Electronic Supplementary Information

Strong electron-phonon coupling driven charge density wave states in stoichiometric 1T-VS\textsubscript{2} crystals

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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.

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Figure S1. Left panel: Representative selective area electron diffraction (SAED) patterns of as-grown VS$_2$ bulk crystal before low temperature vacuum annealing treatment. Right panels: Simulated SAED patterns of trigonal VS$_2$ (top) and monoclinic V$_5$S$_8$ (bottom). The lattice planes in the red and green circles are indexed for the trigonal VS$_2$ (top) and monoclinic V$_5$S$_8$, respectively. Note that lattice information of VS$_2$ and V$_5$S$_8$ are as following: trigonal VS$_2$ – $P_{3m1}$ space group, $a = 3.221$ Å, $c = 5.755$ Å and monoclinic V$_5$S$_8$ – $C_{2/m}$ space group, $a = 11.399$ Å, $b = 6.668$ Å, $c = 7.919$ Å, $\beta = 134.4^\circ$. 
**Figure S2.** (a) Plot of ARPES intensity for bulk VS$_2$ along the $\Gamma_M$ ($h\nu = 128$ eV) direction at 15 K. (b) Electronic band structure obtained from the DFT calculations of bulk VS$_2$. 
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 4×4 CDW lattices, respectively.