Supporting Information

Crossover between rigid and reconstructed moiré lattice in *h*-BNencapsulated twisted bilayer WSe₂ with different twist angles

Kei Kinoshita^{1,*}, Yung-Chang Lin², Rai Moriya^{1,*}, Shota Okazaki³, Momoko

Onodera¹, Yijin Zhang¹, Ryosuke Senga², Kenji Watanabe⁴, Takashi Taniguchi⁵,

Takao Sasagawa³, Kazu Suenaga⁶, and Tomoki Machida^{1,*}

¹ Institute of Industrial Science, University of Tokyo, 4-6-1 Komaba, Meguro, Tokyo 153-8505, Japan

² National Institute of Advanced Industrial Science and Technology, 1-1-1 Higashi, Tsukuba 305-8565, Japan

³ Laboratory for Materials and Structures, Tokyo Institute of Technology, 4259 Nagatsuta, Yokohama, Kanagawa 226-8503, Japan

⁴ Research Center for Electronic and Optical Materials, National Institute for Materials Science, 1-1 Namiki, Tsukuba 305-0044, Japan

⁵ Research Center for Materials Nanoarchitectonics, National Institute for Materials Science, 1-1 Namiki, Tsukuba 305-0044, Japan

⁶ The Institute of Scientific and Industrial Research, Osaka University, 8-1 Mihogaoka, Ibaraki, Osaka 567-0047, Japan

*E-mail:

kkino@iis.u-tokyo.ac.jp; moriyar@iis.u-tokyo.ac.jp; tmachida@iis.u-tokyo.ac.jp



Fig. S1 Illustration of the moiré lattice plotted with respect to the magnitude of reconstruction (from rigid to reconstructed) and twist angle. While magnitude of reconstruction changes the size of W/Se (colored blue) and Se/W (colored light blue) domains, the twist angle changes the size of the moiré unit cell.



Fig. S2 Schematic illustration of sample fabrication methods and TEM images. Top row: non-encapsulated tBL-WSe₂. Middle row: tBL-WSe₂ capped with h-BN. Bottom row: tBL-WSe₂ encapsulated with top and bottom h-BN layers.







Fig. S3 STEM images of tBL-WSe₂ at different twist angles: (a) 2.4° , (b) 2.6° , (c) 2.9° , (d) 3.3° , (e) 3.6° , (f) 3.8° , (g) 4.5° , and (h) 5.3° . All scale bars are 5 nm. In right figures, reconstructed domains are highlighted by blue and light blue marks. Additionally, the moiré unit cell is outlined by the green solid line, the non-reconstructed region centered on the Se/Se stacking region is outlined by the purple solid line, and the positions of the Se/Se stacking region are indicated by orange circles (these are consistent with those in Fig. 2 of the main text). By connecting the centers of triangles formed by three adjacent Se/Se

stacking points, the moiré unit cell can be determined (green outlined hexagon). Then, find the reconstructed domain and draw a hexagon with a purple outline centered on the Se/Se stacking so as not to include that domain. We have confirmed that these green and purple hexagons can be tiled over the entire TEM image and can even be applied to different moiré lattices.



Fig. S4 Sample structure (top panels) and STEM image (bottom panels) of different tBL-WSe₂ devices: (a) tBL-WSe₂ encapsulated with top and bottom h-BN, (b) tBL-WSe₂ capped with top h-BN, and (c) non-encapsulated tBL-WSe₂. All scale bars are 5 nm. The positions of the Se/Se stacking region are indicated by orange circles in each image.



Fig. S5 (a) Schematics of domain walls between W/Se and Se/W. (b,c) Image of (b) before and (c) after transformation at the domain wall in tBL-WSe₂ ($\theta_{BL} = 0.5^{\circ}$) without *h*-BN sample. A movie of this transformation is provided as Supplementary Movie 1.



Fig. S6 (a,c) STEM images for (a) monolayer (ML) WSe₂, (c) *h*-BN, and ML-WSe₂ with *h*-BN. (b,d,e) Fast Fourier transform (FFT) patterns for (b) ML-WSe₂, (d) *h*-BN, and (e) ML-WSe₂ with *h*-BN.