1 \times 1 \times 0.5$
2	45:55	1000	900	2	Tiny CrSb crystals	$0.1 \times 0.1 \times 0.1$
3	45:55	1000	670	2	Large hexagonal-shape crystals and lamellar crystals	$1.5 \times 2 \times 2.5 \times 1$
4	45:55	1000	670	2	Large hexagonal-shape crystals and lamellar crystals	$1 \times 2.5 \times 2 \times 1$
5	45:55	1000	600	2	Flux-colated crystals	$3 \times 2.5 \times 1.5$
6	45:55	1110	670	1.5	Tiny CrSb crystals	$0.1 \times 0.5 \times 0.1 \times 0.2$

Growth conditions

We observed a systematic variation in crystal formation as a function of growth temperature and stoichiometric ratio of the starting materials. The optimal molar ratio of Cr to Sb was determined to be 45:55. The sealed tube was placed inside a furnace, where the temperature was raised to 1110°C at a rate of $200^{\circ}\text{C}/\text{h}$ and held for 36 hours. After this, the sample was slowly cooled at a rate of $2^{\circ}\text{C}/\text{h}$. At 900°C , the tube was quickly taken out from the furnace, and inverted by 180° into a centrifuge cup, and rapidly spun. This process drives the molten flux through the quartz wool, effectively separating it from the crystals, which remain in the crucible. The schematic of the growth process is shown in Fig. 2. We observed very tiny crystals inside the crucible. In the subsequent trial, we reduced the centrifugation temperature to 670°C and quenched the tube in ice water after centrifugation. Rapid quenching makes

the flux more brittle and easier to separate the residual flux mechanically from the crystals. This resulted in the formation of well-defined hexagonal crystals, typically around $2 \times 2.5 \times 1 \text{ mm}^3$ in size. We also performed centrifugation at 600°C ; although crystals were obtained, their separation from the residual flux was more challenging. This is likely due to the solidification of Sb, which occurs at 630°C . Below this temperature, solidified Sb tends to coat the crystal surfaces, hindering clean separation. Therefore, centrifuging above 670°C is essential; otherwise, the rapid temperature drop from furnace to centrifuge can adversely affect the flux removal process. The growth conditions and their outcomes are summarized in Table II.

RESULTS AND DISCUSSION

Crystal characterization

Inset of Fig. 3a shows an optical image of a flat, hexagonal-shaped CrSb crystal with typical dimensions of approximately $2 \times 2.5 \times 1 \text{ mm}^3$. The crystallographic orientation was determined using Laue backscattering photos recorded using a RIGAKU RASCO-BLII system, as shown in Fig. 3a, where the diffraction pattern indicates that the broad facet corresponds to the (001) plane.

Elemental analysis was conducted on both the as-grown surface and a freshly cleaved surface using an electron microscope equipped with tungsten filaments (WSEM, JSM-6010LA; JEOL) to determine the precise stoichiometry of Cr and Sb. As presented in Fig. 3b, the analysis for both surfaces shows a Cr:Sb atomic ratio of 1:1. The phase purity and crystal structure were further examined through powder X-ray diffraction (XRD) on crushed crystals, using a desktop XRD spectrometer (RIGAKU MiniFlex 600-C) with $\text{Cu-K}\alpha$ radiation. The diffraction pattern collected down to $2\theta = 3^{\circ}$ reveals no impurity phases (Fig. 3c). We carried out Rietveld refinement of the diffraction profile using the FullProf suite [29] to obtain the lattice parameters $a = 4.12 \text{ \AA}$ and $c = 5.47 \text{ \AA}$, consistent with previously reported values of $a = 4.10\text{--}4.12 \text{ \AA}$ and $c = 5.45\text{--}5.47 \text{ \AA}$ [12–15, 35].

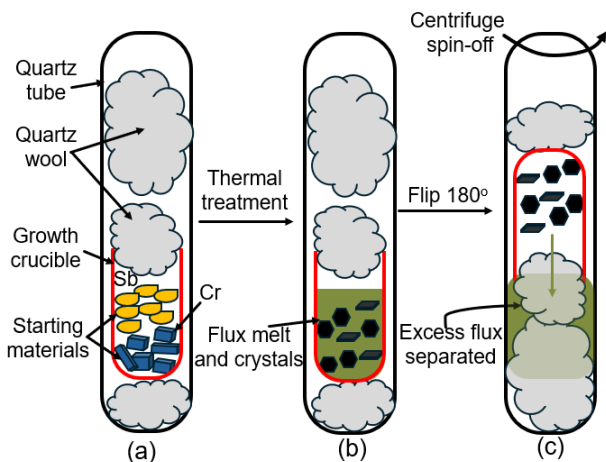


Figure 2. Schematic description of the CrSb single crystal growth process based on the self-flux method. (a) Raw materials (Cr and Sb) are placed in a crucible inside a quartz ampoule for thermal treatment. (b) Upon heating, the flux melts, and CrSb crystals form. (c) The quartz ampoule is inverted by 180° and centrifuged to separate the residual flux from the CrSb crystals.

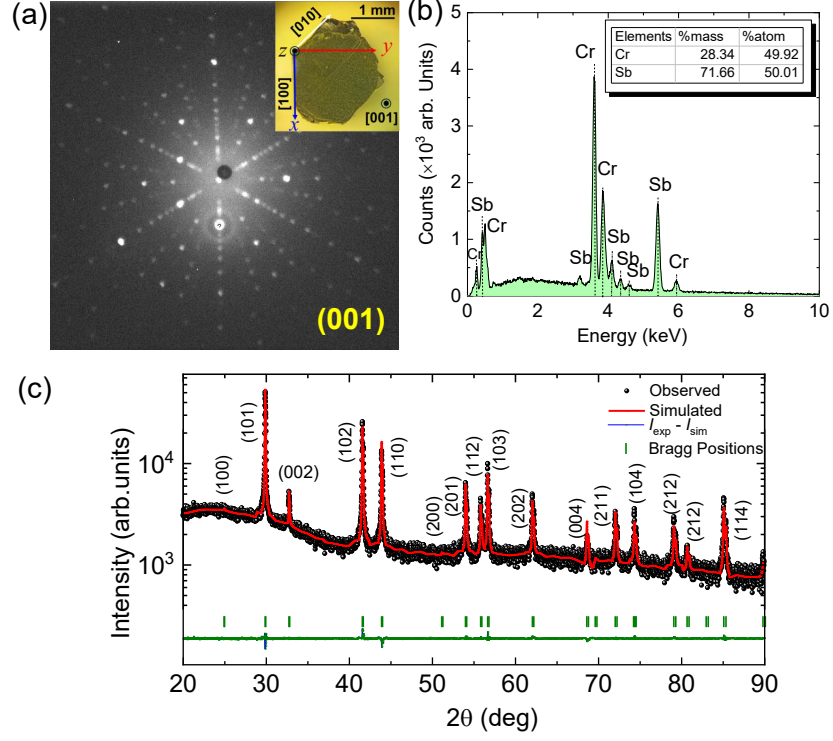


Figure 3. (a) Laue diffraction pattern of a CrSb crystal, showing a diffraction pattern characteristic of the (001) plane. Inset: optical image of the CrSb single crystal. (b) Result of energy dispersive spectroscopy (EDS) of the same crystal indicating a stoichiometric CrSb composition. (c) Powder XRD pattern of crushed CrSb single crystals (black points), shown together with the simulated profile (red curve). The difference between the experimental and simulated intensities is also shown with the blue curve. The green vertical lines represent the calculated Bragg reflection positions. The near coincidence of the two green bars arises from the K_{α_1} and K_{α_2} characteristic X-ray emission lines.

Resistivity

Figure 4a shows the temperature-dependent longitudinal resistivity measured from 1.8 K to 300 K using the AC four-probe technique [5, 36]. Resistivity measurements were performed using a custom-made transport probe mounted on a Quantum Design MPMS-XL system. We defined the x axis as along one of the a axis, the y perpendicular to x within the ab -plane, and the z axis as along the c -axis, as illustrated in the inset of Fig. 3a. The electric current was applied along the x axis. The resistivity shows a monotonic increase with temperature, indicating metallic conduction. In zero magnetic field, the longitudinal resistivity $\rho_{xx}(T)$ decreases from $135 \mu\Omega \text{ cm}$ at 300 K to $12 \mu\Omega \text{ cm}$ at 4 K, corresponding to a residual resistivity ratio (RRR: $\rho_{300 \text{ K}}/\rho_{4 \text{ K}}$) of about 11. This value is higher than those reported in recent studies [14–16, 18, 19, 37], indicating high crystal quality of our samples. At high temperatures, electron–phonon scattering predominates, resulting in a linear dependence on T , while at low temperatures, electron–electron interactions take over, leading to a T^2 dependence. The blue and red dashed curves in Fig. 4a represent fits to the

data using $\rho = \rho_0 + BT$, and $\rho = \rho_0 + AT^2$, respectively. The overall temperature dependence in the entire range is well described by the empirical relation $\rho = \rho_0 + AT^{1.7}$.

Figure 4b shows the magnetic field-dependent magnetoresistance ($\text{MR} = \frac{\rho_{xx}(H) - \rho_{xx}(\mu_0 H=0)}{\rho_{xx}(\mu_0 H=0)}$) in the temperature range 3.5–300 K. A pronounced positive MR of nearly 80% at 6 T and 3.5 K was observed, higher than values commonly reported for antiferromagnetic metals [17, 39, 40] and those in recent studies on CrSb single crystals [14–17, 38], indicating the superior quality of

Table III. Variation of crystal quality and MR at low temperature (below 10 K) for single crystals and thin films of CrSb.

RRR	MR (%) at 6 T	Thickness/size	Ref.
1.7	0.1	Thin film (25 nm)	[19]
4.3	1.25	Thin film (30 nm)	[18]
5	15	$1 \times 1 \times 0.3 \text{ mm}^3$	[15]
7.3	16	$3 \times 0.5 \times 0.1 \text{ mm}^3$	[38]
8.9	45	$2 \times 2 \times 0.05 \text{ mm}^3$	[17]
11	80	$2 \times 2.5 \times 1 \text{ mm}^3$	This work

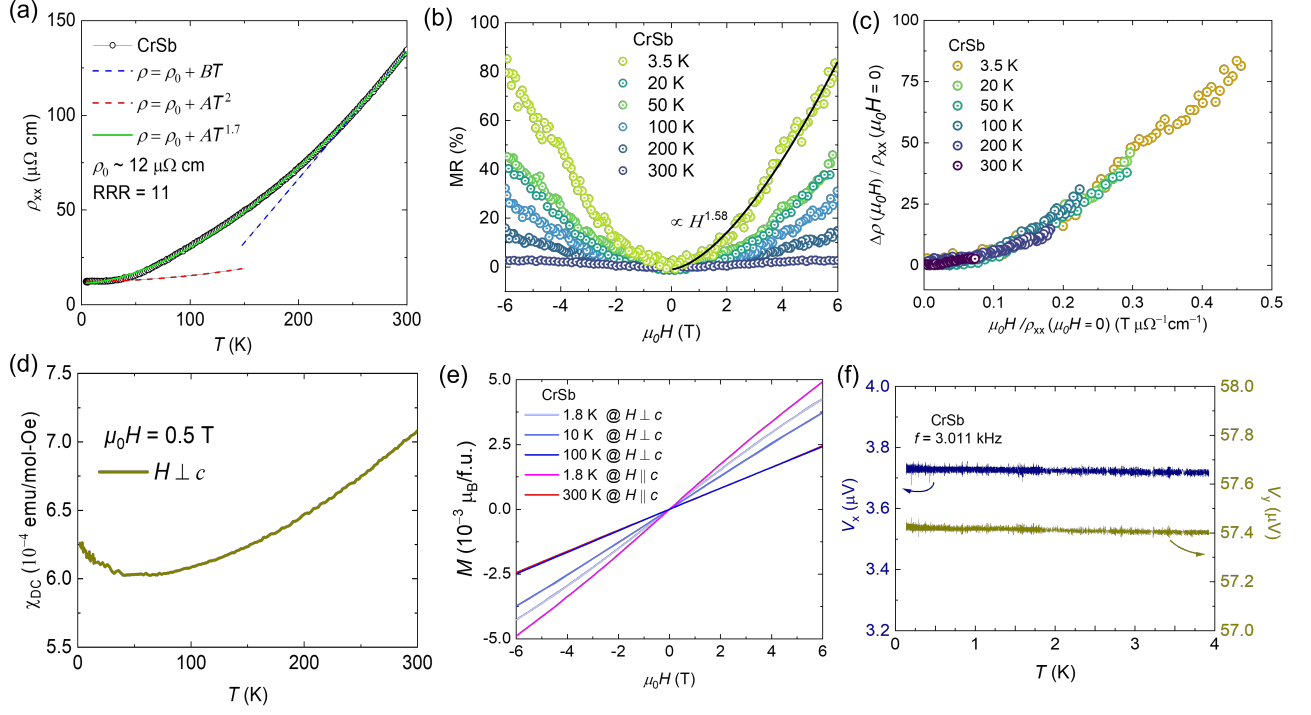


Figure 4. (a) Temperature dependence of the zero-field longitudinal resistivity. (b) MR at various temperatures ranging in 3.5–300 K for $H \parallel c$ and $I \parallel x$. (c) Kohler's plots of the MR at different temperatures. (d) Temperature dependence of the DC magnetic susceptibility (1.8–300 K) for $H \perp c$. (e) Field-dependent magnetization measured at various temperatures for $H \parallel c$ and $H \perp c$. (f) Susceptometer signal, which is proportional to the AC susceptibility, for a single crystal of CrSb. This measurement was performed using an AC excitation current of 0.50 mA rms ($\sim 17 \mu\text{T}$ rms) at a frequency of 3.011 kHz.

the present crystal. We compare our MR results with recent reports on CrSb single crystals [15, 17, 38] and thin films [18, 19] (see Table III). Clearly, higher-RRR samples exhibit larger MR. This supports the attribution of higher MR in our samples to the improved crystal quality. As the temperature increases, the MR gradually decreases in magnitude yet remains positive in the entire field range. As shown by solid curve in Fig. 4b, we found that MR varies with $B^{1.58}$, consistent with previous reports [14, 15]. According to classic Kohler's rule, magnetoresistance measured at different temperatures is expected to merge onto a universal curve when expressed as a function of the scaled field ($\mu_0 H/\rho_{xx}(\mu_0 H = 0)$). As shown in Fig. 4c, our MR data collapse onto a single curve, demonstrating strong adherence to Kohler's rule. This behavior suggests that the magnetoresistance is primarily governed by the orbital motion of charge carriers with a common scattering time in a magnetic field [14–16].

Magnetization

The temperature-dependent DC susceptibility (M/H) of a CrSb single crystal was measured with the magnetic

field applied perpendicular to the crystallographic c -axis ($H \perp c$), as presented in Fig. 4d. Magnetic properties were measured with a SQUID magnetometer (MPMS-XL, Quantum Design). Néel temperature is expected to be well above room temperature, beyond the measured range of 1.8–300 K. Magnetization was found to decrease gradually upon cooling, consistent with the behavior expected for an antiferromagnetic material.

Figure 4e presents the magnetization as a function of magnetic field at 1.8–300 K along $H \perp c$ and $H \parallel c$. The magnetization increases linearly with the applied magnetic field and shows no signs of magnetic hysteresis, spin-flop, or metamagnetic transitions. Even at 6 T, magnetization remains relatively small, approximately $10^{-3} \mu_B$ per Cr atom, notably lower than earlier neutron diffraction studies [12, 41]. Moreover, the magnetization remains far from saturation even at 6 T, consistent with the typical behavior of antiferromagnetic materials. The $M-H$ curve at 1.8 K is showing a weak non-linear trend, suggesting that the spins in the antiferromagnetic state are strongly coupled, allowing only a slight canting in response to the applied magnetic field [14].

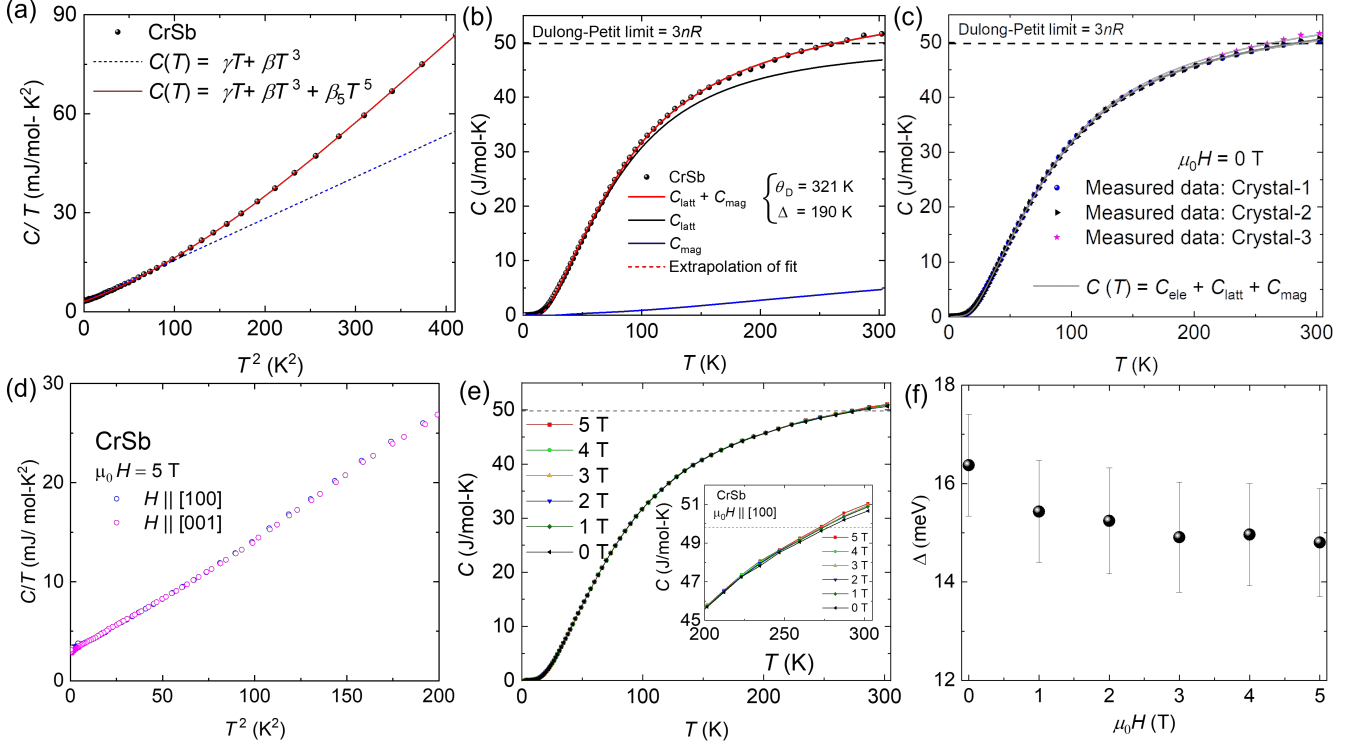


Figure 5. (a) C/T plotted against T^2 from 0.45 to 20 K. Data is fitted with Eq. (2). We also show the result of fitting with $C = \gamma T + \beta T^3$ below 8 K (blue broken curve). (b) Temperature-dependent specific heat of a CrSb crystal measured between 1.8 K and 300 K under zero magnetic field. The black dashed line corresponds to the Dulong Petit limit. The red solid line represents the fit obtained over the temperature range 25–300 K using Eq. (4). The black and blue solid curves indicate the individually extracted lattice and magnon contributions, respectively. (c) Specific heat results from several samples. The solid curves are the result of fitting with Eq. (4). The specific heat of RuO₂, expressed as (C/T) , is plotted as a function of T^2 for temperatures between 0.5 and 10 K under a 5 T magnetic field applied along the [001] and [100] crystallographic directions. The data for the different orientations overlap within experimental accuracy, indicating isotropic behaviour. (e) Field and temperature dependence of the specific heat of CrSb. The inset shows the zoom-in view of the $C(T, H)$. (f) Variation of Δ with field.

AC susceptibility down to 0.1 K

In order to examine the superconductivity in CrSb, the AC susceptibility of the sample was measured using the mutual inductance technique with a lock-in amplifier, integrated into a Quantum Design Physical Property Measurement System (PPMS) [42]. The susceptometer is compatible with both the conventional PPMS sample chamber and the adiabatic demagnetization refrigerator (ADR) module. The ADR, employing a paramagnetic salt (chromium alum), allows rapid cooling from room temperature to below 0.1 K within two hours. The susceptometer coil is made of an outer primary coil and inner secondary coils, detailed configuration outlined in [42]. The output signal from the susceptometer was detected using a lock-in amplifier (SR830, Stanford Research Systems), which enables measurement of both the in-phase (V_x) and out-of-phase (V_y) components of the AC voltage at a frequency of 3.011 kHz. Figure 4f shows susceptome-

ter response in the range 0.1 K to 4.0 K measured using the ADR option. Our results show that there is no superconductivity in a stoichiometric CrSb single crystals down to 0.1 K.

Specific heat capacity

Figure 5a presents the temperature-dependent specific heat of a CrSb single crystal measured under zero magnetic field, with data collected in logarithmic temperature intervals from 0.45 to 20 K. The specific heat measurements were carried out using a commercial calorimetry setup (Quantum Design PPMS) equipped with a ³He refrigeration option. We do not observe any phase transition down to 0.45 K. The observed specific heat primarily consists of two contributions and is given by

$$C(T) = C_{\text{ele}}(T) + C_{\text{latt}}(T). \quad (1)$$

The first term corresponds to the electronic specific heat associated with conduction electrons (C_{ele}), expressed as γT , where the Sommerfeld coefficient $\gamma = \pi^2 N_A k_B^2 D(\epsilon_F)/3$, with k_B as Boltzmann's constant and $D(\epsilon_F)$ the density of states (DOS) at the Fermi energy for both spin directions. The second term represents lattice-specific heat arising from phonons (C_{latt}). We extracted the electronic contribution to the specific heat (γ) by plotting C/T against T^2 in the 0.45–20 K range, as shown in Fig. 5a. The low-temperature data were fitted using a standard model with linear electronic and cubic lattice terms (blue broken curve), but deviated possibly due to higher-order phonon effects above ~ 8 K. As shown by the red curve, the low-temperature specific heat is well fit up to 20 K by:

$$C(T) = \gamma T + \beta T^3 + \beta_5 T^5, \quad (2)$$

where β is associated to the Debye temperature θ_D as [43]

$$\beta = \frac{12\pi^4 n k_B N_A}{5\theta_D^3}, \quad (3)$$

where $n = 2$ corresponds to the total number of atoms in a formula unit (f.u.) for CrSb. Fitting the zero-field data using Eq. (2) gives $\gamma = 4.0 \pm 0.08 \text{ mJ mol}^{-1} \text{ K}^{-2}$ and a phonon coefficient $\beta = 0.12 \pm 0.02 \text{ mJ mol}^{-1} \text{ K}^{-4}$, corresponding to a Debye temperature θ_D is 318 K. We compare the obtained γ with the electronic density of states (DOS) at the Fermi level, $D(\epsilon_F)$. Using γ , we estimate $D(\epsilon_F) = 1.7 \text{ states/eV/f.u.} = 23.1 \text{ states/Ry/f.u.}$, consistent with DOS of about 1.6–1.7 states/eV/f.u. = 21.8 states/Ry/f.u. obtained from band-structure calculations without spin-orbit coupling [44]. We expect that spin-orbit interaction plays a minor role in DOS since it has been reported that band-structure calculations with and without SOC exhibit subtle differences in band structure near the Fermi level [25, 45].

Now, we examine the high-temperature specific heat data (25 K to 300 K), presented in Fig. 5b. We found that the specific heat exceeds the Dulong–Petit limit ($C_V = 3nR = 49.8 \text{ J mol}^{-1} \text{ K}^{-1}$, where R is the gas constant) near room temperature. This behaviour cannot be explained by only lattice and electronic heat capacity contributions. To explain the specific heat exceeding the Dulong–Petit limit, it is necessary to consider the magnetic contribution. Thus, the total specific heat can be expressed as:

$$C(T) = \gamma T + C_{V(\text{Debye})}(T) + C_{\text{mag}}(T), \quad (4)$$

where $C_{V(\text{Debye})}(T)$ is the specific heat at constant volume formulated as

$$C_{V(\text{Debye})}(T) = 9nR \left(\frac{T}{\theta_D} \right)^3 \int_0^{\theta_D/T} \frac{x^4}{(e^x - 1)^2} dx. \quad (5)$$

Table IV. Extracted parameters from the specific heat data of CrSb using the combination of lattice, electronic, and magnetic contributions. The value of θ_D in this table was evaluated from high-temperature fitting using Eq. (4).

Parameters	Crystal-1	Crystal-2	Crystal-3
mass (mg)	23.2	24.9	28.8
γ (mJ mol ⁻¹ K ⁻²)	4.0	4.02	3.12
β (mJ mol ⁻¹ K ⁻⁴)	0.12	0.11	0.10
β_5 (mJ mol ⁻¹ K ⁻⁶)	2.48×10^{-4}	2.58×10^{-4}	2.92×10^{-4}
θ_D (K)	321	320	323
$D(\epsilon_F)$ (States/Ry/f.u.)	23.1	23.2	18.0
Δ (K)	190	190	180
a_1 (J mol ⁻¹ K ^{-3/2})	0.31	0.40	0.32

For $C_{\text{mag}}(T)$, the magnetic specific heat due to magnetic excitation, we attempted to use various models listed in Table I and found that the gapless models cannot fit the experimental data. Thus, we used the gapped magnon model, namely

$$C_{\text{mag}}(T) = a_1 T^{\frac{1}{2}} \exp\left(-\frac{\Delta}{T}\right), \quad (6)$$

which accounts for the specific heat of magnons with a moderate energy gap Δ . The red solid line in Fig. 5b shows the best fit over 25–300 K using Eq. (4), while the black and blue solid curves represent the extracted lattice and magnon contributions, respectively. At room temperature, the zero-field C_{mag} of CrSb is about 3.2 J/mol-K, consistent with earlier report [12]. Within the considered temperature range, the best fit yields a Debye temperature of about 321 ± 5 K, in good agreement with the low-temperature estimate and the magnon energy gap of $\Delta = 190 \pm 10$ K ($\sim 16 \pm 1$ meV), consistent with inelastic neutron-scattering measurements [22, 46]. The corresponding fitted parameters for all samples are summarized in Table IV. All samples exhibit similar values of θ_D and Δ , showing the high reproducibility of our investigation. Among the gapped models, since the expected specific heat behaviour ($C_{\text{mag}}(T) \propto T^{\frac{1}{2}} \exp(-\frac{\Delta}{T})$) is identical for all of anisotropic FM, AFM, and AM, it is difficult to distinguish the types of magnetic order only from the temperature dependence of specific heat. Nevertheless, the deduced gap size is in good agreement with previous experimental and theoretical studies on CrSb. This fact provides strong thermodynamic support for the altermagnetic order in CrSb.

Figure 5d presents the specific-heat response measured under a 5 T magnetic field for different field orientations. To probe the role of spin fluctuations, the field was applied parallel to the [001] and [100] crystallographic axis. Notably, at 5 T, the measurements show no detectable anisotropy among these directions. The Zeeman energy at 5 T (~ 0.6 meV $\equiv 7$ K) is much smaller than the obtained magnon gap (16 meV). Therefore, a 5 T field does not change the magnon gap or dispersion enough

to produce an observable difference in specific heat along different directions. To understand the variation of Δ with field, we measured the $C(T)$ under magnetic fields up to $\mu_0 H = 5$ T as shown in Fig. 5e. The specific heat shows a small variation above 100 K (inset of Fig. 5e). The field- and temperature-dependent $C(T, H)$ is fitted using Eq. (4), with θ_D fixed to the zero-field value, as the phonon contribution is expected to be field-independent. We observe a small reduction of Δ with increasing field as shown in Fig. 5f, whereas the coefficient a_1 remains nearly constant. This reduction in Δ is likely due to Zeeman splitting of the lowest-energy magnon mode rather than magnetic anisotropy [22, 23, 32, 47]. Nevertheless, due to relatively large uncertainty in Δ , precise discussion on the origin of Δ change is difficult and urges for in-field spectroscopies.

The magnetic ordering in CrSb is strong, so magnetic excitations are likely to make a significant contribution to the specific heat over a wide temperature range, not just near the Néel temperature. In collinear antiferromagnets, the absence of magnetic anisotropy preserves continuous spin-rotation symmetry, leading to gapless Goldstone magnon modes, while spin-orbit induced anisotropy opens a finite but degenerate magnon gap [32, 48, 49]. In this regard, two prototypical collinear AFMs, NiO and MnO, preserve combined $\mathcal{PT}$ symmetry. In the absence of magnetic anisotropy, these systems host degenerate magnon branches. However, the inclusion of weak spin-orbit-induced magnetocrystalline anisotropy lifts this degeneracy near the high symmetry Γ point, resulting in a small magnon gap of less than 5 meV [49]. Altermagnets fundamentally differ from this paradigm [22, 23, 32]. In CrSb, the crystal symmetry enforces alternating exchange interactions that lift magnon spin degeneracy even in the collinear and fully compensated state [22, 23]. Within linear spin-wave theory (LSWT), an effective Heisenberg Hamiltonian with symmetry-inequivalent exchange couplings and single-ion anisotropy supports two spin-split magnon modes. In the long-wavelength limit, the magnon gap [50] is $\Delta_{\text{LSWT}} \approx 2S\sqrt{D J_{\text{eff}}}$, where S is the local moment of the Cr ions, D is the spin-orbit-induced single-ion anisotropy, and $J_{\text{eff}} = \sum_n z_n J_n$ denotes the effective exchange scale determined by the symmetry-inequivalent Cr-Cr exchange interactions. Using reported parameters for CrSb ($S=3/2$, $J_1=23$ meV, $J_2=-5.4$ meV, $J_3=5.2$ meV, and $D \sim 0.15$ meV) [22], the resulting altermagnon gap is estimated to be $\Delta_{\text{LSWT}} \sim 9$ -10 meV [22, 23], consistent with our specific heat fitting. These suggest that CrSb is a prototypical system where gapped magnons coexist with intrinsic spin-splitting driven by nonrelativistic exchange symmetry. Based on theoretically predicted non-degenerate magnon dispersion and the experimentally observed large magnon gap supports the realization of the altermagnetic order due to exchange interactions.

CONCLUSION

In conclusion, we described details of a high-quality CrSb crystal growth with the self-flux method. We obtained large hexagonal facet (001) crystal, up to $2 \times 2.5 \times 1$ mm³ with RRR exceeding 10. A large positive magnetoresistance in CrSb of about 80% at 3.5 K and 6 T is observed due to improved sample quality. Magnetization shows a linear field dependence, consistent with the antiferromagnetic order in CrSb. The low-temperature specific heat deviates from the conventional $C(T) = \gamma T + \beta T^3$ behavior, which can be well accounted for by higher-order phonon contributions $\beta_5 T^5$. The extracted Sommerfeld coefficient, $\gamma = 4.0 \pm 0.08$ mJ mol⁻¹ K⁻², suggests modest electronic correlations among conduction electrons. At high temperatures, the specific heat surpasses the Dulong–Petit value, which can be attributed to a broad magnon contribution associated with the altermagnetic state. Analysis of the data yields a Debye temperature of 321 ± 5 K and a magnon excitation gap of approximately 16 ± 1 meV. Furthermore, no signature of superconductivity is observed in stoichiometric CrSb down to 0.1 K. Our thermodynamic and transport results advance the understanding of CrSb and establish a robust bases for engineering its altermagnetic order.

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AUTHOR CONTRIBUTIONS

Shubhankar Paul: Conceptualization (lead), Methodology (lead), Data curation (lead), Formal analysis (lead), Visualization (equal), Writing – original draft (lead), Writing – review and editing (equal). **Atsutoshi Ikeda:** Data curation (equal), Writing – review and editing (supporting). **Giordano Mattoni:** Visualization

(equal), Data curation (supporting), Writing – review and editing (equal). **Shingo Yonezawa:** Conceptualization (equal), Methodology (equal), Visualization (equal), Project administration (equal), Fund acquisition (equal), Writing, review, and editing (equal). **Chanchal Sow:** Conceptualization (equal), Methodology (equal), Visualization (equal), Fund acquisition (equal), Project administration (equal), Writing – review and editing (equal).

ADDITIONAL INFORMATION

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DATA AVAILABILITY

The data is available on request to the corresponding author.

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