# Supporting information for: Soft chemical synthesis of H<sub>x</sub>CrS<sub>2</sub>: an antiferromagnetic material with alternating amorphous and crystalline layers

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#### **1** Experimental Details

#### 1.1 NaCrS<sub>2</sub> synthesis

Polycrystalline NaCrS<sub>2</sub> powders were synthesized by solid-state reaction of Na<sub>2</sub>S (Alfa Aesar, anhydrous), Cr (Alfa Aesar, ~100 mesh, 99 % metal basis) and S (Alfa Aesar, sublimed, ~100 mesh, 99.5 %) powders, which were mixed in a molar ratio of 0.5: 1: 3.5. Reagents were mixed homogeneously in an argon filled glove box before being placed in alumina crucibles with a diameter of 13 mm and a height of 25 mm and sealed in a silica quartz tube with a outer diameter of 16 mm and inner diameter of 14 mm under vacuum. The length of a sealed quartz tube was about 8 cm. The sealed quartz tube was heated to 800 °C with a ramp rate of 2 °C/hour and dwelled for 48 hours before cooling to room temperature.

NaCrS<sub>2</sub> single crystals were grown from Na-S flux based on a published literature procedure.<sup>S1</sup> Na<sub>2</sub>S, Cr and S powders were mixed in a molar ratio of 3: 1: 4. The powders were ground carefully in an argon-filled glove box before being placed in alumina crucibles and sealed in a silica quartz tube under vacuum. The size of the alumina curcibles and the sealed quartz tube were the same as described in the above paragraph. The sealed quartz tube was heated to 1000 °C with a ramp rate of 2 °C/hour and dwelled for 24 hours before being cooled down to 300 °C with a rate of 25 °C/hour. After cooling to room temperature, the silica quartz tube was opened in the air, and the alumina crucibles were placed in the deionized water to dissolve the excess Na-S flux. NaCrS<sub>2</sub> single crystals were picked out from the solutions and washed with deionized water thoroughly before being dried in a vacuum oven at 150 °C overnight.

### 1.2 Proton exchange of the $NaCrS_2$ polycrystalline powders and single crystals

Proton exchanged polycrystalline and single crystalline samples were prepared by shaking  $NaCrS_2$  in an 1 M HCl 25 vol% water/ethanol solutions for three days. The acid to solid

ratio was 1 mL/mg, and the acid was exchanged daily. After the acid exchange, the sample was washed with an 25 vol% water/ethanol solution and dried under vacuum at 150 °C overnight. Polycrystalline NaCrS<sub>2</sub> powders were treated with the same chemical conditions to extract the Na cations.

### 1.3 Chemical exfoliation of the proton-exchanged polycrystalline samples

The proton-exchanged polycrystalline sample was first shaken in an 400 mM tetramethylammonium hydroxide (TMAOH) solution on a SCILOGEX SK-O330-Pro shaker at the speed of 400 rpm for two weeks. Then, the exfoliation was achieved by replacing TMAOH with 8 mM tetrabutylammonium hydroxide (TBAOH) and continuing shaking at the same speed for at least one week.

#### **1.4** Characterizations and Instrumentation

The morphology of both NaCrS<sub>2</sub> and the proton-exchanged single crystals were studied with a Leica DM2700M optical microscope with white light. The in-plane crystal structure of the pristine NaCrS<sub>2</sub> single crystal as well as the proton-exchanged crystal were measured with an STOE PXRD with Mo  $K_{\alpha 1}$  radiation and a single-Mythen detector, working in Debye-Scherrer geometry. For crystal quality evaluation, in-plane and out of plane diffraction images were collected on a crystal using a Bruker APEX II CCD system equipped with a Mo  $K_{\alpha}$ fine focus tube. The surface morphology and chemical composition of both the pristine and acid treated single crystals were studied with a Verios 460 Extreme High-Resolution Scanning Electron Microscope (XHRSEM) with an Oxford energy dispersive X-ray spectrometer (EDS) and the Titan Cubed Themis 300 double Cs-corrected S/TEM operated at 300 kV with a point resolution of 0.07 nm and an energy resolution of 0.8 eV. The Titan Cubed Themis S/TEM is equiped with a super-X EDS system for elemetal mapping and a Gatan Quantum SE/963 P post-column energy filter for for energy filtered TEM (EFTEM) and electron energy-loss spectra (EELS) data acquisition. The SEM and EDS characterizations were performed on freshly made surfaces by Scotch tape to avoid the potential influences from surface contamination and surface oxidation. To study the cross-section of the compounds, the samples were cut and transferred onto a Cu TEM grid with a Helios NanoLab G3 UC DualBeam FIB/SEM. The Raman measurements were performed on a Horiba Raman Spectrometer with a 532 nm laser source.

The X-ray photoelectron spectroscopy (XPS) data was collected on a ThermoFisher K- $\alpha$  X-Ray Photoelectron Spectrometer. The surface of the sample was cleaned by argon ions to remove the influence of possible surface contamination and oxidation. The data was analyzed by CasaXPS and calibrated so that the C 1s transition occurred at 284.8 eV binding energy. Peaks of the  $2p_{3/2}$  and  $2p_{1/2}$  transitions were fitted simultaneously in 2: 1 intensity ratios with the same peak shape and width. The sample consists of alternating layers of crystalline and amorphous components as well as an oxidized surface layer. The oxide layer was reduced by ion sputtering, but small amounts remained after. While sputtering removed the oxide signal of the surface layer, the variations in thickness of the crystalline and amorphous components in combination with the short sampling depth of XPS impeded the exact determination of elemental ratios. Additionally, NaCrS<sub>2</sub> contains traces of the sodium (poly)sulfide flux (BE 162.4 eV) and H<sub>x</sub>CrS<sub>2</sub> contains a surface layer of oxidized S-O species (BE 166.8-168.6 eV) prior to argon ion sputtering.

The nanosheets were characterized with a Bruker Dimension ICON3 Atomic Force Microscope (AFM) operating in tapping mode and Talos F200X S/TEM. The acquired AFM images were processed with Gwyddion software. For the magnetic susceptibility measurement, several pieces of the NaCrS<sub>2</sub> single crystals were glued together along the c-axis with GE varnish. The proton-exchanged samples were prepared in the same manner. The magnetic susceptibility data were acquired on a Quantum Design DynaCool Physical Property Measurement Setup (PPMS) with a Vibrating Sample Magnetometer (VSM) model. To determine the molar mass of the proton-exchanged phase, we performed a thermogravimetric analysis on the proton-exchanged single crystals under air flow with a ramp rate of 10 °C/min from 50 °C to 700 °C with a PerkinElmer TGA-8000 Thermogravimetric Analyzer (TGA). The sample was dewelled at 700 °C for 30 minutes to allow for full oxidation. The structure of the oxidized product was confirmed with the STOE PXRD.

# 2 In-plane XRD of the pristine $NaCrS_2$ single crystal and the proton-exchanged crystal

Due to the presence of the amorphous phase and the stacking faults in the c direction of the proton-exchanged crystal, it is hard to obtain powder X-ray diffraction (XRD) data of the new phase. Here we show the in-plane XRD pattern (measuring in transmission geometry) of the proton-exchanged crystal in comparison to the pristine NaCrS<sub>2</sub> single crystal's in-plane XRD pattern to show that the in-plane lattice parameters shrink from a = 0.3548 nm to a = 0.3474 nm upon proton exchange as the (110) reflection shifts to higher angles. The intensity of the new phase's XRD pattern decreases due to loss of crystallinity in the system. Thus, the intensity of the proton-exchanged crystal's XRD pattern has been magnified 5 times to better exemplify the peak shift. The reflection indicated by the star is caused by the polymer film that holds the sample.



Figure S1: In-plane XRD of the pristine  $NaCrS_2$  single crystal (black) and the proton-exchanged crystal (blue)

#### 3 XPS spectra of the pristine $NaCrS_2$ and proton-exchanged



#### crystals

Figure S2: (A)-(C) Na 1s, Cr 2p, S 2p spectra of the pristine NaCrS<sub>2</sub> single crystal, (D)-(F) Na 1s, Cr 2p, S 2p spectra of the proton-exchanged crystal

# 4 Measured and calculated Raman spectra for the pristine $NaCrS_2$ and proton-exchanged crystals

Quantum chemical calculations were performed in the framework of Density Functional Theory (DFT) using a linear combination of Gaussian-type functions (LCGTF) scheme as implemented in CRYSTAL17.<sup>S2,S3</sup> Models for the 2D H<sub>x</sub>CrS<sub>2</sub> sheets were constructed from the CrS<sub>2</sub>-partial structure of the parent compound NaCrS<sub>2</sub> with AA stacking of the sheets, without sodium atoms and an increased interlayer distance of more than 13 Å. Different amounts of hydrogen functionalization (HCrS<sub>2</sub>, H<sub>0.5</sub>CrS<sub>2</sub>) were modeled via 2 × 2 supercells. The resultant structural details are listed in Table S1. Grimmes D3 dispersion correction  $^{S4-S6}$  was added to full structural optimizations with the GGA (PBE)  $^{S7}$  xc-functional. The basis sets were taken from the literature.  $^{S8-S10}$  The convergence criterion considering the energy was set to  $1 \cdot 10^{-8}$  a.u. with a *k*-mesh sampling of at least  $4 \times 4$  for the supercells. Vibrational frequencies including Raman intensities were computed on the basis of the relaxed structures using the coupled-perturbed Kohn-Sham (CPKS) mode.  $^{S11,S12}$  The modes were visualized and animated with the J-ICE application.  $^{S13}$ 



Figure S3: (A) Raman spectrum of the NaCrS<sub>2</sub> single crystal (blue) and the theoretical values (red) calculated by DFT. (B) Raman spectrum of the proton-exchanged crystal (blue) and the theoretical values (red) calculated by DFT. The highest measured peak at 537 cm<sup>-1</sup> is attributed to the S-H bending vibration. S-S vibrations in transition metal pyrites or S<sub>8</sub> appear at lower frequencies, between 380 and 490 cm<sup>-1</sup>.<sup>S14</sup>



Figure S4: Visualization of selected calculated Raman active modes of the  $H_{0.5}CrS_2$  model with Raman shift (color code: Cr grey, S yellow, H white, vibrational vectors red)

Model	Space group	lattice parameters	atomic coordinates x y z
$\mathrm{HCrS}_2$	P2/m (no. 10)	$a=18.55~{ m \AA}$	S1: 0.4337 0. 0.3156
		$b=3.51~{ m \AA}$	S2: 0.4319 0.5 -0.1907
		$c=6.13~{ m \AA}$	H: -0.3601 00.3278
		eta=95.05 °	Cr1: 0.5 -0.5 0.5
			Cr2: 0.5 0. 0.
$H_{0.5}CrS_2$	C2/m (no. 12)	$a=12.13~{ m \AA}$	S1: -0.0820 0. 0.4349
		$b=7.09~{ m \AA}$	S2: 0.0838 -0.5 -0.4320
		$c=18.53~{ m \AA}$	S3: 0.1674 0.2508 0.4333
		eta=90.48 °	H: 0.0840 00.3608
			Cr1: 00.2607 0.5
			Cr2: $-0.2516 \ 0. \ -0.4997$

Table S1: Structural details of the models used for the simulation of the Raman spectra



Figure S5: (A) Top view, (B) side view of the  $HCrS_2$  model as obtained from DFT; (C) top view and (D) side view of the  $H_{0.5}CrS_2$  model as obtained from DFT; (color code: Cr grey, S yellow, H blue)

# 5 Additional STEM information of the proton-exchanged phase

In the main text, we use the well-studied  $MoS_2$  as a reference for the two known van der Waals bonded layered TMD structures for octahedrally-coordinated transition metals for the discussion of the stacking order in the new proton-exchanged phase based on the in-plane atomic-resolution HAAFD-STEM studies. These structures are shown below.



Figure S6: (A) The structure of  $3R-MoS_2^{S15}$  viewed from above the planes. (B) The structure of  $3R-MoS_2^{S15}$  viewed parallel to the planes. (C) The structure of  $1T-MoS_2^{S16}$  viewed from above the planes. (D) The structure of  $1T-MoS_2^{S16}$  viewed parallel to the planes.

A pristine as-synthesized NaCrS<sub>2</sub> single crystal sample was also studied with the HAADF-STEM to compare the results to a material with ABC stacking order (Figure S7(A)). In some of the proton-exchanged samples, we observed the coexistence of 2 crystalline phases. A typical image is shown in Figure S7(B). The region highlighted in red maintained the ABC stacking order (shown in Figure S7(C)-(D)), while Cr atoms (indicated by orange spheres) migrated to the Na<sup>+</sup> vacancies (the Na<sup>+</sup> are indicated by cyan spheres in Figure S7(A), (C)). An illustration of the structure after the Cr migration is shown in Figure S7(D). In the region highlighted in blue, the structure rearranged to a different stacking order as discussed is the main text. This provides insight about the phase transformation process. Upon Na<sup>+</sup> removal, some Cr atoms in the  $[CrS_2]^-$  layers migrate to the empty Na<sup>+</sup> position, but the stacking order does not change right away. As a second step, this phase will transform to the structure with ABA type stacking. This likely happens upon de-intercalation of the Na<sup>+</sup>, and drying of the sample in a vacuum oven further separated the amorphous lamellae from the crystalline lamellae. The structure of the ABA stacked phase is isostructural to CrS, but the Cr positions are only partially occupied (see Figure S7(E)).



Figure S7: (A) A HAADF-STEM image of the cross-section of a pristine NaCrS<sub>2</sub> single crystal. The Na, Cr and S atoms are indicated by the cyan, red and yellow spheres, respectively. (B) A HAADF-STEM image of the cross-section of a proton-exchanged crystal with two different structural phases coexisting at the same time. The region highlighted in red maintained the ABC stacking order (shown in Figure 3(C)), while Cr atoms (indicated by orange spheres) migrated to the Na vacancies (the Na cations are indicated by cyan spheres in Figure 3(A)). The region highlighted in blue has transformed to the new ABA stacking order (shown in Figure 3(E)). (C) An illustration of the structure of NaCrS<sub>2</sub>. (D) An illustration of structure with the ABC stacking order after the Cr migration. (E) An illustration of structure with the ABA stacking order after the Cr migration. The bi-colored spheres indicate partial vacancies of Cr, which is based on the measured overall Cr: S ratio of 1: 2. Note that the actual vacancy distribution is not known and just loosely estimated based on STEM image contrast.



Figure S8: (A) A HAADF-STEM image of the cross-section a proton-exchanged sample. (B)-(C) EDS mapping of Cr and S distribution of sample in the Figure S8(A). (D) EDS atomic fraction of the S (blue line) and Cr (red line) distribution along the cross-section (indicated by the green line in the Figure S8(A)) of the proton-exchanged crystal. (E) EDS spectrum of a typical proton-exchanged crystal for the HAADF-STEM studies.

Elements	- ·	Out-of-plane		Out-of-
	In-plane	crystalline	Out-of-plane	plane overall
	overall	phase (at.%)	amorphous	(at.%)
	(at.%)	1 ( )	phase (at.%)	
S		65.1		67.2
	66.4		70.9	
$\operatorname{Cr}$		34.9		32.8
	33.6		29.1	

Table S2: STEM-EDS data of the proton-exchanged phase

In order to further confirm the chemical environment of Cr and S in the proton-exchanged crystal, we performed Electron energy loss spectroscopy (EELS) on single crystalline NaCrS<sub>2</sub> and a proton-exchanged crystal, which contains both, crystalline ( $H_xCrS_{1.9}$ ) and amorphous ( $H_yCrS_{2.4}$ ) phases. Energy loss near edge structures (ELNES) of S and Cr are shown in Figure S9 (A) and (B), respectively. Both crystalline phases of NaCrS<sub>2</sub> and  $H_xCrS_{1.9}$  almost show the same fine structures of S L<sub>2,3</sub>, L<sub>1</sub> and Cr L<sub>2,3</sub> edges. It indicates that S and Cr in the two crystalline phases keep the same electronic structures (or chemical bonding environment) during the chemical treatment. However, in the amorphous phase ( $H_yCrS_{2.4}$ ), the S L edges are a little weaker than those of the crystalline phases and the Cr L edges shift ~ 0.4 eV in comparison to those of the crystalline phases.

In combination with the XPS analysis, it can be concluded that S and Cr in the two crystalline phases keep the same valence states ( $S^{2-}$  and  $Cr^{3+}$ ) while the disordered coordination of atoms in the amorphous phase could change the valence states of S and Cr. Note that hydrogen atoms might still be present at the edges of crystalline phases as drawn in Figure 1, since these would directly reside at the interpose between crystalline and amorphous layer, which makes exact probing challenging.



Figure S9: EELS analysis of single crystalline  $NaCrS_2$  and a proton-exchanged crystal including crystalline ( $H_xCrS_{1.9}$ ) and amorphous ( $H_yCrS_{2.4}$ ) phases. Energy loss near edge structures (ELNES) of S and Cr are shown in (A) and (B), respectively.

#### 6 Electrical resistivity of the proton-exchanged crystal

The temperatures depended resistivity of the proton-exchanged crystal was measured in a standard two-terminal geometry in a Quantum Design PPMS DynaCool system. Figure S10(A) show that the resistivity decreases with temperature, indicative of semiconducting behavior. Below 125 K, the sample became too insulting to obtain a signal. To better understand the conduction mechanism, the resistivity data were analyzed by fitting them to Arrhenius-type model and the Mott variable range hopping (VRH) type model as shown in the Figure S10(B). These two models can be expressed as:  $\rho = \rho_0 exp(\frac{T_0}{T})^{1/(d+1)}$ , where  $\rho$  is the resistivity. In the case of the Arrhenius model, d = 0, and  $T_0$  is the activation energy. This model is usually applicable for a typical semiconductor. For the Mott VRH model, d = 3 in case of three-dimensional conductance, and  $T_0$  represents the characteristic energy for hopping. Here, the transport can be better described by a Mott VRH model as the plot of the logarithm of resistivity against  $T^{-1/4}$  (red) is more linear compared with the plot of the sample, since amorphous and highly disordered samples usually follow the Mott VRH mechanism.



Figure S10: (A) Temperature dependence of the resistivity of a proton-exchanged crystal between 125 K to 300 K. (B) The temperature dependent resistivity plotted according to the Arrhenius (blue) and the Mott variable range hopping (VRH) (red) mechanisms. The Mott VRH model yields a more linear curve, indicating that this is a better model to describe the transport in  $H_xCrS_2$ .

#### 7 Magnetic properties



Figure S11: Temperature dependent magnetic susceptibility and inverse susceptibility of  $NaCrS_2$  (A) and the proton-exchanged crystals (B), where the field is applied perpendicular to the plane. ZFC data is shown in blue and FC in green. The inverse magnetic susceptibility is shown in black. The red line indicates the Curie-Weiss fit. The insets highlight the Néel transition.

#### 7.1 Curie-Weiss fits

The paramagnetic regions (150 K to 300 K) of the inverse temperature dependent magnetic susceptibility of NaCrS<sub>2</sub> and the proton-exchanged crystals were fitted according to the Curie-Weiss law to derive the Weiss temperature and the effective magnetic moment. The Curie-Weiss law is given by  $\chi = C/(T - \Theta)$ , where  $\chi$  is the magnetic susceptibility (unit: emu/mol Oe), T is the temperature (Unit: K),  $\Theta$  is the Weiss temperature (Unit: K) and C is the Curie constant. The magnetic susceptibility is calculated from the measured magnetization data M (emu), as  $\chi$  (emu/mol Oe)= $M(emu) \times \frac{Molar \ mass \ of \ the \ sample \ (mol/g)}{Mass \ of \ the \ sample \ (g) \times Applied \ magnetic \ field \ (Oe)}$ . The effective magnetic moment of the magnetic cations,  $\mu_{eff}$  (Unit:  $\mu_B$ ), can be derived from

the Curie constant by  $\mu_{eff} = 2.827 \times \sqrt{C}$ .

Even though the proton-exchanged crystals we used for magnetic measurement have been dried in a vacuum oven at 150 °C overnight to obtain a more accurate sample mass for the Curie-Weiss fit, it is hard to estimate the molar mass of the new phase due to the possible crystalline water existing in the system. Thus, we performed a thermogravimetric analysis on the dried proton-exchanged crystals under air flow to fully oxidize the samples. The final product was confirmed with PXRD to be  $Cr_2O_3$  (Figure S12(B)). The resulting mass was used to back calculate the amount of Cr.



Figure S12: Thermogravimetric analysis of the proton-exchanged crystals. The analysis was carried out with a ramp rate of 10 °C/min in air flow. The inset shows the derivation of the sample mass versus sample temperature. (B):PXRD of the oxidized products (blue) compared to a simulated  $Cr_2O_3$  XRD pattern<sup>S17</sup> (red) to confirm the sample was completely oxidized to  $Cr_2O_3$ .

In order to gain a more comprehensive understanding of the parent material's magnetic behaviors we supply additional information here. Similarly to previous reports, we observe that NaCrS<sub>2</sub> exhibits a field induced spin-flop transition if the field is aligned within the plane, at around 3 T and below the Néel temperature (Figure S13(A)). This field induced transition does not appear if the field is aligned perpendicular to the plane as shown in the insets of Figure S13(B). The presence of a spin-flop transition within the lattice plane at low temperature suggests that the spins align antiferromagnetically within lattice plane below the transition.  $NaCrS_2$ 's magnetic phase diagrams for both applied field directions are given in the Figure S14.



Figure S13: In-plane magnetic susceptibility at 2 K (blue) and at 20 K (black) of the pristine NaCrS<sub>2</sub> single crystal stack versus applied field, insets show dM/dT of M-H data from 0 T to 9 T at both temperatures. (B): Out-of-plane magnetic susceptibility at 2 K of the same pristine sample versus applied field, inset shows dM/dT of M-H data from 0 T to 9 T at 2 K. (C) and (D) In-plane and out-of-plane magnetic susceptibility at 2 K (blue) of the proton-exchanged crystal stack versus applied field, insets show low-field regions to highlight the weak hysteresis loop.



Figure S14: (A): Magnetic phase diagram of the pristine  $NaCrS_2$  single crystals. The field is applied along the plane. (B): Magnetic phase diagram of the pristine  $NaCrS_2$  single crystals. The field is applied across the planes.

#### 8 Chemical information of the Cr-S based nanosheets



Figure S15: (A) An image of the Cr-S based nanosheets suspension solution. (B)-(C) HAADF-STEM EDS mapping of S and Cr in the sheet shown in Figure 5(D).

We measured the Cr-S based nanosheet suspension's zeta potential with a Zetasizer Nano-ZS (Malvern Instrument, Malvern, UK) to analyze the stability of the exfoliated sheets and to gain information about the nature of their charge. The zeta potential of our dilute nanosheet suspension is  $-54 \pm 5$  mV, suggesting that the sheets are negatively charged and stabilized

in suspension.<sup>S18,S19</sup> In order to estimate the yield of the nanosheet, we used a PerkinElmer TGA-8000 Thermogravimetric Analyzer (TGA) to heat a known amount of concentrated nanosheet suspension to 700 °C to evaporate solvents, burn out the alkalyammonium cations and fully oxidize the nanosheets to  $Cr_2O_3$ . The measured mass of  $Cr_2O_3$  can then be used to calculate the nanosheet yield. However, the final mass was too low to give an accurate measurement - the error bar for the final mass was above 100%. Based on the noisy data, we estimate that the exfoliation yield is roughly 5 wt.%. Yields this low are common for soft chemical exfoliation methods.<sup>S20</sup>

#### 9 Electrochemical Lithium intercalation



Figure S16: Cyclovoltamogram of  $\rm H_xCrS_2$  at a sweep rate of 0.1 mV/s. WE:H\_xCrS\_2-C, CE: Li, electrolyte LiPF\_6 in EC/DMC 50:50

2D-layered transition metal chalcagonides are of major interest as active cathode materials in Lithium-Ion Batteries (LIB). The weak van der Waals interactions between their layers promises easy (low over-potentials) and reversible intercalation of lithium into the structure. In order to describe the capability of  $H_xCrS_2$  to reversibly intercalate lithium, we build a 2-probe battery cell using 2023 stainless steel coin cells. First, a composite working electrode was prepared by combining  $H_xCrS_2$  as the electrochemical active ingredient, carbon black as conductive additive and PVDF as binder in a ratio of 8:1:1. The dry powders were combined with N-Methyl-2-pyrrolidone (NMP) to form a highly viscous slurry. By using

a doctor blade, an even 30  $\mu$ m thick film was cast on a 0.1 mm thick copper foil. The electrode was dried over night in a vacuum oven and punched into 7/16 inch disks. The mass loading was approximately 1 mg. Coin cells were produced by using the  $H_xCrS_2$  electrode as working electrode (WE), a porous polymer Separator (Celgard 3220) and a lithium metal disc as counter electrode (CE), which also served as the reference electrode (RE). A 1M solution of  $LiPF_6$  in mixed solvent of Ethylene Carbonate (EC) and Dimethyl Carbonate (DMC) in a 1:1 ratio, was used as electrolyte. The cells were let to rest for 48 hours before cyclovoltamograms (CV) were recorded. All electrochemical measurements were performed on an Arbin BT2000 potentiostat. All Potentials are reported against Li/Li<sup>+</sup>. The CV in Figure S16 was recorded with a sweep rate of 0.2 mV/s. The measurement started at an open circuit voltage of 2.53 V and was first sweeped in the cathodic direction. At 0.6 V a faradaic peak is observed, signaling a reduction on the working electrode, presumably the reduction of  $H_xCrS_2$  to  $H_xLi_vCrS_2$ . While changing the sweep direction a significant non faradaic current increase was observed, which was attributed to a high capacitance of the cell. Further increase of the potential leads to an oxidation peak at 1.93 V where  $H_x Li_v CrS_2$ is presumably delithiated.

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