

Supplementary information

Temperature dependent Raman study of antiferromagnetic CrPS₄

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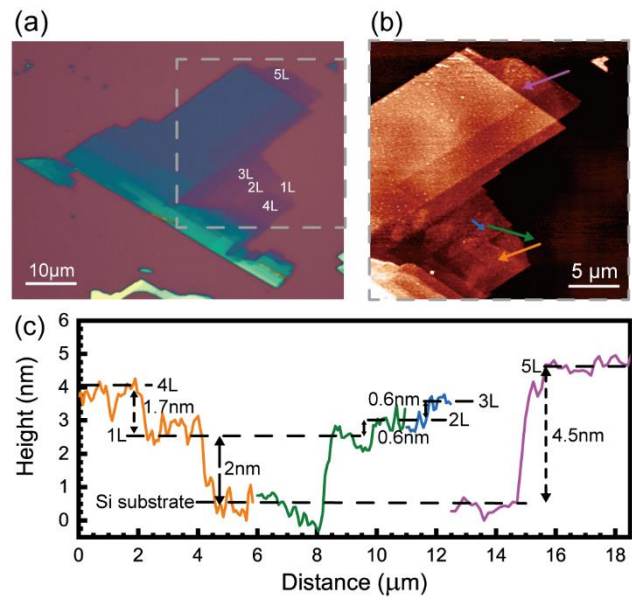


Figure S1. (a) Optical image and (b) AFM image of a few-layer CrPS₄ sample. (c) Height profiles measured along the lines indicated in (b).

Table S1. Comparison of Raman peaks measured at 293 K with calculated phonon modes of bulk CrPS₄.

Our work		Theoretical work by S. N. Neal <i>et al.</i> ¹	
Peak label	Frequency (cm ⁻¹)	Frequency (cm ⁻¹)	Vibrational motion
B1	86.0	78.3	Out-of-plane Cr translation + Out-of-plane S displacement
B2	116.0	106.1	P-S rocking motion
A1	119.0	107.7	In-plane Cr translation + S shearing
B3	156.9	142.0	Out-of-plane Cr translation + Out-of-plane S displacement
A2	170.7	162.9	In-plane P-S translation
A3	179.9	172.9	Out-of-plane P-S motion
A4	217.3	207.8	
B4	233.2	225.3	In-plane Cr translation + P translation + Out-of-plane S motion
B5	258.3	249.8	In-plane S shearing
A5	269.4	259.2	In-plane Cr translation + P translation+ In-plane and Out-of-plane motion
B6	300.1	294.3	Cr motion + S In-plane counter motion
A6	307.5	296.2	In-plane Cr translation + S scissoring
A7	325.0	310.9	In-plane Cr translation + In-plane S shearing + Out-of-plane P translation
B7	350.3	342.9	Out-of-plane Cr translation + S In-plane motion

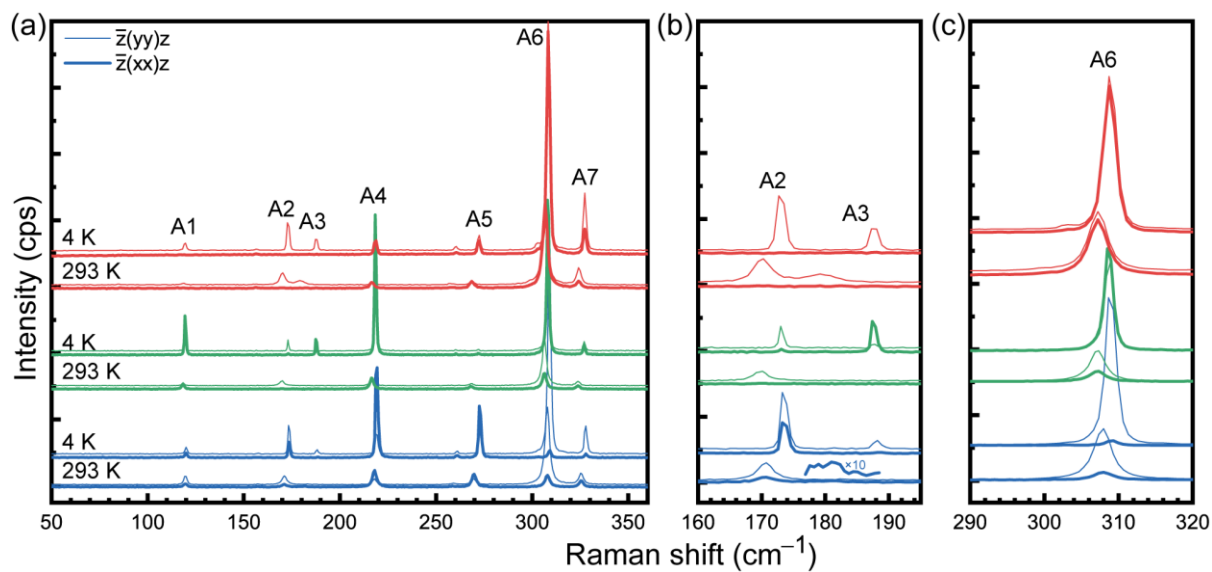


Figure S2. Raman spectra of bulk CrPS₄ with three different excitations: 1.96-eV (red), 2.41-eV (green) and 2.71-eV (blue) excitations in $[\bar{z}(xx)z]$ (thick lines) and $[\bar{z}(yy)z]$ (thin lines) polarization configurations at 293 and 4 K.

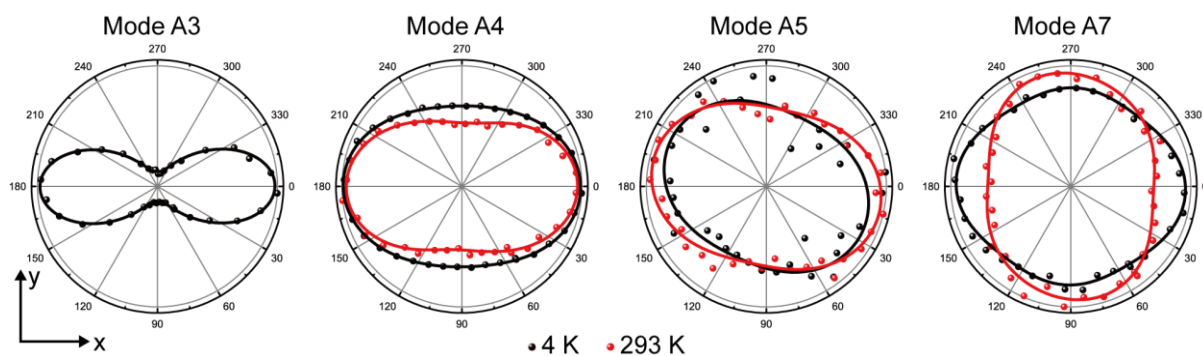


Figure S3. Polarization dependences of A modes in CrPS₄ at 4 K (black) and 293 K (red). The dots and lines are experimental data and fitting, respectively. These data were collected with the 2.41-eV excitation.

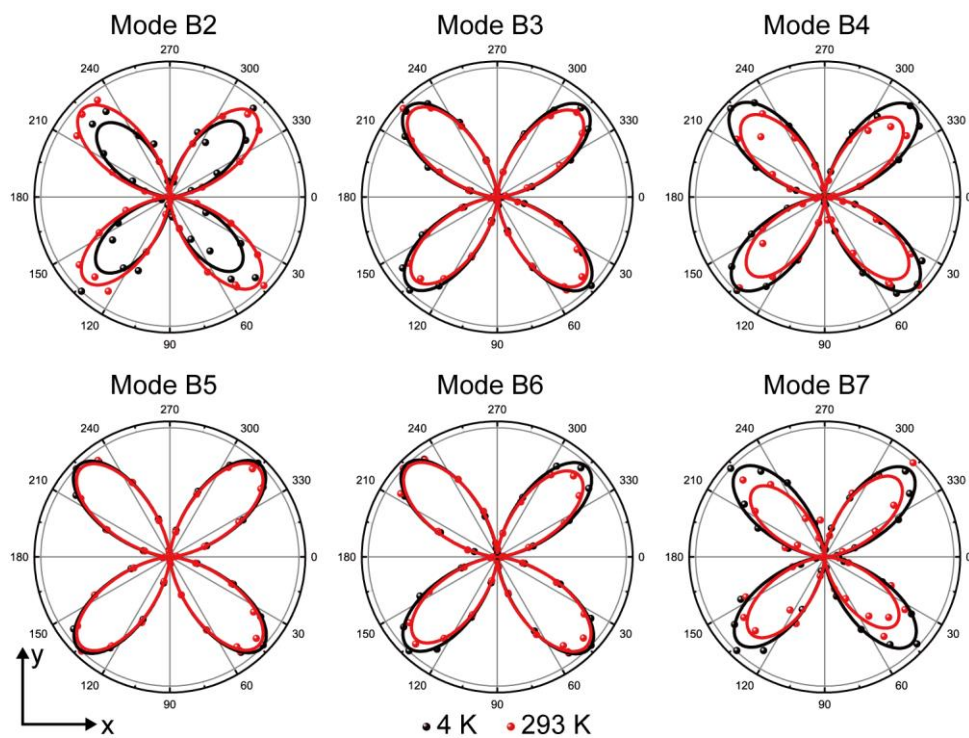


Figure S4. Polarization dependences of the B modes in CrPS₄ at 4 K (black) and 293 K (red). The dots and lines are experimental data and fitting, respectively. These data were collected with the 2.41-eV excitation.

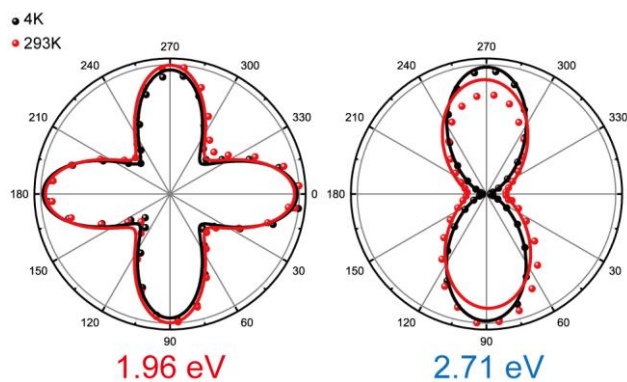


Figure S5. Polarization dependences of the A6 mode of a bulk CrPS₄ sample at 4 K (black) and 293 K (red) measured with 1.96-eV and 2.71-eV excitations, respectively. The dots and lines are experimental data and fitting, respectively.

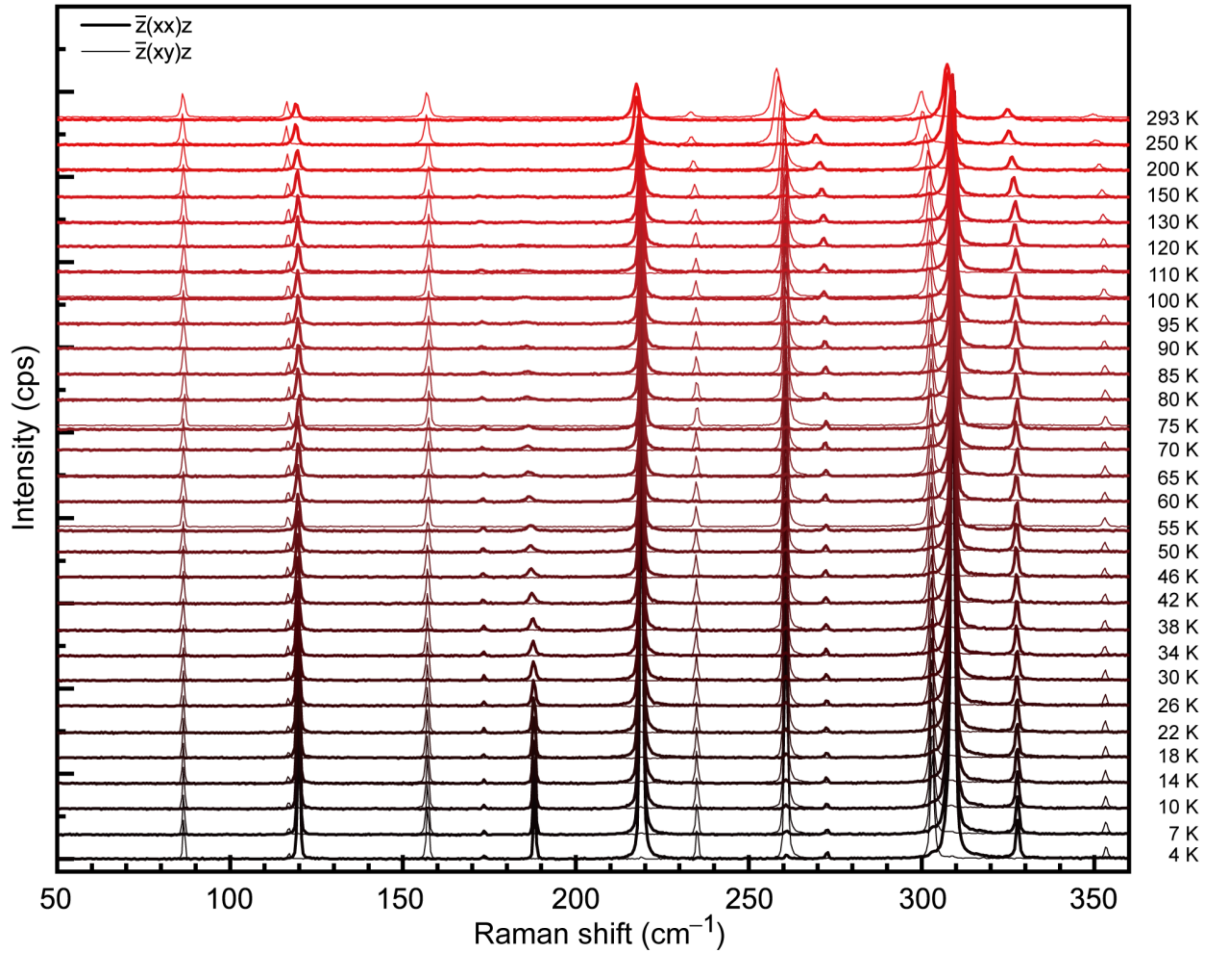


Figure S6. Temperature dependence of Raman spectra of a bulk sample, measured with the 2.41-eV excitation in the parallel [$\bar{z}(xx)z$] (thick lines) and cross [$\bar{z}(xy)z$] (thin lines) polarization configurations.

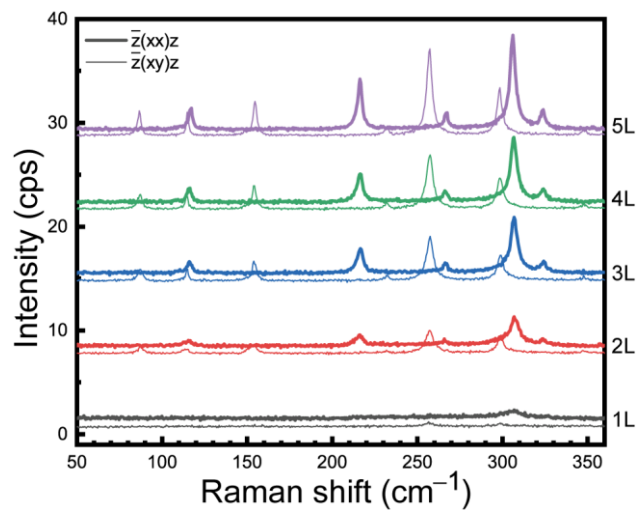


Figure S7. Raman spectra of few-layer CrPS₄ samples at 293 K, measured with the 2.41 eV-excitation in the parallel [$\bar{z}(xx)z$] (thick lines) and cross [$\bar{z}(xy)z$] (thin lines) polarization configurations.

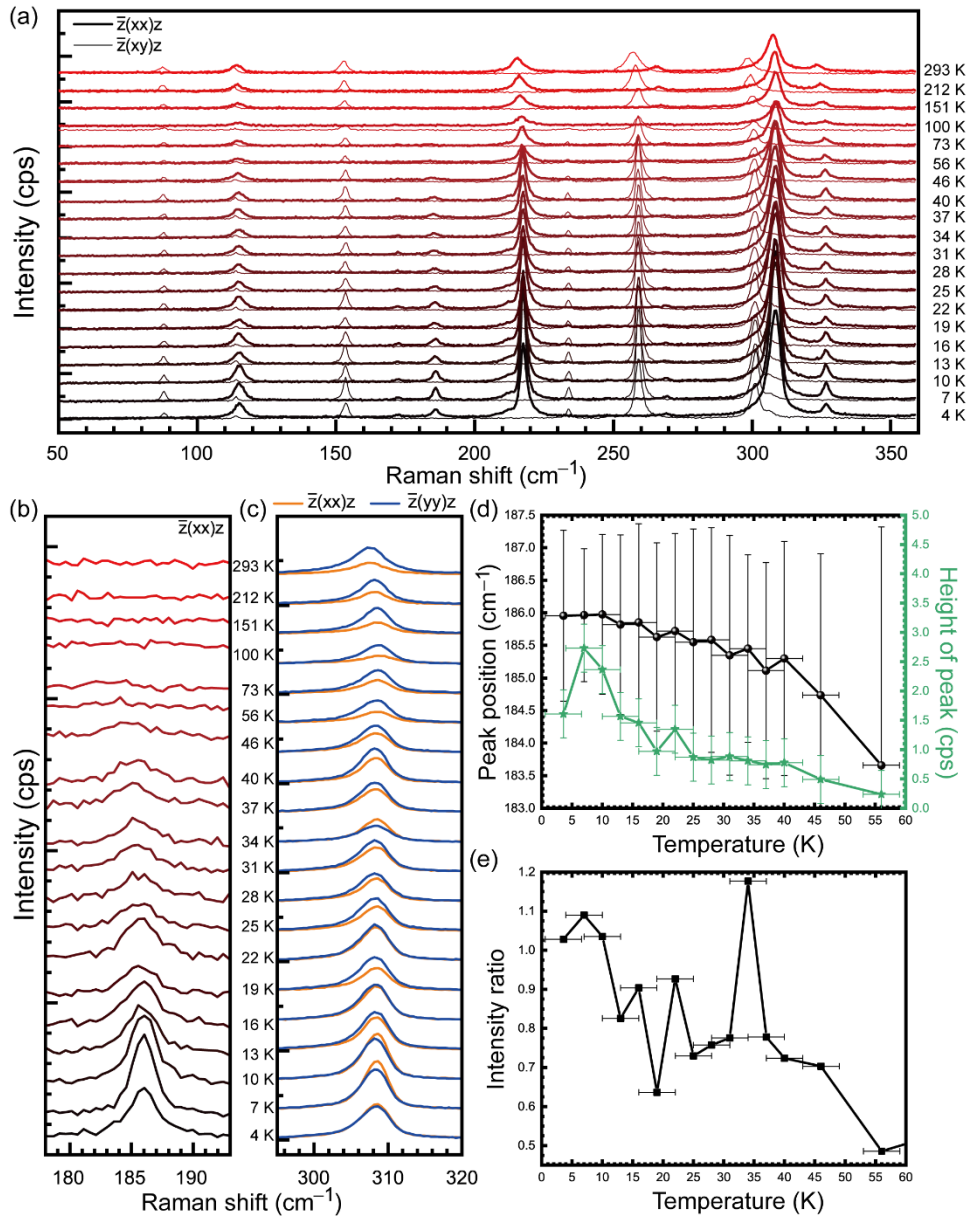


Figure S8. (a) Temperature dependence of Raman spectra of a 2L CrPS₄ sample in the parallel $[\bar{z}(xx)z]$ (thick lines) and cross $[\bar{z}(xy)z]$ (thin lines) polarization configurations. (b) Temperature dependence of the A₃ mode of a 2L sample. (c) Temperature dependence of the A₆ mode of a 2L sample for two different polarizations of $[\bar{z}(xx)z]$ (orange) and $[\bar{z}(yy)z]$ (blue). The Raman spectra were recorded with the 2.41-eV excitation. (d) Temperature dependence of the peak position (black) and the height (green) extracted from (b). (e) Temperature dependence of the intensity ratio of the two polarizations in (c). The error bars indicate the experimental uncertainties.

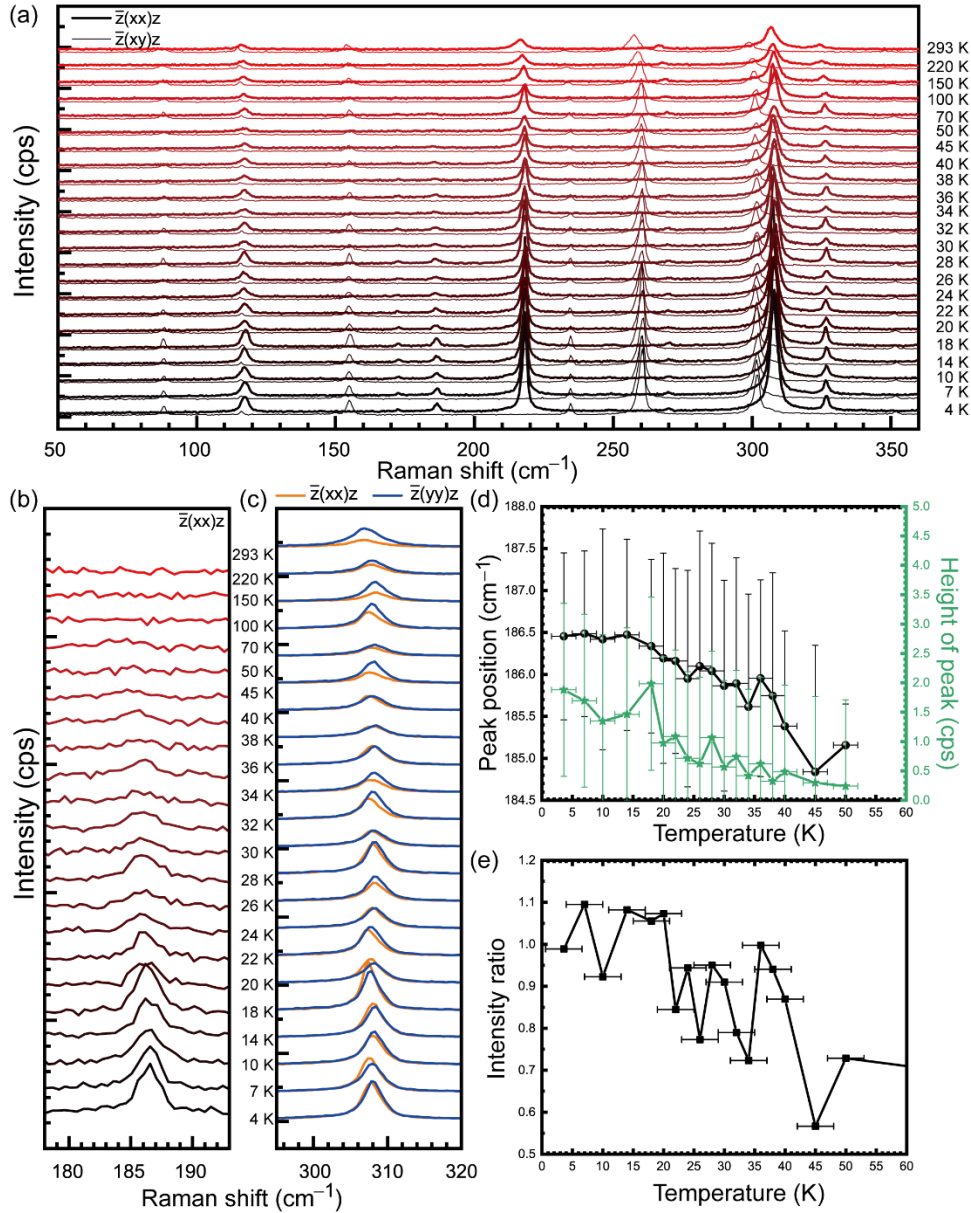


Figure S9. (a) Temperature dependence of Raman spectra of a 3L CrPS₄ sample in the parallel [$\bar{z}(xx)z$] (thick lines) and cross [$\bar{z}(xy)z$] (thin lines) polarization configurations. (b) Temperature dependence of the A3 mode of a 3L sample. (c) Temperature dependence of the A6 mode of a 3L sample for two different polarizations of [$\bar{z}(xx)z$] (orange) and [$\bar{z}(yy)z$] (blue). The Raman spectra were recorded with the 2.41-eV excitation. (d) Temperature dependence of the peak position (black) and the height (green) extracted from (b). (e) Temperature dependence of the intensity ratio of the two polarizations in (c). The error bars indicate the experimental uncertainties.

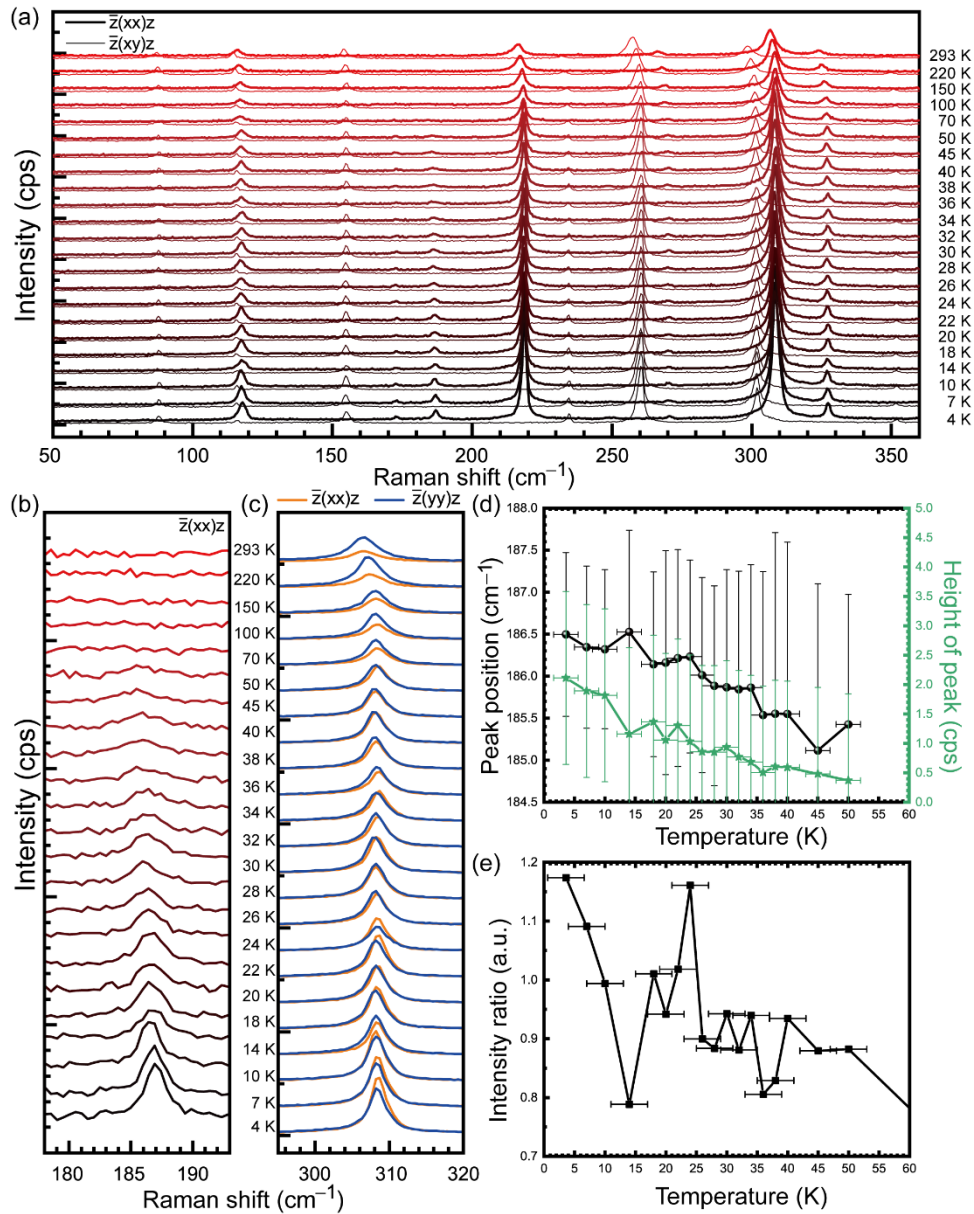


Figure S10. (a) Temperature dependence of Raman spectra of a 4L CrPS₄ sample in the parallel [$\bar{z}(xx)z$] (thick lines) and cross [$\bar{z}(xy)z$] (thin lines) polarization configurations. (b) Temperature dependence of the A3 mode of a 4L sample. (c) Temperature dependence of the A6 mode of a 4L sample for two different polarizations of [$\bar{z}(xx)z$] (orange) and [$\bar{z}(yy)z$] (blue). The Raman spectra were recorded with the 2.41-eV excitation. (d) Temperature dependence of the peak position (black) and the height (green) extracted from (b). (e) Temperature dependence of the intensity ratio of the two polarizations in (c). The error bars indicate the experimental uncertainties.

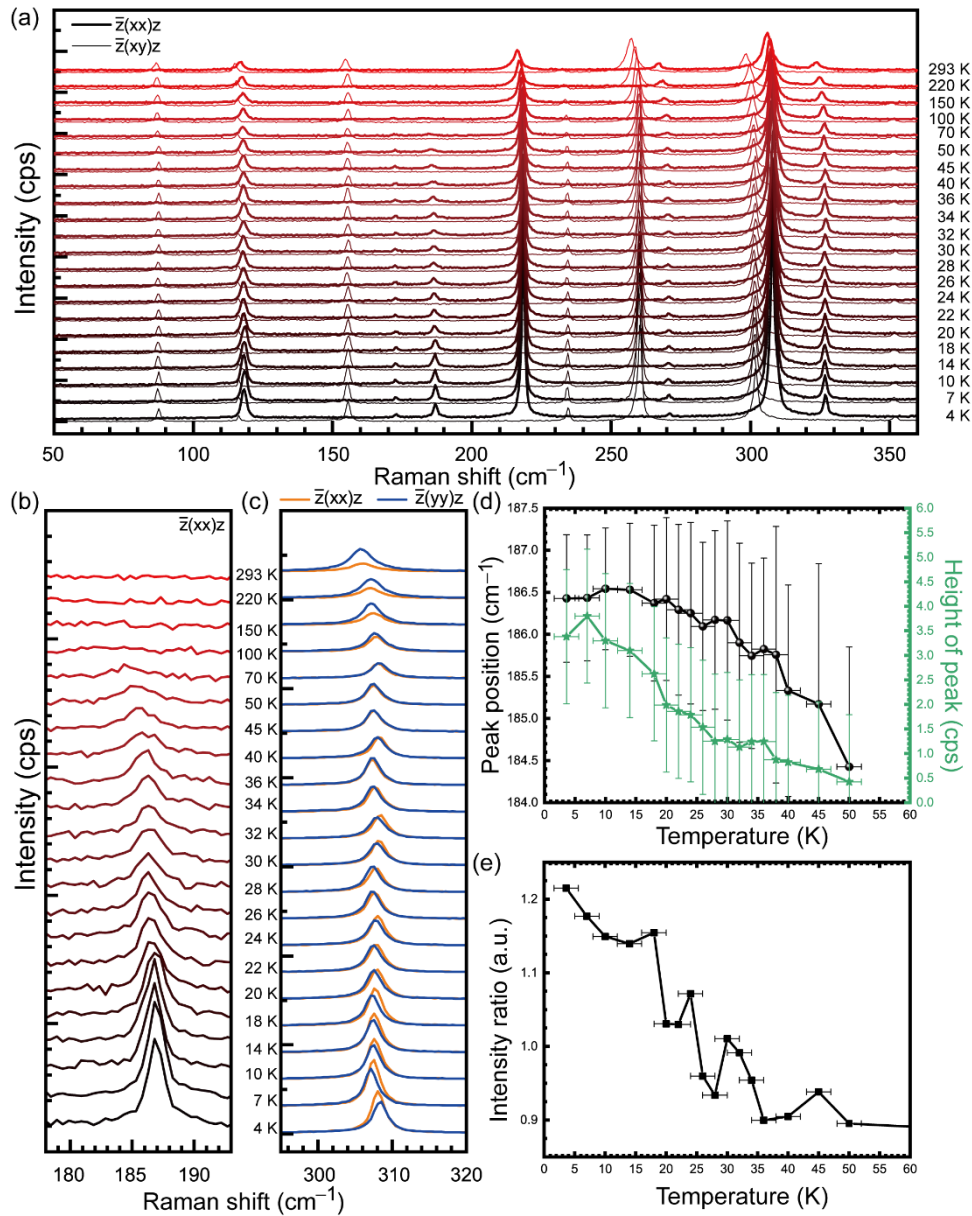


Figure S11. (a) Temperature dependence of Raman spectra of a 5L CrPS₄ sample in the parallel [$\bar{z}(xx)z$] (thick lines) and cross [$\bar{z}(xy)z$] (thin lines) polarization configurations. (b) Temperature dependence of the A₃ mode of a 5L sample. (c) Temperature dependence of the A₆ mode of a 5L sample for two different polarizations of [$\bar{z}(xx)z$] (orange) and [$\bar{z}(yy)z$] (blue). The Raman spectra were recorded with the 2.41-eV excitation. (d) Temperature dependence of the peak position (black) and the height (green) extracted from (b). (e) Temperature dependence of the intensity ratio of the two polarizations in (c). The error bars indicate the experimental uncertainties.

Table S2. Raman peak frequency (cm^{-1}) of few-layer CrPS4 samples that were recorded with 2.41-eV excitation at 293 K in our work and comparison with a previous report.

Our work					Work by Lee <i>et al.</i> ²				
Peak label	2L	3L	4L	5L	Peak label	2L	3L	4L	5L
B1	87.3	87.3	87.0	86.6	--	--	--	--	--
B2	113.7	114.7	114.5	114.9	--	--	--	--	--
A1	115.1	116.1	116.0	116.7	A	112.7	114.7	115.6	116.7
B3	153.4	154.1	154.1	154.6	B	151.0	153.3	154.5	155.7
A2	169.8	169.8	169.7	169.7	C	171.6	169.4	169.7	170.3
A3	--	--	--	--	--	--	--	--	--
A4	216.0	216.4	216.3	216.2	D	215.4	215.6	215.9	216.4
B4	231.9	232.2	232.0	232.1	E	230.2	231.6	232.1	232.3
B5	257.2	257.4	257.3	257.1	F	256.9	256.7	256.9	257.4
A5	265.9	266.5	266.3	266.9	G	263.5	265.6	266.6	267.6
B6	298.9	298.9	298.6	298.3	H	297.9	297.7	298.3	298.9
A6	307.0	307.0	306.5	305.8	I	306.5	306.0	306.1	306.7
A7	324.0	324.0	323.6	323.5	J	323.1	323.2	323.4	323.8
B7	347.4	347.8	347.8	347.9	K	346.7	347.4	347.8	348.6

Table S3. Raman peak linewidth (cm^{-1}) of few-layer CrPS₄ samples that were recorded with 2.41-eV excitation at 293 K in our work and comparison with a previous report.

Our work					Work by J.Lee <i>et al.</i> ²				
Peak label	2L	3L	4L	5L	Peak label	2L	3L	4L	5L
B1	3.5	3.4	3.1	2.3	--	--	--	--	--
B2	4.5	1.7	1.6	1.4	--	--	--	--	--
A1	6.8	3.3	3.0	2.6	A	6.5	4.9	5.6	6.1
B3	5.5	2.9	2.8	2.6	B	7.0	4.7	4.5	3.8
A2	2.9	3.1	3.2	2.8	C	8.2	5.2	5.0	4.6
A3	--	--	--	--	--	--	--	--	--
A4	5.6	4.3	4.1	3.3	D	14.7	5.7	5.1	5.2
B4	2.1	2.7	2.3	2.3	E	7.3	5.3	5.8	5.2
B5	5.6	4.8	4.7	3.9	F	10.8	6.3	5.7	5.3
A5	2.9	2.9	3.1	2.5	G	3.5	3.9	4.4	4.2
B6	5.0	4.4	4.5	3.6	H	9.9	6.7	5.9	5.0
A6	4.8	4.9	4.9	3.9	I	9.1	6.3	5.6	5.0
A7	2.9	2.8	3.1	2.8	J	4.7	4.5	5.3	3.7
B7	1.8	0.9	3.1	3.6	K	3.9	3.0	4.2	5.1

References

- 1 S. N. Neal, K. R. O’Neal, A. V Haglund, D. G. Mandrus, H. A. Bechtel, G. L. Carr, K. Haule, D. Vanderbilt, H.-S. Kim and J. L. Musfeldt, *2D Mater.*, 2021, **8**, 035020.
- 2 J. Lee, T. Y. Ko, J. H. Kim, H. Bark, B. Kang, S.-G. Jung, T. Park, Z. Lee, S. Ryu and C. Lee, *ACS Nano*, 2017, **11**, 10935–10944.