

Supplementary Materials

Symmetry-Engineering-Induced In-Plane Polarization Enhancement in Ta₂NiS₅/CrOCl van der Waals Heterostructure

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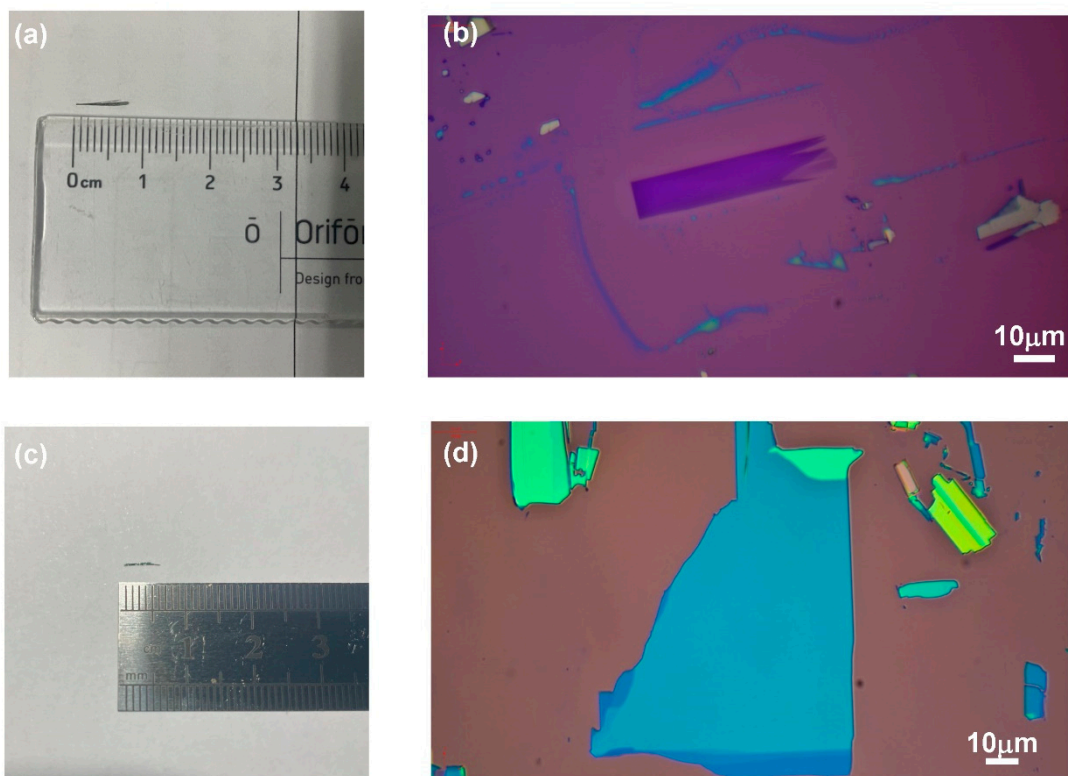


Figure S1. Photograph of bulk (a) Ta_2NiS_5 and (c) CrOCl crystal. Optical microscopy image of few-layer (b) Ta_2NiS_5 and (d) CrOCl flake. The scale bar is $10\mu\text{m}$.

Table S1. Comparison of Raman frequencies of Ta_2NiS_5 and $\text{Ta}_2\text{NiS}_5/\text{CrOCl}$.

	$\text{B}_{2g,a\text{-axis}}$	${}^2\text{A}_{g,a\text{-axis}}$	${}^3\text{A}_{g,a\text{-axis}}$	$\text{B}_{2g,c\text{-axis}}$	${}^2\text{A}_{g,c\text{-axis}}$	${}^3\text{A}_{g,c\text{-axis}}$
Ta_2NiS_5	61.6cm^{-1}	123.7cm^{-1}	146.1cm^{-1}	60.7cm^{-1}	123.7cm^{-1}	146cm^{-1}
$\text{Ta}_2\text{NiS}_5/\text{CrOCl}$	61.2cm^{-1}	124.1cm^{-1}	146.5cm^{-1}	60.9cm^{-1}	123.9cm^{-1}	146.4cm^{-1}
Frequency shift	-0.4cm^{-1}	0.4cm^{-1}	0.4cm^{-1}	0.2cm^{-1}	0.2cm^{-1}	0.4cm^{-1}

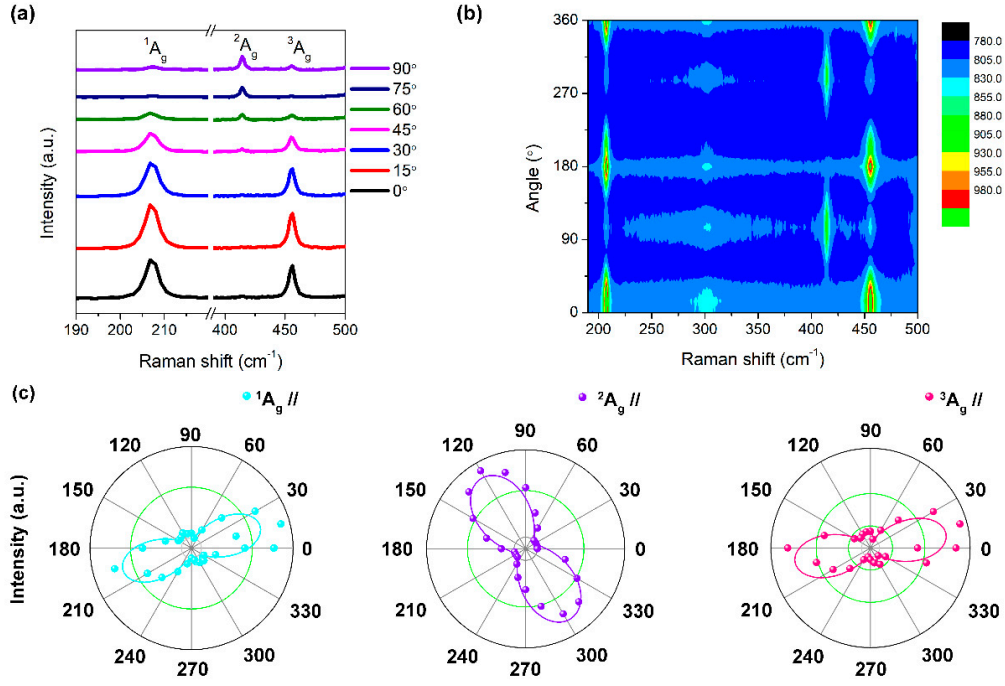


Figure S2. In parallel configurations, (a) polarized Raman spectra, (b) corresponding contour map and (c) polar plots of CrOCl flake.

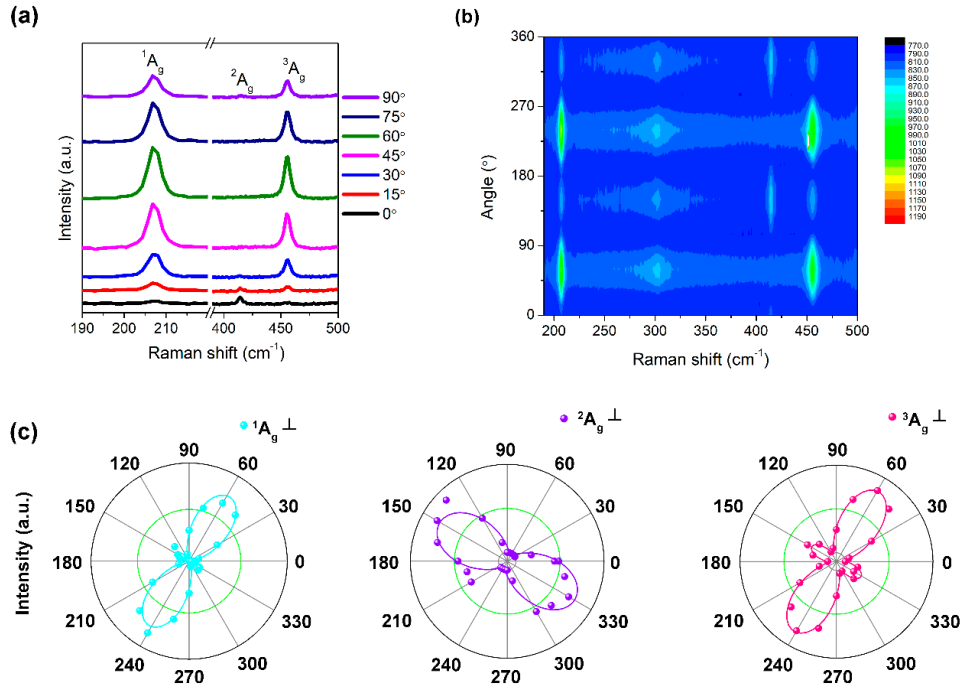


Figure S3. In perpendicular configurations, (a) polarized Raman spectra, (b) corresponding contour map and (c) polar plots of CrOCl flake.

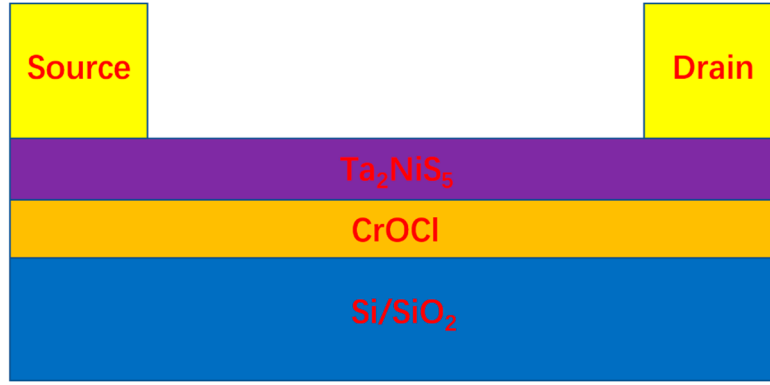


Figure S4. Schematic diagram of Ta₂NiS₅/CrOCl device in side view.

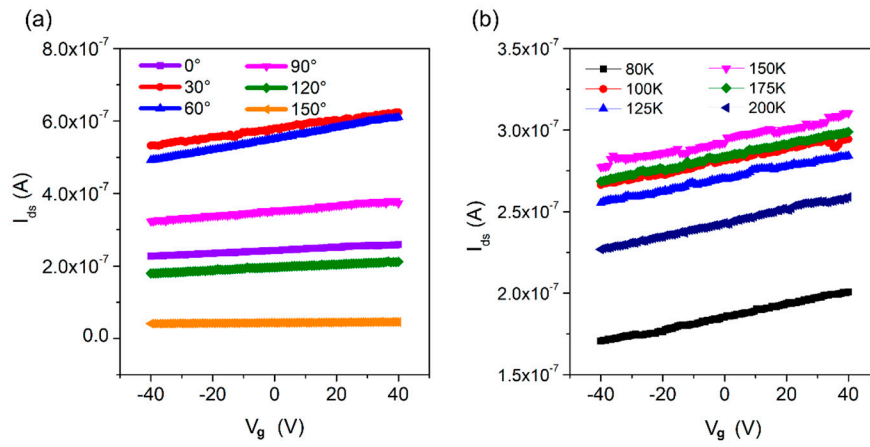


Figure S5. (a) Transfer characteristic curves of Ta₂NiS₅/CrOCl along *a*-axis at different temperatures. (b) Transfer characteristic curves of Ta₂NiS₅/CrOCl along different angles at 200K.

Table S2. Comparison of the anisotropy ratio of Ta₂NiS₅ and Ta₂NiS₅/CrOCl heterostructure.

Materials	σ_a/σ_c	μ_a/μ_c	Reference
Ta ₂ NiS ₅	1.78	/	[1]
Ta ₂ NiS ₅	2.1	2.7	This work
Ta ₂ NiS ₅ /CrOCl	15	32	This work

References:

1. Li, L.; Gong, P.; Wang, W.; Deng, B.; Pi, L.; Yu, J.; Zhou, X.; Shi, X.; Li, H.; Zhai, T. Strong In-Plane Anisotropies of Optical and Electrical Response in Layered Dimetal Chalcogenide. *ACS Nano* **2017**, *11*, 10264-10272, doi:10.1021/acsnano.7b04860.