Anisotropic magnetic properties and tunable conductivity in two-dimensional layered NaCrX₂ (X=Te,Se,S) single crystals

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Monolayer NaCrX₂ (X=Te,Se,S) were theoretically proposed to be two-dimensional intrinsic ferromagnetic semiconductors while their physical properties have not been thoroughly investigated in bulk single crystals. We report the single-crystal growth, structural, magnetic and electronic transport properties of NaCr(Teₓ−ₓSeₓ)₂ (0 ≤ x ≤ 1) and NaCrS₂. For NaCr(Teₓ−ₓSeₓ)₂, the strong perpendicular magnetic anisotropy of NaCrTe₂ can be gradually tuned to be a nearly isotropic one by Se-doping. Meanwhile, a systematic change in the conductivity with increasing x is observed, displaying a doping-induced metal-insulator-like transition. Under magnetic field larger than 30 kOe, both NaCrTe₂ and NaCrSe₂ can be polarized to a ferromagnetic state. While for NaCrS₂, robust antiferromagnetism is observed up to 70 kOe and two field-induced metamagnetic transitions are identified along H||ab. These intriguing properties together with the potential to be exfoliated down to few-layer thickness make NaCrX₂ (X=Te,Se,S) promising for exploring spintronic applications.

INTRODUCTION

Magnetism in two dimensions has been a fascinating topic in condensed matter physics for decades. From the initial investigations on thin-film magnets to the recent discovery of two-dimensional (2D) magnetic order in ultra-thin van der Waals (vdW) materials, a wide range of possibilities for both spintronic applications and fundamental research have been opened up. For few-layer vdW crystals, 2D magnetism has been realized in metallic Fe₃GeTe₂, semiconducting CrI₃/Gr₂Ge₂Te₆, and insulating FePS₃ due to the large magnetic anisotropy which could counteract thermal fluctuations. These materials can serve as different building blocks of vdW heterostructures depending on their conductivity and exploring potential applications in novel magnetoelectronic devices. Therefore magnetic anisotropy and conductivity are two key properties of 2D magnetic materials. Finding new 2D materials and ways to tune these properties would be quite important in the research of 2D magnetism.

Bulk CrTe₂ with 1T phase is a vdW ferromagnet with in-plane magnetic anisotropy and Curie temperature of 310 K. Remarkably, in ultra-thin flakes or films, the easy axis of CrTe₂ changes from in-plane to out-of-plane and room-temperature ferromagnetism is retained. Moreover, a recent study identified a zigzag type antiferromagnetic order in monolayer CrTe₂, demonstrating the intricacy of 2D magnetism in this material. On the other hand, the intercalations of metal atoms into the vdW gap of CrX₂ (X=Te,Se,S) can form plenty of new phases. Although the structures of these intercalated CrTe₂ phases are non-vdW type, many of them are still able to be exfoliated into nanosheets while keep intriguing physical properties. For example, the room-temperature ferromagnetism in 10 nm-thick Cr-intercalated CrTe₂ and the superionic behavior in 1.1 nm-thick AgCrS₂ have been reported recently. Especially for the latter, the structure of so-called AgCrS₂ monolayer consists of one Ag layer sandwiched between two [CrS₂] layers with actual formula AgCrS₂4, which has been shown to be stable experimentally.

The Na-intercalated NaCrX₂ (X=Te,Se,S) are recently proposed to be a class of monolayer ferromagnetic semiconductors by first-principles calculations. Although the crystal structures are non-vdW type, their cleavage energies are comparable with other 2D materials. It also should be mentioned that, in previous research the vdW magnetic material CrTe₂ is mainly made by deintercalating the alkali metal of KCrTe₂. However the physical properties of NaCrTe₂ have not been thoroughly investigated until recently. Experimentally, NaCrTe₂ is determined to be an A-type antiferromagnet with T_N=110 K and perpendicular magnetic anisotropy. With applied magnetic field, spin-flip-driven giant negative and angle-dependent magnetoresistance are observed for single crystals of NaCrTe₂. For NaCrSe₂ and NaCrS₂, although their antiferromagnetic transition temperature and structure have already been determined, the anisotropic and field-induced magnetic properties are still unclear due to the lack of single crystals. Furthermore, chemical doping has been proved to be an effective way to tune the magnetic and transport properties of layered magnetic materials. It would be interesting to explore the Se-doping effect on NaCrTe₂.

In this paper, we report the successful growth of single crystals of NaCr(Teₓ−ₓSeₓ)₂ and NaCrS₂. These crystals show intriguing magnetic and transport properties, including field-induced spin-flip transitions, giant negative magnetoresistance, chemical doping controlled magnetic anisotropy and conductivity. Our findings suggest the NaCrX₂ series are promising candidates for further
investigations in 2D limit.

**METHODS**

Single crystals of NaCr(\(\text{Te}_{1-x}\text{Se}_x\))\(_2_\) and NaCrS\(_2_\) were grown by melting stoichiometric elements. High-purity Na, Cr, Te and Se/S were mixed in the mole ratio 1:1:2(1-x):2x. These reagents were mixed in alumina crucibles and sealed into an evacuated quartz tube. The assembly was heated up to 1050 °C and maintained at this temperature for 24 h. Then it was slow-cooled to 800 °C at a rate of 3 °C/h and annealed at this temperature for one day before furnace-cooled to room temperature. In order to make the reaction adequate, before heating to 1050 °C, the assembly would stay for ten hours at the temperatures slightly below the melting or boiling point of each reactant. The plane size of the obtained NaCr(\(\text{Te}_{1-x}\text{Se}_x\))\(_2_\) single crystals is up to 6 mm×6 mm, while the size of NaCrS\(_2_\) is smaller which is typically 1 mm×1 mm. NaCrTe\(_2_\) is air-sensitive, its shining surface could be oxidized and discolored after being put in the air for a few hours. With Se-doping, the crystal become less air-sensitive and it takes about two days for NaCrSe\(_2_\) to become degenerative in the air. NaCrS\(_2_\) is air-stable.

We characterized all samples with energy dispersive x-ray spectroscopy (EDS, Oxford X-Max 50). For NaCr(\(\text{Te}_{1-x}\text{Se}_x\))\(_2_\) with x=0.1 and x=0.2, the EDS value is quite close to the nominal value. For x>0.3, the doping concentration may slightly deviate from the nominal value (the estimated error is about 20%). In order to be accurate, all measured samples have been carefully checked by EDS. The descriptions in this paper about doping level x all refer to the EDS values.

X-ray diffraction (XRD) of the samples were collected from a Bruker D8 Advance X-ray diffractometer and a Bruker D8 VENTURE single-crystal diffractometer using Cu K\(_\alpha\) radiation. Magnetization and electrical transport measurements were carried out in Quantum Design MPMS3 and PPMS-14T, respectively. The dimensions of exfoliated NaCrTe\(_2_\) and NaCrS\(_2_\) nanoflakes were checked by a Bruker edge dimension atomic force microscope.

**RESULTS AND DISCUSSIONS**

Crystal structure of NaCrX\(_2_\)

As shown in Fig. 1(a) and confirmed by XRD analysis, NaCrX\(_2_\) crystallizes in a hexagonal structure with the space group of P-3\(\text{m}1\) for X=Te and R-3\(\text{m}\) for X=Se/S, same as previous reports\[19,21\]. The crystal structure of NaCrTe\(_2_\) can be considered as the intercalation of Na atoms between 1\(\text{T}-\)CrTe\(_2_\) layers. Na and Cr atoms stack alternately along the c-axis in the same site. The Cr atoms form a triangular lattice in the \(\text{ab}\)-plane. When Te was substituted by Se or S, the major change in crystal structure is the interlayer stacking order. Comparing with NaCrTe\(_2_\), for NaCrSe\(_2_\) and NaCrS\(_2_\), both Na- and Cr-triangular lattice layers are stacked along the c-
axis with (1/3 1/3) translation in the ab-plane. So the c-lattice parameter has almost tripled. These two different structures are referred as ‘Te-phase’ and ‘Se-phase’ respectively in the following descriptions.

For NaCr(Se_x Te_{1-x})_2 (0 \leq x \leq 1), the lattice parameters were obtained by refining the single crystal x-ray diffraction data and plotted in Fig. 1(c) and (d). The results show that the samples with x < 0.5 maintain the Te-phase while the samples with x > 0.68 are confirmed to have the Se-phase. Therefore the phase boundary may exist near x=0.6, although it has not been accurately determined. The precession images from the single crystal x-ray diffraction data display streaking features for most doped samples, especially for the heavily doped ones. This indicates that notable stacking disorders and possible phase separations may exist in NaCr(Se_x Te_{1-x})_2 (Figure S2 in Supplementary Materials). In the Te-phase zone, both the a- and c-lattice parameters do not follow a monotonic change with increasing x. Compare with NaCrTe_2, there is a 7% shrinkage for the a-axis of NaCrSe_2. The c-lattice parameter for NaCrSe_2 is 19.485 Å, which decreases about 4% compared with that of NaCrSe_2.

Previous calculations of the cleavage energies suggest that NaCrX_2 can be exfoliated to a thickness of a few layers. We performed the mechanical exfoliation of bulk NaCrX_2 single crystals using Scotch tape. Nanosheets of NaCrTe_2 and NaCrSe_2 with thickness 20-30 nm could be obtained, which is demonstrated by the atomic force microscopy images in Fig. 1(e) and (f). Recently Jing Peng et al. demonstrated that isostructural AgCrS_2 can be exfoliated into 1.1 nm nanosheet which consists of one Ag-layer sandwiched between two CrS_2-layers. Therefore this class of materials are promising 2D materials for further investigations. Future investigation on whether NaCrX_2 is stable under similar exfoliation method as AgCrS_2 could be stimulated.

**Magnetic properties of NaCr(Se_x Te_{1-x})_2**

The temperature dependent magnetic susceptibility \( \chi(T) \) and isothermal magnetization \( M(H) \) of NaCr(Se_x Te_{1-x})_2 single crystals are shown in Fig. 2. Demagnetization corrections with methods used in our previous publication have been applied on the \( H \parallel c \) data and the applied magnetic field \( H_{app} \) in \( M(H) \) curve is replaced by the internal field \( H_{int} \) in Fig. 2. For NaCrTe_2, \( \chi(T) \) curve under \( H \parallel c \) exhibits a sharp drop down to nearly zero below \( T_N=110 \) K in contrast to the weak cusp and plateau-like feature under \( H \parallel ab \). This suggests the development of an A-type antiferromagnetic order (ferromagnetic intralayer and antiferromagnetic interlayer couplings) with moment aligned along c-axis. The Curie-Weiss fit on the high-temperature paramagnetic susceptibility reveals \( \mu_{eff}/Cr = 3.8 \mu_B \) and \( \theta_{CW} = 155 \) K. The large positive \( \theta_{CW} \) value indicates the strong intralayer ferromagnetic correlations. The hysteresis loops at 2 K under \( H \parallel c \) and \( H \parallel ab \) demonstrate the strong perpendicular magnetic anisotropy (PMA) and a spin-flip transition to the ferromagnetic state near \( H_{ab}=30 \) kOe. These observations are similar as previous report. The saturation moment for NaCrTe_2 is 2.6 \( \mu_B/Cr \), which is a bit lower than the theoretical value 3.0 \( \mu_B \) for Cr^{3+} in a localized model.

For x=0.1, \( T_N \) decreases to 78 K while the features of A-type antiferromagnetic order and perpendicular magnetic anisotropy still persist. The spin-flip transition near...
and (d) Magnetization isotherms measured at selected temperatures. On the other hand, fitting the high-temperature paramagnetic data to the Curie-Weiss law yields $\mu_{eff}/Cr = 3.8\,\mu_B$ and $\theta_{CW} = 108\,K$. The positive $\theta_{CW}$ temperature indicates ferromagnetic correlations are still strong in each individual layer, while the interlayer coupling is antiferromagnetic, similar as NaCrTe$_2$ and many other 2D layered magnetic materials.

In contrast to the strong anisotropic magnetization under $H//c$ and $H//ab$ for NaCrTe$_2$, the magnetic anisotropy of NaCrSe$_2$ is much smaller. Without applying a demagnetization correction, NaCrSe$_2$ appears to have essentially zero anisotropy. After the correction, a weak PMA could be identified from the $M(H)$ curves at 2 K [Fig. 3(b)]. Under $H > 30\,kOe$ for both directions and $T=2\,K$, the magnetization becomes saturated with a saturation moment of $3.0\,\mu_B/Cr$, which is larger than that of NaCrTe$_2$ and accurately equals to the theoretical value of $Cr^{3+}$ in a localized model. It should be noted that, before reaching saturated value, the magnetization increases quickly and linearly with increasing field along both directions. This indicates that although the spins of NaCrSe$_2$ are antiferromagnetically aligned at low field, they could be continuously canted along the field direction with increasing field. Fig. 3(c) and (d) show the $M(H)$ curves at different temperatures. The saturation field (c) also exists but with a much larger hysteresis comparing with that of $x=0$. An important modification of magnetic anisotropy is that the PMA seems to get weakened with Se-doping. If we choose the ratio $M_r/M_{ab}$ at $H=60\,kOe$ and $T=2\,K$ as a criterion, it decreases from 2.3 for $x=0$ to 1.5 for $x=0.10$.

With increasing doping concentration $x$, $T_N$ continuously shifts to lower temperature and the spin-flip transition gradually vanishes as shown in Fig. 2(c)-(d) and (h)-(j). The $M_r/M_{ab}$ at 60 kOe and 2 K also gradually decreases from 1.4 ($x=0.19$) to 1.2 ($x=0.30$), and finally to 0.95 ($x=0.42$). This means the magnetic anisotropy gradually evolves from PMA to a slightly preferred in-plane magnetization. On the other hand, for $x \geq 0.19$, there is a bifurcation between zero-field-cooling (ZFC) and field-cooling (FC) magnetization below $T_N$, which implies the emergence of a spin-glass state. In addition, the Curie-Weiss fit on the doped samples reveals similar values of effective moment but lower $\theta_{CW}$ values (131 K-106 K), suggest a slightly weakened ferromagnetic correlations.

The samples with $0 \leq x \leq 0.42$ discussed above all belongs to the Te-phase. Our XRD analysis reveal that at least from $x=0.68$, NaCr(Se$_{1-x}$Te$_x$)$_2$ enters the Se-phase. Let us first discuss the magnetic properties of NaCrSe$_2$ which is shown in Fig. 3. A cusp at $T_N=39\,K$ is observed in the $\chi(T)$ curve indicating an antiferromagnetic transition. On the other hand, fitting the high-
transition moment gradually decreases with increasing temperature. At 50 K which is well above $T_N$, the M(H) curve still exhibits a nonlinear curvature and a large moment ($\sim 2.2 \mu_B/\text{Cr}$) at 60 kOe. In addition, the M(T) curve also deviates from Curie-Weiss behavior below 200 K, which are actually observed for all NaCr$X_2$ (X=Te,Se,S) samples. These observations suggest that strong magnetic fluctuations or short-range magnetic order may develop well above $T_N$.

For $x=0.68$ and $x=0.79$ with Se-phase, the spin-glass behavior are quite similar as $x=0.42$ with Te-phase. The related magnetization data is not shown here but the results are plotted in the phase diagram of Fig. 6. Further discussions about the evolution of anisotropic magnetic properties of NaCr$(\text{Te}_{1-x}\text{Se}_x)_2$ will be presented in the following section with the phase diagram.

**Tunable conductivity in NaCr$(\text{Te}_{1-x}\text{Se}_x)_2$**

Previously, NaCrTe$_2$ was reported to exhibit a metal-insulator-like transition due to the formation of $A$-type antiferromagnetic ordering[20]. Similarly, the resistivity of $x=0.1$ sample has a typical metallic behavior above 130 K [Fig. 4(a)], while a sharp jump appears at $T_N=78$ K with decreasing temperature which should result from the localization of charge carriers by long-range antiferromagnetic order. With further decreasing temperature, the resistivity continues to increase with a smaller slope which might be attributed to the impurity scattering effect by chemical-doping. Under magnetic field along c-axis, a giant negative magnetoresistance appears with maximal value up to 48% at 5 K and 70 kOe. This behavior is directly associated with the field-induced spin-flip transition and due to the reduced spin scattering to the electrons in the ferromagnetic state comparing with that in the antiferromagnetic states.

Comparing with the previously reported resistivity of NaCrTe$_2$[20], we noticed that the value for $x=0.1$ approximately increases by an order of magnitude. As shown in Fig. 5(a), intriguingly, with further increasing Se-doping concentration $x$, the temperature dependent resistivity of NaCr$(\text{Te}_{1-x}\text{Se}_x)_2$ becomes fully semiconducting-like and the absolute value of resistivity continuously increases by several orders. For NaCrSe$_2$ with $x=1$, the resistivity is about $\sim 10 \Omega \cdot cm$ and the data below 95 K cannot be obtained due to the upper limit of PPMS measurement. Thus a Se-doping induced metal-insulator transition is observed for NaCr$(\text{Te}_{1-x}\text{Se}_x)_2$.

Density functional theory calculations have shown that both NaCrTe$_2$ and NaCrSe$_2$ are semiconductors, their band gaps are 0.59 eV and 0.77 eV respectively[18]. In order to check whether the doping controlled conductivity is due to the gradual increasing of the band gap, we have tried to fit the resistivity data using the thermal activation model as described by $\rho=\rho_0\exp(E_a/k_BT)$. Where $E_a$ is the energy gap and $k_B$ is the Boltzmann constant. Therefore a fine fitting result using this formula can only be obtained when $\ln\rho$ and $T^{-1}$ follows a linear relationship. However, as shown in Fig. 5(b), there are not well defined linear relations between $\ln\rho$ and $T^{-1}$ for all samples, especially for $x=0.42$ and $x=0.76$.

For chemical doped samples, the doping-induced strong disorder potential might trap itinerant electrons and lead to a metal-insulator transition, which is the famous Anderson localization[27]. For an Anderson insulator at low temperatures, there are electronic states trapped in the vicinity of the Fermi surface and the hopping transport by localized electrons would be described by the variable range hopping (VRH) model $\rho=\rho_0\exp(T_0/T)^{1/(\nu+1)}$. In this formula, $\nu=0$ denotes the traditional insulator with band gap (same as thermal activation model described above). $\nu=1$, 2 and 3 correspond to one-, two-, and three-dimensional material

![FIG. 5. (a) Temperature dependent resistivity data for NaCr$(\text{Te}_{1-x}\text{Se}_x)_2$ ($H=0$). (b) $\ln\rho$ versus $T^{-1}$ plot of $x=1$ for all measured temperatures. The inset shows same plots for $x=0.42$ and $x=0.76$. (c,d) Low temperature $\ln\rho$ versus $T^{-1/(\nu+1)}$ plots of $x=0.76$ and $x=0.42$, respectively. $\nu$ is the parameter from VRH formula.](image-url)
with Anderson localization, respectively. In Fig. 5(c) and (d), the lnρ versus T−1/(ν+1) plots for x=0.42 and 0.76 from 2 K to 10 K are presented. Although a perfect straight line is not observed, the formula using ν=2 or ν=3 would clearly give a much better fitting results comparing with that using ν=0, especially for x=0.76. Our results indicate that the significant enhancement of resistivity in NaCr(Te1−xSex)2 may partially originate from the Anderson localization. Besides, the tuning of band gap by Se-doping would also be possible. The electronic transport properties of NaCr(Se1−xTex)2 may contains contributions from these two aspects.

Discussion on the phase diagram of NaCr(Se1−xTex)2

The T-x phase diagram of NaCr(Se1−xTex)2 is presented in Fig. 6, which summarizes the major experimental results above. Three features should be mentioned about this phase diagram. Firstly, considering either from Se-doped NaCrTe2 or Te-doped NaCrSe2, the antiferromagnetic transition temperature gradually decreases and the system enters a spin-glass state with increasing doping concentration. Secondly, the magnetic anisotropy can be effectively tuned by chemical doping in this system. NaCrTe2 possesses a strong PMA and it continuously gets weakened and evolves into a nearly isotropic magnetic behavior at x=0.42 while maintain the Te-phase. For NaCrSe2 at the other end, a weak PMA is identified. Thirdly, A systematic change in the conductivity of NaCr(Se1−xTex)2 occurs on increasing x, from bad metallic behavior in NaCrTe2 to the semiconducting or insulating behavior in NaCrSe2.

The doping-induced disorder effect and stacking faults should be responsible for the emergence of the spin glass state[29]. It also may play an important role in tuning the magnetic anisotropy in NaCr(Se1−xTex)2. In previous investigations on materials for high-density magnetic recording media, chemical disorder has been shown to have important influence on the magnetocrystalline anisotropy energy (MAE) [31–32]. It may either drastically reduce the MAE or tune the MAE to a maximum value[33–36]. For vdW material, recent studies reveal that Ni1−xFexPS3 and Fe5(1−x)Co5xGeTe2 enable chemical tuning of easy-plane and easy-axis anisotropies [24, 25, 37]. Magnetic anisotropy is a key property of 2D magnets, which is required for counteracting thermal fluctuations. There have been reports about pressure control of magnetic anisotropy on CrGeTe3[34] and tensile strain-tunable magnetic anisotropy in monolayer CrX3 (x=Cl, Br, I)[35]. Our study provides a new example for chemical disorder or stacking fault controlled magnetic anisotropy in 2D magnetic material NaCr(Se1−xTex)2.

In addition, one should notice that the magnetic anisotropy also makes a big difference between NaCrTe2 and NaCrSe2, both of them seem to have no evident chemical disorder. A possible speculation is that, for NaCrSe2 with relatively much lighter Se element, the spin-orbit coupling effect may become weak and results in a weak PMA[34]. As we mentioned above, the saturation moment of NaCrSe2 agrees well with the expectation of Cr3+ spin S=3/2 model without orbital moment while that of NaCrTe2 has a notable smaller value which might be due to the enhanced spin-orbital coupling.

The tuning of conductivity by chemical doping in 2D magnetic material has been rarely reported. This effect found in NaCr(Se1−xTex)2, possibly due to Anderson localization and the change of band structure, may have important applications in designing novel spintronic devices. Particularly, the chemical doping in NaCr(Se1−xTex)2 could simultaneously tune both the conductivity and magnetic anisotropy.

Field-induced metamagnetic transitions in NaCrS2

For the magnetic properties of NaCrS2, so far as we know, there has not been any investigations on the single crystals. We present the anisotropic magnetization data on NaCrS2 single crystals in Fig. 7. The different temperature dependent features of χab and χc below TN=19 K indicate the ordered magnetic moment should be confined within ab-plane [Fig. 7(a)]. Fig. 7(b) presents the magnetic hysteresis loops at 2 K. Under magnetic field applied along the hard-axis (H || c), a linear relationship between magnetization and field is observed. But for field applied along the easy ab-plane, a sudden magnetization jump is revealed at around 25 kOe suggest the occurrence of field-induced metamagnetic transition. From the magnetization isothersms at higher temperatures [Fig. 7 (c) and (d)], the linear behavior persists for H || c and the magnetization jump gradually weakens with increasing temperature for H || ab. The contour
This is supported by the cusp-like feature in the $\chi_{ab} - T$ curve at $T_N$, as shown in Fig. 7 (f). The antiferromagnetic transition seems to be quite robust under field. $T_N$ only has a slight shift to 16 K under $H = 70 \text{kOe}$. In addition, $M(H)$ curve follows a linear relationship up to 70 kOe with a small value of moment ($\sim 0.7 \mu_B/\text{Cr}$) under this field. Secondly, for $H_{ab} \leq 20 \text{kOe}$ the magnetic susceptibilities have a sharp drop below $T_N$. In contrast, for $H_{ab} = 30 \text{kOe}$ and 70 kOe, the magnetic susceptibilities exhibit a plateau below $T_N$, which is a typical feature of canted antiferromagnetic magnetic order. Therefore, new types of antiferromagnetic order are expected under in-plane magnetic field, which would be an interesting topic for further investigations using neutron scattering.

CONCLUSIONS

In summary, the physical properties of NaCrX$_2$ (X=Te,Se,S) are investigated in single-crystal form. For NaCr(Se$_{1-x}$Te$_x$)$_2$, a field-induced spin-flip transition together with a giant negative magnetoresistance are observed at $x \leq 0.1$. At higher doping level, a spin glass state emerges. The most prominent feature is that both the magnetic anisotropy and the conductivity can be effectively tuned by $x$. For NaCrS$_2$, two magnetic field induced metamagnetic transitions are identified. We further demonstrate that these crystals can be mechanically-exfoliated into nano-flakes. These properties would make NaCrX$_2$ a promising material playground for further investigations on 2D magnetism and designing novel...
magneto-electronic devices. Furthermore, our samples also provide a route to vdW layered CrX₂ through deintercalation.

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[1] K. S. Burch, D. Mandrus, and J.-G. Park, Nature 563, 47 (2018).
[2] H. Li, S. Ruan, and Y.-J. Zeng, Advanced Materials 31, 1900065 (2019).
[3] D. L. Cortie, G. L. Causer, K. C. Rule, H. Fritzschke, W. Kreuzpaitner, and F. Klose, Advanced Functional Materials 30, 1901414 (2020).
[4] Y. Deng, Y. Yu, Y. Song, J. Zhang, N. Zhang, Z. Sun, Y. Yi, Y. Z. Wu, S. Wu, J. Zhu, J. Wang, X. H. Chen, and Y. Zhang, Nature 563, 94 (2018).
[5] Z. Fei, B. Huang, P. Malinowski, W. Tang, T. Song, J. Sanchez, W. Yao, D. Xiao, X. Zhu, A. F. May, W. Wu, D. H. Cobden, J.-H. Chu, and X. Xu, Nature Materials 17, 778 (2018).
[6] B. Huang, G. Clark, E. Navarro-Moratalla, D. R. Klein, R. Cheng, K. L. Seyler, D. Zhong, E. Schmidgall, M. A. McGuire, and D. H. Cobden, Nature 546, 270 (2017).
[7] C. Gong, L. Li, Z. Li, H. Ji, A. Stern, Y. Xia, T. Cao, W. Bao, C. Wang, and Y. Wang, Nature 546, 265 (2017).
[8] J.-U. Lee, S. Lee, J. H. Ryoo, S. Kang, Y. T. Kim, P. Kim, C.-H. Park, J.-G. Park, and H. Cheong, Nano Letters 16, 7433 (2016).
[9] X. Wang, K. Du, Y. Y. F. Liu, P. Hu, J. Zhang, Q. Zhang, M. H. S. Owen, X. Lu, C. K. Gan, P. Sengupta, K. Kloc, and Q. Xiong, 2D Materials 3, 031009 (2016).
[10] D. C. Freitas, R. Weht, A. Sulpice, G. Remenyi, P. Strobel, F. Gay, J. Marcus, and M. Nuñez-Regueiro, Journal of Physics: Condensed Matter 27, 176002 (2015).
[11] X. Sun, W. Li, X. Wang, Q. Sun, T. Zhang, Z. Wang, L. Liu, D. Li, S. Feng, S. Zhong, H. Wang, V. Bouchiat, M. Nuñez-Regueiro, N. Rougemaille, J. Coraux, A. Purbawati, A. Hadj-Azzem, Z. Wang, B. Dong, X. Wu, T. Yang, G. Yu, B. Wang, Z. Han, X. Han, and Z. Zhang, Nano Research 13, 3358 (2020).
[12] X. Zhang, Q. Lu, W. Liu, Y. Niu, J. Sun, S. Cook, M. Vaninger, P. F. Miceli, D. J. Singh, S.-W. Lian, T.-R. Chang, X. He, J. Du, L. He, R. Zhang, G. Bian, and Y. Xu, Nature Communications 12, 2492 (2021).
[13] L. Meng, Z. Zhou, M. Xu, S. Yang, K. Si, L. Liu, X. Wang, H. Jiang, B. Li, P. Qin, P. Zhang, J. Wang, Z. Liu, P. Tang, Y. Ye, W. Zhou, L. Bao, H.-J. Gao, and Y. Gong, Nature Communications 12, 809 (2021).
[14] F. Fabre, A. Finco, A. Purbawati, A. Hadj-Azzem, N. Rougemaille, J. Coraux, I. Philip, and V. Jacques, Phys. Rev. Materials 5, 034008 (2021).
[15] J.-J. Xian, C. Wang, J.-H. Nie, R. Li, M. Han, J. Lin, W.-H. Zhang, Z.-Y. Liu, Z.-M. Zhang, M.-P. Miao, Y. Yi, S. Wu, X. Chen, J. Han, Z. Xia, W. Ji, and Y.-S. Fu, Nature Communications 13, 257 (2022).
[16] M. Huang, Z. Ma, S. Wang, S. Li, M. Li, J. Xiang, P. Liu, G. Hu, Z. Zhang, Z. Sun, Y. Lu, Z. Sheng, G. Chen, Y.-L. Chueh, S. A. Yang, and B. Xiang, 2D Materials 8, 031003 (2021).
[17] J. Peng, Y. Liu, H. Lv, Y. Li, Y. Lin, Y. Su, J. Wu, H. Liu, Y. Guo, Z. Zhuo, X. Wu, C. Wu, and Y. Xie, Nature Chemistry 13, 1235 (2021).
[18] W. Xu, S. Ali, Y. Jin, X. Wu, and H. Xu, ACS Applied Electronic Materials 2, 3853 (2020).
[19] S. Kobayashi, H. Ueda, C. Michioka, and K. Yoshimura, Inorganic Chemistry 55, 7407 (2016).
[20] J. Wang, J. Deng, X. Liang, G. Gao, T. Ying, S. Tian, H. Lei, Y. Song, X. Chen, J.-g. Guo, and X. Chen, Phys. Rev. Materials 5, 091401 (2021).
[21] F. Engelsman, G. Wiegers, F. Jellinek, and B. Van Laar, Journal of Solid State Chemistry 5, 574 (1973).
[22] C.-K. Tian, C. Wang, W. Ji, J.-C. Wang, T.-L. Xia, L. Wang, J.-J. Liu, X.-Z. Zhang, and P. Cheng, Phys. Rev. B 99, 184428 (2019).
[23] J. Sheng, X. Li, C. Tian, J. Song, X. Li, G. Sun, T. Xia, J. Wang, J. Liu, D. Xu, H. Zhang, X. Tong, W. Luo, L. Wu, W. Bao, and P. Cheng, Phys. Rev. B 101, 174516 (2020).
[24] C. Tian, F. Pan, S. Xu, C. Ai, T. Xia, and P. Cheng, Applied Physics Letters 116, 202402 (2020).
[25] S. Lee, J. Park, Y. Choi, K. Raju, W.-T. Chen, R. Sankar, and K.-Y. Choi, Phys. Rev. B 104, 174412 (2021).
[26] See Supplemental Material at [URL will be inserted by publisher] for details about structural and composition analysis of NaCr(Te₁₋₂Se₂)₂ through single-crystal x-ray diffraction and EDS.
[27] P. W. Anderson, Phys. Rev. 109, 1492 (1955).
[28] T. Ying, Y. Gu, X. Chen, X. Wang, S. Jin, L. Zhao, W. Zhang, and X. Chen, Science Advances 2, e1501283 (2016).
[29] K. Binder and A. P. Young, Rev. Mod. Phys. 58, 801 (1986).
[30] C. J. Aas, L. Sønnygod, J. S. Chen, and R. W. Chantrell, Applied Physics Letters 99, 132501 (2011).
[31] M. Si, A. Izdarkar, and C. Ederer, arXiv:2111.12492 (2021).
[32] C. Neise, S. Schonecker, M. Richter, K. Koepenick, and H. Eschrig, physica status solidi (b) 248, 2398 (2011).
[33] A. F. May, M.-H. Du, V. R. Cooper, and M. A. McGuire, Phys. Rev. Materials 4, 074008 (2020).
[34] T. Sakurai, B. Rubrecht, L. T. Corredor, R. Takehara, M. Yasutani, J. Zeisner, A. Alfonsov, S. Selter, S. Aswartham, A. U. B. Wolter, B. Büchner, H. Ohta, and V. Kataev, Phys. Rev. B 103, 024404 (2021).
[35] L. Webster and J.-A. Yan, Phys. Rev. B 98, 144411 (2018).
[36] F. Xue, Y. Hou, Z. Wang, and R. Wu, Phys. Rev. B 100, 224429 (2019).
[37] D. C. Johnston, Phys. Rev. B 95, 094421 (2017).