## **Supplementary Information**

## Atomistic real-space observation of van der Waals layered

## structure and tailored morphology in VSe<sub>2</sub>

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**Figure S1.** (a) X-ray diffraction (XRD) pattern of VSe<sub>2</sub> nanosheets ensemble synthesized on a mica substrate. All the peaks are indexed to hexagonal VSe<sub>2</sub> phase (JCPDS card no. 89-1641), except the peaks from mica substrate, marked with asterisks. (b) Combined XRD patterns with VSe<sub>2</sub> on mica (black solid line) and just mica substrate (red solid line). The predominant XRD peaks of VSe<sub>2</sub> can be detected solely.



**Figure S2.** (a) HAADF-STEM image of  $VSe_2$  nanosheet and brightness profiles obtained at the line of the red box (b) and green box (c) in (a). The height of the peaks in the profiles indicates the intensity of high-angle scattered electron beams from the specimen's atoms. We conjecture the inplane lattice constant is in 3.26±0.2 Å, and out-of-plane lattice contant is in 6.27±0.3 Å.

