Supporting Information

Epitaxial growth and electronic properties of antiferromagnetic semiconducting VI₂ Monolayer

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Fig. S1 Moiré superstructures formed by single-layer VI$_2$ on iodine buffer layer. (a, b) The feasible schematic illustration of the dot phase. Yellow balls are iodine atoms of buffer layer on Au(111), and its period is 4.59 Å. Orange balls are iodine atoms in the bottom layer of VI$_2$, the other atoms are not displayed for simplicity. The Moiré pattern with periodicity of ~ 3.3 nm, it is consistent with the experimental results. (c) The dI/dV spectra obtained on different Moire superstructures have a band gap of 2.8 eV and no obvious difference was observed.

Fig. S2 Magnetic properties of VI$_2$ ML regulated by in-plane lattice constants. (a to d) DFT calculated exchange $J$ Néel temperature $T_N$, magnetic anisotropy energy (MAE) and gap as a function of in-plane a.
**Fig. S3** The Heisenberg exchange interactions between V$^{2+}$ ions near vanadium and iodine vacancy defects. (a) Top view of one V vacancy defect in the 5 × 5 supercell VI$_2$ ML. The V vacancy defect is shown by the red empty circle. The back, blue and green double arrowed lines represent the considered Heisenberg exchange interactions $J_{v1}$, $J_{v2}$ and $J_{v3}$ that are near the V vacancy defect. (b) Top view of one I vacancy defect in the 5 × 5 supercell VI$_2$ ML. The I vacancy defect is shown by the green empty circle. In (a) and (b), red, green and yellow balls represent V, upper and bottom I atoms, respectively. The back, blue and green double arrowed lines represent the considered Heisenberg exchange interactions $J_{i1}$, $J_{i2}$ and $J_{i3}$ that are near the I vacancy defect.

\[ J_{v1} = -2.96 \text{ meV} \]
\[ J_{v2} = 0.82 \text{ meV} \]
\[ J_{v3} = 5.08 \text{ meV} \]
\[ J_{i1} = -24.15 \text{ meV} \]
\[ J_{i2} = 5.84 \text{ meV} \]
\[ J_{i3} = 4.49 \text{ meV} \]