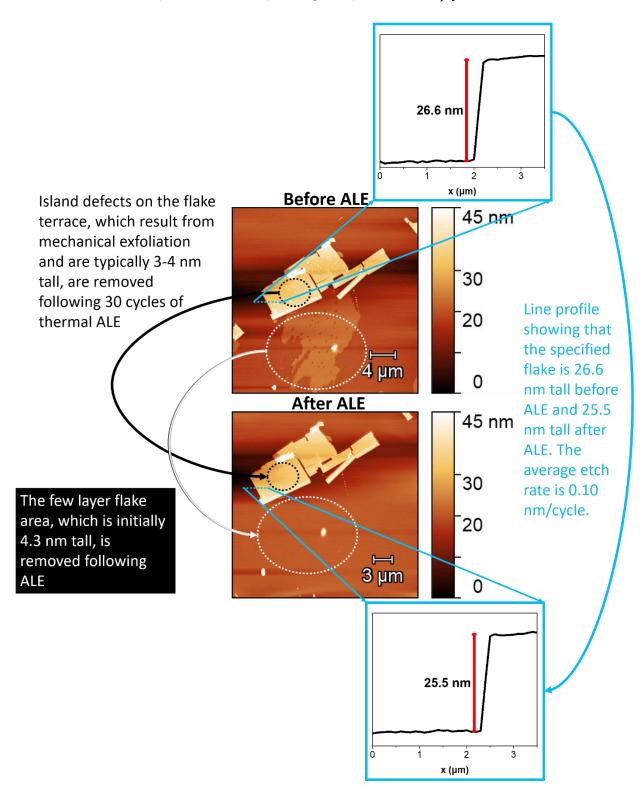
Title: Designing an ALE process and uncovering the etching mechanism for a 2D van der Waals material: Ternary transition metal chalcogenide CrPS<sub>4</sub>

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**Figure 1.** AFM images of a CrPS<sub>4</sub> flake  $\sim$ 25 nm thick and  $\sim$ 20 um wide near a 4 nm tall few-layer flake (circled in white) before and after ALE. The few-layer area is completely or almost completely removed after 30 ALE cycles. The island defects (circled in black) atop the  $\sim$ 25 nm thick flake surface are also removed following ALE.