

Electronic Supplementary Material (ESI) for Materials Chemistry Frontiers.

## Raman Spectroscopy and Carrier Scattering in 2D Tungsten

### Disulfides with Vanadium Doping

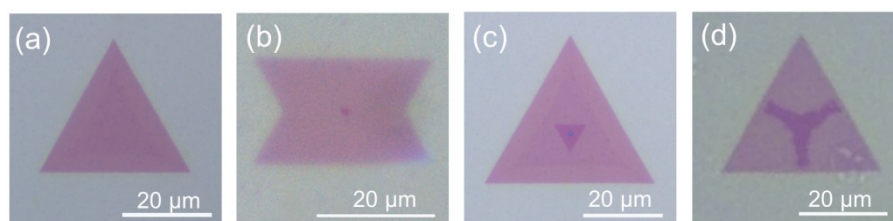
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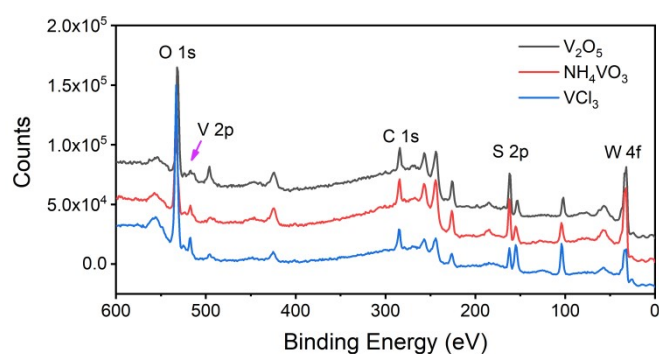
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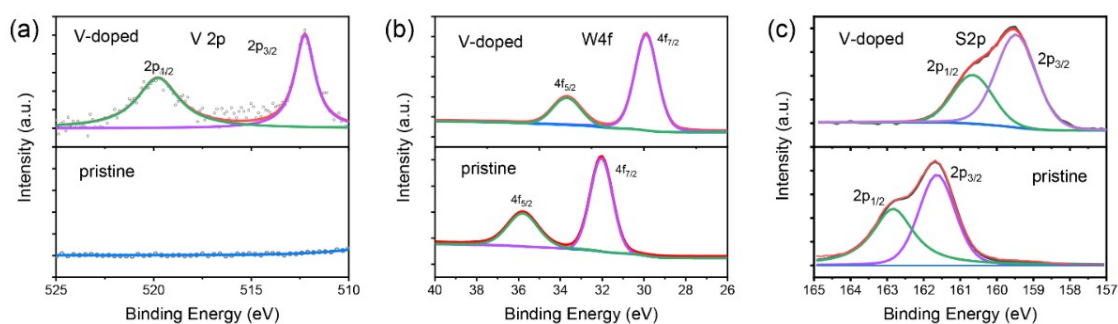
**Figure S1.** Optical microscope images of the as-grown V-WS<sub>2</sub> flakes with different morphologies. (a) A triangle monolayer, (b) a twinned monolayer V-WS<sub>2</sub>, (c) a triangle bilayer, and (d) a windmill-like bilayer flake, respectively. We find that V-WS<sub>2</sub> flakes with different morphologies and layer numbers can be grown by varying the growth parameters, including temperature and additive amount of the V precursors.



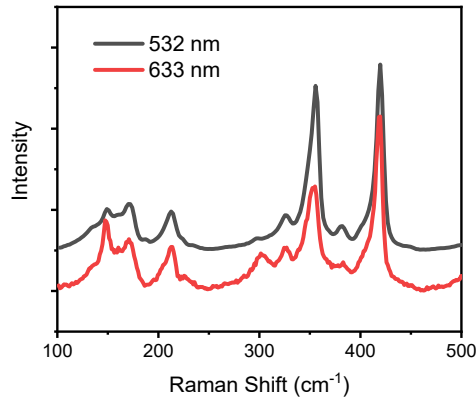
**Figure S2.** Survey XPS spectra of V-WS<sub>2</sub> flakes doped with V<sub>2</sub>O<sub>5</sub>, NH<sub>4</sub>VO<sub>3</sub>, and VCl<sub>3</sub>, respectively.

**Table S1.** Atomic concentration obtained from the XPS characterization in the V-WS<sub>2</sub> flakes

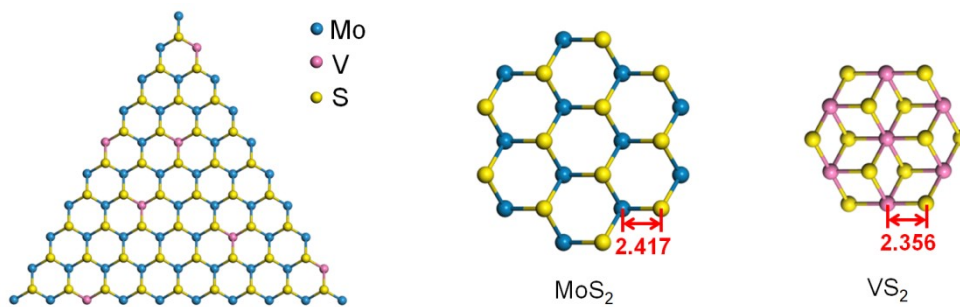
Dopants	W	S	V
V <sub>2</sub> O <sub>5</sub>	32.3 at%	65.5 at%	2.2 at%
NH <sub>4</sub> VO <sub>3</sub>	31.1 at%	62.9 at%	6.0 at%
VCl <sub>3</sub>	29.3 at%	59.6 at%	11.1 at%



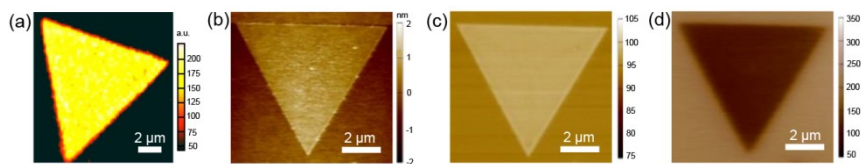
**Figure S3.** XPS analysis of V-WS<sub>2</sub> doped by VCl<sub>3</sub>. (a-c) XPS spectra of the V 2p, W 4f, and S 2p orbitals, respectively. XPS signal of vanadium is observed, confirming the existence of substituted V atoms in the V-WS<sub>2</sub> flake. Meanwhile, both the XPS peaks of W 4f and S 2p orbitals shift toward the low binding energy direction, showing the strong *p*-type doping effect.



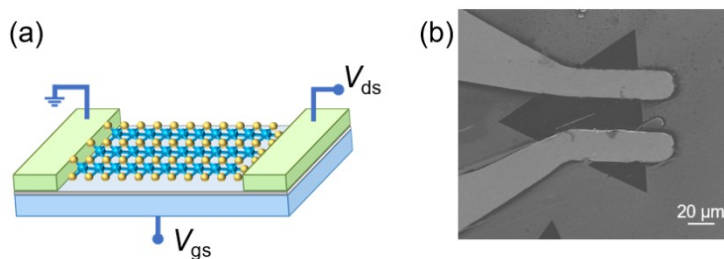
**Figure S4.** Raman spectra of the V-WS<sub>2</sub> excited by the 532-nm and 633-nm lasers. The two spectra were normalized according to the  $A_1'(I)$  peak. Obviously, though the intensity of  $E'(I)$  and  $ZA(M)$  peaks varies with the changed excitation lasers, both the peak position and intensity of  $P_c$  nearly do not change, revealing that this Raman peak has no dependence with the wavelength of the excitation lasers.



**Figure S5.** Schematics of the atomic crystal structure of (a) V-doped WS<sub>2</sub>, (b) WS<sub>2</sub>, and (c) VS<sub>2</sub>. The substituted vanadium atoms are uniformly distributed throughout the whole flake. The bond length of VS<sub>2</sub> is 2.356 Å, small than that of MoS<sub>2</sub> (2.417 Å). So, the substituted V atoms will stretch the surrounding Mo-S bonds. A tensile strain is generated in the V-WS<sub>2</sub>, which causes the shift of the Raman  $E_{2g}$  peak to the low wavenumber direction.



**Figure S6.** (a) Raman mapping image acquired at  $214\text{ cm}^{-1}$ , in which the uniform color profile demonstrates the uniform doping effect. (b) AFM topography image of a monolayer  $\text{V-WS}_2$  flake doped with  $\text{NH}_4\text{VO}_3$  taken at room temperature, and the corresponding (c) EFM and (d) SKPM images.



**Figure S7.** (a) Schematic and (b) SEM image of the back-gated FET with a monolayer  $\text{V-WS}_2$  flake as the channel material. The width ( $W$ ) and length ( $L$ ) of the channel are  $11.6\text{ }\mu\text{m}$  and  $36.9\text{ }\mu\text{m}$ , respectively, yielding an  $L/W$  ratio of 3.18.