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Article

Fe-Rich Ferromagnetic Cleavable Van der Waals Telluride Fe₅AsTe₂

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= 48 K. In the ordered state, the magnetization shows a dual-slope behavior in low magnetic fields, indicating the compensated or canted nature of magnetism. Magnetic susceptibility and magnetization measurements reveal perpendicular magnetic anisotropy. The large Rhodes–Wohlfarth ratio of 4.6 indicates the itinerant nature of ferromagnetism in Fe_5AsTe_2 .

INTRODUCTION

Layered van der Waals (vdW) compounds, such as $\text{CrSiTe}_{3,1}^{1}$ CrGeTe₃,² CrI₃,³ and Fe₃GeTe₂,^{4–7} with embedded 3d transition metals are a platform for emergent two-dimensional (2D) nanomaterials with peculiar magnetic and electronic properties. While the synthesis of 2D nanomaterials is a challenging route, involving nontrivial methods, such as chemical vapor deposition, magnetic sputtering, molecular beam epitaxy, and others,⁸ mechanical exfoliation of bulk cleavable crystals is still the simplest strategy that provides high-quality functional nanomaterials. This method was successfully employed to obtain atomically thin CrSiTe₃,⁹ CrGeTe₃,¹⁰ Ta₃FeS₆,¹¹ Fe₃GeTe₂,¹² and Fe₅GeTe₂¹³ with the persistence of ferromagnetism in two dimensions.

Remarkably, Fe-rich layered vdW tellurides contain a number of ferromagnets with high Curie temperatures and metallic-type conductivity, which are able to produce the spinpolarized current for spin filtering in tunnel junctions and heterostructures. Here, Fe₃AsTe₂ with bulk $T_{\rm C} = 123$ K,¹⁴ Fe₃GeTe₂ (225 K),⁵ Fe₅Ge₂Te₂ (250 K),¹⁵ and Fe₅GeTe₂ (310 K)^{13,16-19} are suitable for designing novel spintronic devices with tailored functionalities. For example, 2D Fe₃GeTe₂/*h*-BN/Fe₃GeTe₂ vdW heterostructures demonstrate the spin polarization of 0.66 at 4 K, corresponding to 83 and 17% of majority and minority carriers, respectively.¹² Furthermore, topological vortex phases,²⁰ skyrmion bubbles,²¹ and Neel-type skyrmions²² were observed in Fe₃GeTe₂ bulk crystals and 2D nanomaterials. The Curie temperature of Fe_3GeTe_2 is significantly reduced in the atomically thin 2D nanomaterial.²³ However, it can be enhanced up to room temperature by gate tuning,²⁴ Ga doping,²⁵ or patterning methods,²⁰ which is promising for the application of spintronic devices under ambient conditions. Also, the related Fe_5GeTe_2 telluride exhibits ferromagnetism in the vicinity of room temperature both as a bulk material and in two dimensions.¹³ Recently, the As for Ge substitution has been examined in the $Fe_5Ge_{1-y}As_yTe_2$ system, and the new vdW telluride, $Fe_{4.8}AsTe_2$, was reported.²⁶ The compound is the only isostructural analog of the room-temperature ferromagnet Fe_5GeTe_2 .

In this study, we report on the synthesis, crystal growth, crystal and real structure, and magnetic properties of the Fe_sAsTe_2 vdW telluride. Due to the layered crystal structure with only weak van der Waals bonding between the neighboring Fe-rich layers, bulk cleavable crystals can be synthesized and used for future design of vdW heterostructures and novel spintronic architectures.

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EXPERIMENTAL SECTION

Synthesis and Crystal Growth. Fe (powder, 99.98%, Alfa Aesar), As (powder, 99.997%, Merck), and Te (pieces, 99.999%, Merck) were used as starting materials. Handling of the As powder should be taken with a special care using protective gloves. In particular, all operations with As were performed in an argon-filled glove box [Spectro-systems, $p(H_2O, O_2 < 1 \text{ ppm})$]. The elements were weighed in the stoichiometric molar ratio of $\nu(Fe):\nu(As):\nu(Te)$ = 5:1:2 with a total mass of 0.5 g and placed inside a quartz ampule, which was sealed under a vacuum of 5×10^{-3} mbar. The ampule was annealed twice in a programmable furnace at 973 K for 7 days with intermediate grinding of the specimen in an agate mortar. Single crystals of Fe₅AsTe₂ were grown employing chemical vapor transport (CVT) reactions. The transport agent (iodine, 1 mg/mL) along with the presynthesized polycrystalline Fe₅AsTe₂ with a total mass of 0.5 g was vacuum-sealed inside a quartz ampule (8 mm in inner diameter and 200 mm in length), which was annealed in a horizontal two-zone programmable furnace. The transport temperatures were adjusted between 1073 and 1173 K on the source side and between 973 and 1073 K on the sink side. Single crystals in the form of platelets were collected in the cold zone after annealing in the temperature gradient of 1123-1073 K during 6 weeks. After synthesis, the furnace was cooled to room temperature in the shut-off regime.

Sample Characterization. Phase composition was studied using a Stoe STADI P powder X-ray diffractometer ($CoK\alpha_1$ radiation, Ge_{111} monochromator). The STOE WinXPOW program (version 1.06) was used for data collection and evaluation. Single crystals were examined on a JSM JEOL 6490-LV scanning electron microscope equipped with an energy dispersive X-ray detection system INCA x-Sight. Elemental composition was determined by electron probe microanalysis (EPMA) by collecting and averaging of 10–20-point spectra for each sample.

Structure Refinement. Single crystals grown by the CVT method were examined on an Enraf Nonius CAD-4 diffractometer (Mo X-ray source, graphite monochromator, $\lambda = 0.71073$ Å). The Ψ -scan procedure was employed for the absorption correction. The reported structural data of Fe₅GeTe₂¹³ were used as a starting model for crystal structure refinement, which was performed in the full-matrix anisotropic approximation against |F| using the Jana2006 program.²⁷ Polycrystalline Fe₅AsTe₂ was studied by high-resolution powder X-ray diffraction (HRPXRD) at the ID22 beamline [$\lambda = 0.354109(2)$ Å, $2\theta_{\text{max}} = 25^{\circ}$ of the European Synchrotron Radiation Facility (ESRF, Grenoble, France).²⁸ Measurements were conducted on the sample enclosed in a quartz capillary (0.4 mm in diameter), which was purged with pure argon gas (H_2O , $O_2 < 1$ ppm) and flame-sealed. The data were collected at room temperature and under heating from 373 to 973 K with the step of 100 K. Rietveld refinements against the highresolution powder X-ray diffraction data were performed using the Jana2006 program.²⁷ Details of data collection and crystal structure refinement are shown in Table 1. Parameters of atomic positions and selected interatomic distances are given in Tables 2 and 3, respectively.

Magnetic Properties. Magnetization measurements were performed on the oriented single crystals and polycrystalline Fe_5AsTe_2 using a Magnetic Properties Measurement System (MPMS-XL5, Quantum Design). The single-crystal sample was mounted rigidly on a commercial sample holder, when the magnetic field was applied parallel to the crystal's plane, and using a custom sample holder for the perpendicular orientation. The signal from the sample holder was measured independently for each orientation of the magnetic field. Demagnetization correction was applied assuming a flat cylindrical sample according to its highly anisotropic shape. Magnetic susceptibility was measured at temperatures between 2 and 300 K in magnetic fields of 0.1, 1, and 5 T in the zero-field cooling (ZFC) and field-cooling (FC) conditions. Magnetization measurements were performed in the ZFC conditions by a sweeping magnetic field between -5 and 5 T at various temperatures.

Table 1. Crystal Data Collection and Refinement Details for Fe₅AsTe₂

parameter	single crystal	polycrystalline
refined composition	Fe _{4.58(4)} AsTe ₂	Fe _{4.74(2)} AsTe ₂
space group	$R\overline{3}m$ (no. 166)	
T[K]	295	973
a [Å]	3.9937(5)	4.065788(9)
c [Å]	28.858(4)	29.7408(1)
$V [Å^3]$	398.61(9)	425.768(2)
Ζ	3	
$\rho_{\rm calc} ~[{\rm g}~{\rm cm}^{-3}]$	7.33	6.96
ranges in <i>hkl</i>	$-7 \leq h \leq 7$	$0 \leq h \leq 4$
	$-7 \le k \le 6$	$-4 \le k \le 0$
	$-24 \le l \le 52$	$0 \le l \le 36$
2θ range [°]	4.24-40.47	1-25
diffractometer	Enraf Nonius CAD-4	ID 22, ESRF
detector	point	nine-crystal multianalyzer
radiation, λ [Å]	ΜοΚα, 0.71073	synchrotron, 0.35411(2)
refined parameters	16	38
residual peaks [e ⁻ ·Å ⁻³]	2.29/-2.86	2.68/-2.93
$R_1 \left[I > 3\sigma(I) \right]$	0.0469	
$wR_2 [I > 3\sigma(I)]$	0.0590	
R_1 (all)	0.1333	
wR_2 (all)	0.0781	
$R_{\rm p}$ (obs)		0.0660
$R_{\rm wp}$ (obs)		0.0712
$R_{\rm p}$ (all)		0.0721
R_{wp} (all)		0.0713
GOF	1.03	2.36

Table 2. Parameters of Atomic Positions in the Crystal Structure of $Fe_5AsTe_2^{\ a}$

atom	site	x	у	z	occupancy	U_{eq} [Å ²]
Fe1	6c	0	0	0.0672(9)	0.29(2)	0.060(8)
Fe1	6c	0	0	0.0723(2)	0.370(3)	0.051(2)
Fe2	6c	0	0	0.3095(1)	1	0.0224(7)
Fe2	6c	0	0	0.31012(6)	1	0.0505(7)
Fe3	6c	0	0	0.3996(1)	1	0.029(1)
Fe3	6c	0	0	0.39921(6)	1	0.087(1)
As1	6c	0	0	0.0057(2)	0.5	0.0197(8)
As1	6c	0	0	0.0117(2)	0.5	0.047(3)
Te1	6c	0	0	0.21957(6)	1	0.0250(4)
Te1	6c	0	0	0.21961(3)	1	0.0482(4)

^{*a*}The data from the single-crystal and polycrystalline refinements are shown as regular and italic text, respectively.

Table 3. Selected Interatomic Distances for Fe₅AsTe₂ According to the Single-Crystal XRD Data

bond	distance	e [Å]
Fe1	-Fe2 (×3)	2.62(1)
	-Fe3 (×3)	2.3059(3)
	-As1 (×1)	2.10(3)
	-Te1 (×3)	2.67(1)
Fe2	-Fe2 (×3)	2.686(2)
	-Fe3 (×1)	2.601(5)
	-Fe3 (×3)	2.610(2)
	-As1 (×3)	2.458(2)
	-Te1 (×1)	2.594(4)
Fe3	-As1 (×3)	2.894(4)
	-Te1 (×3)	2.682(2)

RESULTS AND DISCUSSION

Crystal Structure. Crystal growth of Fe₅AsTe₂ using the CVT reactions yields bulk single crystals in the form of black hexagonal platelets with metallic luster. The obtained crystals possess the $Fe_{4.56(7)}As_{0.94(5)}Te_{2.06(3)}$ elemental composition according to EPMA, where a slight off-stoichiometry of Fe can be noticed. Indexing of the single-crystal X-ray diffraction data indicates a trigonal R-centered unit cell with a =3.9937(5) Å and c = 28.858(4) Å. These unit cell parameters are consistent with those of the secondary Fe₅AsTe₂ phase reported recently.²⁶ The $R\overline{3}m$ space group was chosen based on the diffraction pattern symmetry and systematic extinction conditions. Notably, Fe₅GeTe₂ has a similar unit cell.¹³ Therefore, the reported structural parameters of Fe₅GeTe₂ were used as a starting model for the refinement of the crystal structure of $\text{Fe}_{s}\text{As}\text{Te}_{2}$. It should be noted that the centrosymmetric $R\overline{3}m^{13,19}$ and noncentrosymmetric $R3m^{18}$ space groups were proposed for Fe₅GeTe₂. Analysis of the Fe₅AsTe₂ single-crystal X-ray diffraction pattern reveals that its symmetry is consistent with the centrosymmetric R3m space group.

The unit cell of Fe_5AsTe_2 , which is shown in Figure 1a, contains five crystallographic positions (Table 2). Remarkably,



Figure 1. Crystal structure of Fe_5AsTe_2 : (a) unit cell and coordination polyhedra, (b) structural block viewed along the *c* axis, and (c) edge-sharing connection of polyhedra. As atoms are shown in green, Te in black, and Fe1, Fe2, and Fe3 in red, blue, and yellow, respectively.

the As1 atoms slightly shift along the [001] direction from their ideal 3*a* position (0; 0; 0). As a result, they are located on the split 6*c* site with the fixed occupancy of 0.50. Similar to As1, the Fe1 position shows atomic disorder, too. Its thermal ellipsoid is elongated along the [001] direction, yielding the large value of U_{eq} at room temperature. For Fe1, $U_{11} = U_{22} =$ 0.023(5) Å², $U_{33} = 0.14(2)$ Å², $U_{12} = 0.011(3)$ Å², and $U_{13} =$ $U_{23} = 0$. The refinement of the crystal structure against the single-crystal data yields an average model, in which the partially occupied and split sites indicate inherent structural disorder. The local structure of Fe₅AsTe₂ presumably contains vacancies, dislocations, stacking faults, and other types of defects, which require a more careful study by means of highresolution transmission electron microscopy or other localprobe methods.

The crystal structure has a pronounced layered character. Here, hexagonal blocks shown in Figure 1b are confined by Te atoms and alternate along the [001] direction. Only weak van der Waals bonding is realized between the adjacent blocks. The measured single-crystal XRD data reveal no residual electron density in the interstitial region (Table 1). Thus, the refined formula unit of $Fe_{4.58(4)}AsTe_2$ indicates the composition of one

hexagonal block, which is enriched with Fe atoms and contains two confining layers of Te and one layer of the split As atoms in the middle. Coordination polyhedra of Fe atoms are shown in Figure 1a. While Fe1 and Fe2 are tetrahedrally coordinated by As and Te, the Fe3 atom center is octahedrally composed of three As and three Te atoms. The noticeably short interatomic distance of 2.10(3) Å is realized between the Fe1 and As1 atomic positions, which is presumably compensated by the partial occupancy of both Fe1 and As1. Interestingly, the facesharing connection of polyhedra is observed: three Fe1@Te₃As tetrahedra and three Fe2@As₃Te tetrahedra are condensed on the triangular faces of the Fe3@As₃Te₃ octahedron (Figure 1c). As a result, short distances between the Fe1 and Fe3 atoms (2.31 Å) and between the Fe2 and Fe3 atoms (2.61 Å) are realized. The latter are comparable with the Fe-Fe distances observed in Fe₃AsTe₂ (2.61 Å),¹⁴ Fe₃GeTe₂ (2.63 Å),⁴ Fe₅GeTe₂ (2.52 Å),¹⁸ and Fe₅Ge₂Te₂ (2.57 Å).¹⁵ At the same time, the short Fe1-Fe3 distance of 2.31 Å comes together with the reduced occupancy of the Fe1 atom, which is 0.29(2). According to Pauling's rule, coordination polyhedra avoid the face-sharing connection to minimize the electrostatic repulsion of cations. This rule can be bypassed in the case of noticeable covalent contribution to the chemical bonding, which may take place in Fe₅AsTe₂, too. In the electronic structure of the related Fe₃AsTe₂ compound, strong d-d hybridization in the vicinity of the Fermi level indicates covalent chemical bonds between the Fe atoms inside the hexagonal structural blocks.¹⁴

Real Structure of Polycrystalline Fe₅AsTe₂. Polycrystalline Fe₅AsTe₂ can be prepared from elements by two-stage annealing at 973 K. The sample contains no admixtures of starting materials or other binary or ternary compounds. Its elemental composition of $Fe_{4.9(1)}As_{1.0(1)}Te_{2.1(1)}$ is in good agreement with the nominal one according to EPMA. However, the as-prepared polycrystalline Fe₅AsTe₂ contains two isomorphous phases with close but slightly different c unit cell parameters, which results in the splitting of the 00l, 10l, and 01l diffraction reflections. At room temperature, the splitting of the 003 peak on the HRPXRD pattern corresponds to the values of c = 28.67 and 29.53 Å, which are in agreement with those obtained from the diffraction measurements of a crystal facet.²⁶ Remarkably, our single-crystal data were indexed using the $R\overline{3}m$ space group with a = 3.9937(5) Å and c = 28.858(4) Å. This crystal structure with the reduced unit cell may be stabilized due to the long-term annealing at high temperature in a small temperature gradient, where the CVT concentrations of Fe, As, and Te are close to equilibrium.

Figure 2 shows temperature-dependent HRPXRD patterns in the vicinity of the 003 diffraction reflection. In the temperature range between 300 and 473 K, this peak is split into two, indicating the coexistence of two different *c* unit cell parameters. At 573 K, the 003 peak has a shoulder, and at higher temperatures, a single peak is observed, indicating the single-phase region. Thus, high-temperature HRPXRD measurements unveil spinodal or other type of solid-to-solid decomposition of Fe₅AsTe₂, which occurs below \sim 673 K. The quenching of a polycrystalline sample into cold water after annealing at 973 K does not stabilize the high-temperature single-phase state. Notably, single crystals of Fe₅GeTe₂ demonstrate similar behavior around the transition temperature of ~550 K. For a given 00l reflection, a sharp single peak is observed only in the high-temperature state.¹⁷ The HRPXRD data collected at 973 K were used for the Rietveld



Figure 2. The 003 reflection of the HRPXRD patterns of Fe₅AsTe₂.

refinement of the crystal structure (Figure 3). Details of data collection and refinement are shown in Table 1. The obtained parameters of atomic positions are given in Table 2.



Figure 3. Experimental (black points) and calculated (red line) HRPXRD patterns of Fe_5AsTe_2 . Peak positions are given by black tips, and the difference plot is shown as a black line in the bottom part.

The refinement of the crystal structure against the HRPXRD data collected at 973 K confirms the structural model deduced from the single-crystal XRD experiment. Larger values of the thermal ellipsoid parameters are observed at 973 K, except the Fe1 position, where both single-crystal and HRPXRD refinements yield similar values of U_{eq} . This fact indicates the difference between single-crystal and HRPXRD refinements. While U_{eq} of Fe1 is comparable with that of Fe2 for the HRPXRD data, the observed large value of U_{eq} of Fe1 at room temperature points at the local disorder present in the singlecrystal sample. In the HRPXRD refinement, the split As1 site is shifted from the (0; 0; 0) position by dz = 0.0117(2), and the partially occupied Fe1 site contains 0.370(3) of Fe atoms. These structural features may be the origin of the phase separation, which occurs at temperatures below 673 K. In the crystal structure, two Fe1-As1 distances are observed due to splitting of the As1 position, 1.801(8) and 2.498(8) Å at 973 K, according to the HRPXRD data. The former is too short to be considered as bonding interaction, suggesting the aggregation of point defects in the crystal structure. The following scenario can be proposed, according to which half of the Fe1@Te₃As tetrahedra contain 74% of Fe atoms and 26% of vacancies with the equilibrium Fe1-As1 distance of

2.498(8) Å, while the other half are completely vacant. The appearance of such defects in the ordered way may yield two alternative values of the *c* unit cell parameter, corresponding to the phase separation in the real structure of Fe_5AsTe_2 .

Magnetic Properties. Magnetic susceptibility measured on single crystals of Fe₅AsTe₂ with the magnetic field applied parallel to the [001] direction (H||c) and in the in-plane orientation (H||ab) is shown in Figure 4. In the paramagnetic state above ~100 K, magnetic susceptibility is not orientationand field-dependent and can be fitted using the modified Curie–Weiss law, $\chi = \chi_0 + C_{CW}/(T - \theta)$, where χ_0 is the temperature-independent contribution, C_{CW} is the Curie-Weiss constant, and θ is the Weiss temperature. Fitting the data in 1 T magnetic field above 100 K yields $\chi_0 = 0.0136(1)$ emu mol⁻¹, $\theta = 47.4(4)$ K, and $C_{CW} = 6.50(4)$ emu K mol⁻¹. The latter corresponds to the effective magnetic moment of $M_{\rm eff}$ = 3.22(2) $\mu_{\rm B}/{\rm Fe}$, which is comparable with that of the Fe₃GeTe₂ layered vdW telluride, where the values between 3.9(2) and 4.9(1) $\mu_{\rm B}$ /Fe were registered depending on the Fe off-stoichiometry.⁶ The positive value of Weiss temperature indicates ferromagnetic interactions between magnetic centers. At low temperatures, magnetic susceptibility exhibits a broad maximum, below which the bifurcation of ZFC and FC signals is observed. The calculated Fisher's heat capacity, $\partial(\gamma T)/\partial T$, which is shown in the inset of Figure 4, middle panel, exhibits a minimum, indicating ferromagnetic phase transition with the Curie temperature of $T_{\rm C}$ = 48 K. The position of this minimum is in perfect agreement with the Weiss temperature of θ = 47.4(4) K. Furthermore, magnetization measurements reveal ferromagnetic hysteresis below 50 K for both orientations of crystals in the magnetic field. Above $T_{\rm C}$, the examined crystals show no remnant magnetization in the zero magnetic field, in agreement with the paramagnetic behavior.

The measured magnetic susceptibility and magnetization curves reveal the anisotropy of magnetic properties in the vicinity of the ferromagnetic phase transition. For $H \parallel c$, the low-temperature anomaly of magnetic susceptibility is sharp, while for $H \| ab$, it is smooth and lower in magnitude. The bifurcation of the ZFC and FC curves is observed below ~40 K in 0.1 T magnetic field for both orientations of crystals. Increasing the magnetic field gradually suppresses the lowtemperature anomaly in accordance with its ferromagnetic origin. Ferromagnetic hysteresis below T_C exhibits anisotropy, too. Values of the coercive field and saturation moment at 2 K are $H_c = 281$ and 100 mT and $M_s = 2.45(3)$ and 1.69(4) $\mu_B/$ f.u. for $H \| c$ and $H \| ab$, respectively. More complicated magnetic behavior is observed as a function of magnetic field for $H \parallel c$, displaying two distinct slopes. A low slope is observed near the coercive field, followed by the increase in slope before saturation. The lower slope may be induced by the spin-lattice coupling, as it was observed in Fe₅GeTe₂.¹³ Alternatively, the observed dual-slope magnetization may indicate a spin-flop transition accompanied by a partial spin reorientation, as it was proposed recently.²⁶ This scenario assumes the noncollinear or compensated nature of magnetism in Fe₅AsTe₂. Overall, the measured magnetic properties indicate perpendicular magnetic anisotropy in Fe₅AsTe₂, similar to the related Fe₃GeTe₂, 6,7 $Fe_5Ge_2Te_2^{15}$ and $Fe_5GeTe_2^{13}$ layered vdW tellurides. The Rhodes-Wohlfarth ratio (RWR) can be calculated as RWR = M_c/M_s , where M_c is defined using the equation $M_c(M_c + 2) =$ $M_{\rm eff}^{2}$, and $M_{\rm s}$ is the saturation moment in the ordered state.^{29,30} The values of $M_{\rm eff}$ = 3.22 $\mu_{\rm B}$ /Fe and $M_{\rm s}$ = 0.52 $\mu_{\rm B}$ /Fe (for H||c) yield RWR = 4.6. While RWR = 1 corresponds to



Figure 4. Top: magnetic susceptibility of Fe_5AsTe_2 . The inset shows a Curie–Weiss fit of the data in 1 T magnetic field. Middle: magnetization of Fe_5AsTe_2 with the magnetic field applied parallel to the [001] direction at various temperatures. The inset shows Fisher's heat capacity plot in the vicinity of the Curie temperature in 0.1 T magnetic field. Bottom: magnetization for the in-plane orientation of crystals at different temperatures.

the localized magnetism, RWR larger than 1 accompanied by low Curie temperature ($T_{\rm C} = 48 \ll 500$ K) suggests that itinerant magnetism is likely present in Fe₅AsTe₂.

The magnetic susceptibility of polycrystalline Fe_5AsTe_2 , which is shown in Figure 5, exhibits the low-temperature anomaly and bifurcation of the ZFC and FC curves, too. The data are in agreement with those reported recently.²⁶ However, the sample contains a ferromagnetic admixture that yields field-



Figure 5. Top: magnetic susceptibility of polycrystalline Fe_5AsTe_2 in comparison with the single-crystal sample for H||ab. Bottom: magnetization of polycrystalline Fe_5AsTe_2 at various temperatures.

dependent susceptibility at high temperatures and remnant magnetization with ferromagnetic hysteresis up to 400 K. Furthermore, the step-like anomaly at 125 K on the magnetic susceptibility curve in 0.1 T magnetic field indicates the presence of the Fe₃O₄ admixture, which exhibits the Verwey transition at this temperature, and shows ferromagnetism up to 853 K.^{31,32} Despite the presence of the ferromagnetic admixture, magnetization curves of polycrystalline Fe₅AsTe₂ exhibit two distinct slopes in low magnetic fields at low temperatures, in agreement with the single-crystal measurements. Above the Curie temperature of $T_{\rm C}$ = 48 K, this dual-slope behavior disappears.

CONCLUSIONS

Fe-rich cleavable van der Waals tellurides, such as Fe_3AsTe_2 , Fe_3GeTe_2 , Fe_5GeTe_2 , and $Fe_5Ge_2Te_2$, are a source of emergent two-dimensional magnetic nanomaterials. They are able to provide the control of spins by filtering through the tunnel junctions realized in heterostructures. Furthermore, novel topological and magnetic solutions, as well as nontrivial arrangements of spins, are expected in the two-dimensional van der Waals architectures. Here, we present the synthesis, crystal growth, crystal structure, and magnetic properties of the Fe_5AsTe_2 van der Waals telluride. Single-crystal and highresolution powder X-ray diffraction experiments reveal a layered crystal structure of the Fe_5GeTe_2 structure type with no residual electron density in the van der Waals gap. Bulk crystals demonstrate high-temperature paramagnetic behavior with the effective magnetic moment of $3.22(2) \mu_B$ per Fe atom. At low temperatures, weak ferromagnetism emerges below the Curie temperature of 48 K with the saturation magnetization of 2.45(3) μ_B per formula unit when the magnetic field is applied perpendicular to the structural layers. The dual-slope behavior of magnetization in low magnetic fields points at possible spin–lattice coupling or spin-flop transition, for which the ferromagnetic state has the compensated nature. Fe₅AsTe₂ demonstrates perpendicular magnetic anisotropy similar to the Fe₃GeTe₂ and Fe₅Ge₂Te₂ layered van der Waals tellurides. The Rhodes–Wohlfarth ratio of 4.6 indicates itinerant ferromagnetism in Fe₅AsTe₂. Its delocalized nature is relevant for the production of spin-polarized current.

ASSOCIATED CONTENT

Accession Codes

CCDC 2156155 and 2156163 contain the supplementary crystallographic data for this paper. These data can be obtained free of charge via www.ccdc.cam.ac.uk/data_request/cif, or by emailing data_request@ccdc.cam.ac.uk, or by contacting The Cambridge Crystallographic Data Centre, 12 Union Road, Cambridge CB2 1EZ, UK; fax: +44 1223 336033.

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Author Contributions

The manuscript was written through contributions of all authors.

Notes

The authors declare no competing financial interest.

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