

**Electronic Supplementary Information**

**Strong electron-phonon coupling driven charge density wave  
states in stoichiometric 1T-VS<sub>2</sub> crystals**

*Si-Hong Lee<sup>1</sup>, Yun Chang Park<sup>2</sup>, Jinwoong Chae<sup>3,4</sup>, Gunn Kim<sup>3,4</sup>, Hyuk Jin Kim<sup>5</sup>, Byoung Ki Choi<sup>5,6</sup>, In Hak Lee<sup>5,7</sup>, Young Jun Chang<sup>5,8</sup>, Seung-Hyun Chun<sup>3</sup>, Minkyung Jung<sup>9</sup>, Jungpil Seo<sup>1\*</sup>, Sunghun Lee<sup>3\*</sup>*

<sup>1</sup>Department of Physics and Chemistry, DGIST, Daegu 42988, Korea.

<sup>2</sup>Department of Measurement and Analysis, National Nanofab Center, Daejeon 34141, Korea.

<sup>3</sup>Department of Physics and Astronomy, Sejong University, Seoul 05006, Korea.

<sup>4</sup>Hybrid Materials Research Center, Sejong University, Seoul 05006, Korea.

<sup>5</sup>Department of Physics, University of Seoul, Seoul 02504, Korea.

<sup>6</sup>Advanced Light Source (ALS), E. O. Lawrence Berkeley National Laboratory, Berkeley, CA, United States.

<sup>7</sup>Center for Spintronics, Korea Institute of Science and Technology, Seoul 02792, Korea.

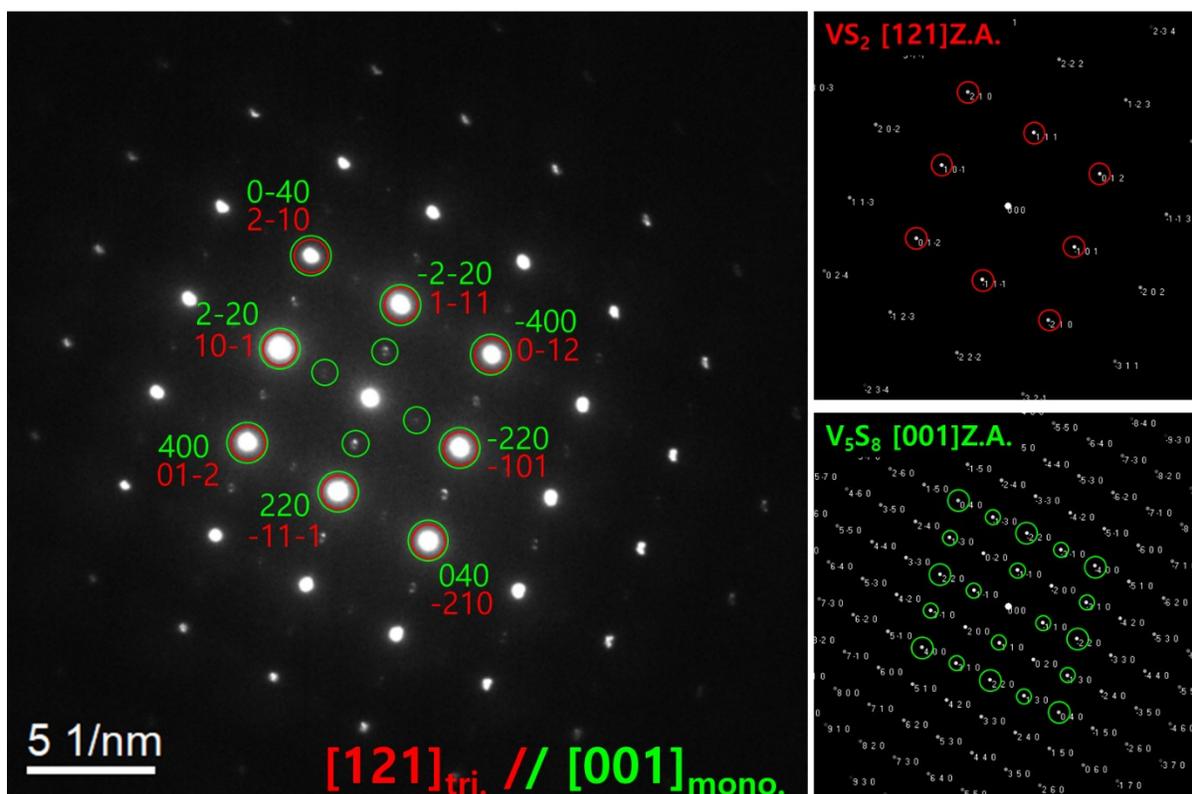
<sup>8</sup>Department of Smart Cities, University of Seoul, Seoul 02504, Korea.

<sup>9</sup>DGIST Research Institute, DGIST, Daegu 42988, Korea.

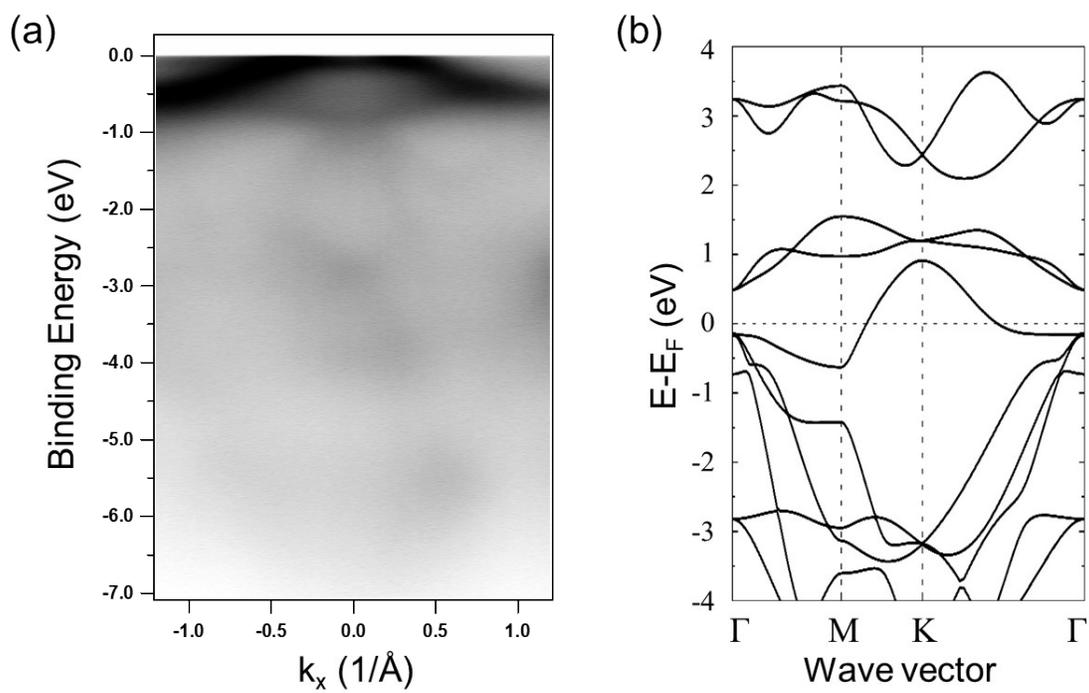
\*Corresponding authors email; jseo@dgist.ac.kr (J.S). kshlee@sejong.ac.kr (S.L.)

Element	line type	k factor	absorption correction	wt%	wt% sigma	atomic %
V	K series	1.1350	1.00	41.04	0.11	32.44
S	K series	0.9804	1.00	58.96	0.18	67.56
<b>Total:</b>				100.00		100.00

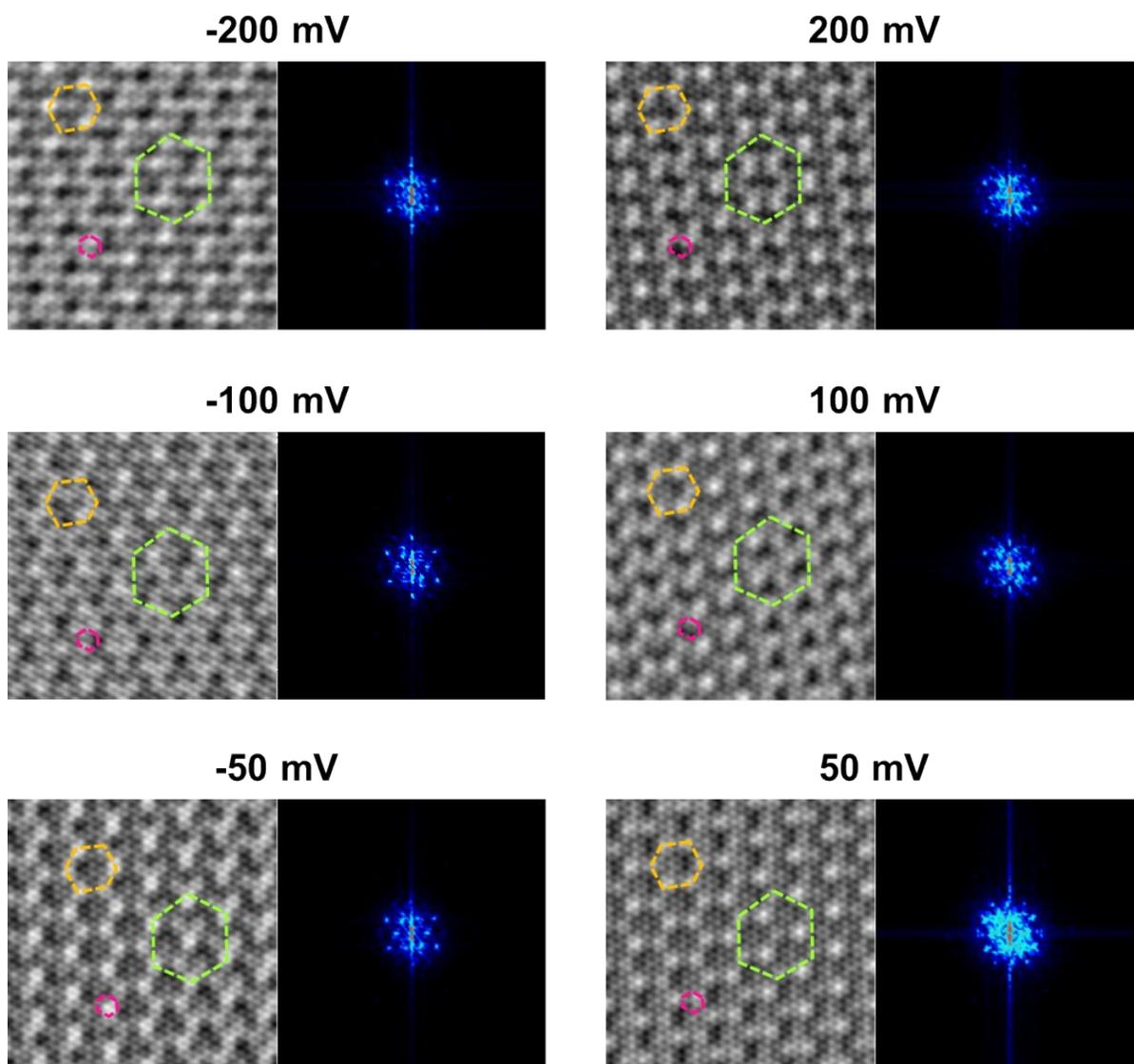
**Table S1.** Elemental composition obtained from Energy-dispersive X-ray spectroscopy (EDX). The atomic ratio of V and S were 32.44% and 67.56%, respectively, indicating V/S = 1/2.08.



**Figure S1.** Left panel: Representative selective area electron diffraction (SAED) patterns of as-grown  $\text{VS}_2$  bulk crystal before low temperature vacuum annealing treatment. Right panels: Simulated SAED patterns of trigonal  $\text{VS}_2$  (top) and monoclinic  $\text{V}_5\text{S}_8$  (bottom). The lattice planes in the red and green circles are indexed for the trigonal  $\text{VS}_2$  (top) and monoclinic  $\text{V}_5\text{S}_8$ , respectively. Note that lattice information of  $\text{VS}_2$  and  $\text{V}_5\text{S}_8$  are as following: trigonal  $\text{VS}_2 - P_{3m1}$  space group,  $a = 3.221 \text{ \AA}$ ,  $c = 5.755 \text{ \AA}$  and monoclinic  $\text{V}_5\text{S}_8 - C_{2/m}$  space group,  $a = 11.399 \text{ \AA}$ ,  $b = 6.668 \text{ \AA}$ ,  $c = 7.919 \text{ \AA}$ ,  $\beta = 134.4^\circ$ .



**Figure S2.** (a) Plot of ARPES intensity for bulk VS<sub>2</sub> along the  $\Gamma$ M ( $h\nu = 128$  eV) direction at 15 K. (b) Electronic band structure obtained from the DFT calculations of bulk VS<sub>2</sub>.



**Figure S3.** STM topographic and corresponding fast Fourier transform (FFT) images of filled states (-200, -100, and -50 mV) and empty states (50, 100, and 200 mV). Magenta, orange, and green dashed hexagons indicate basis,  $\sqrt{6} \times \sqrt{6}R30^\circ$ , and  $4 \times 4$  CDW lattices, respectively.