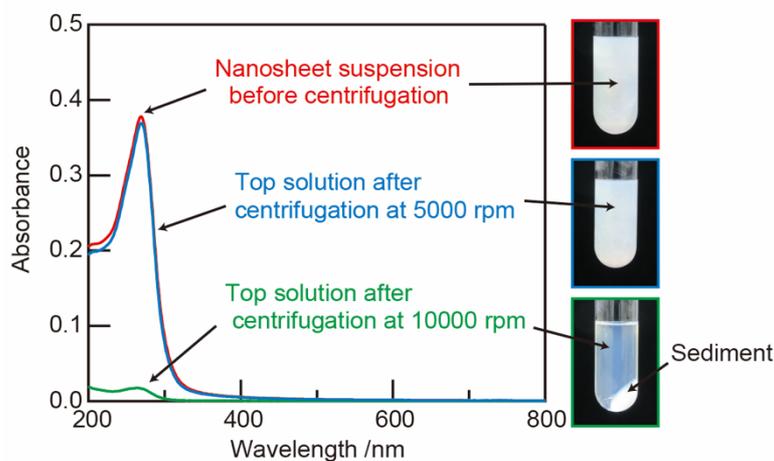


## Guidelines for Arranging 2D Nanosheets into Neatly Tiled Monolayer Films by a Spin-Coating Process

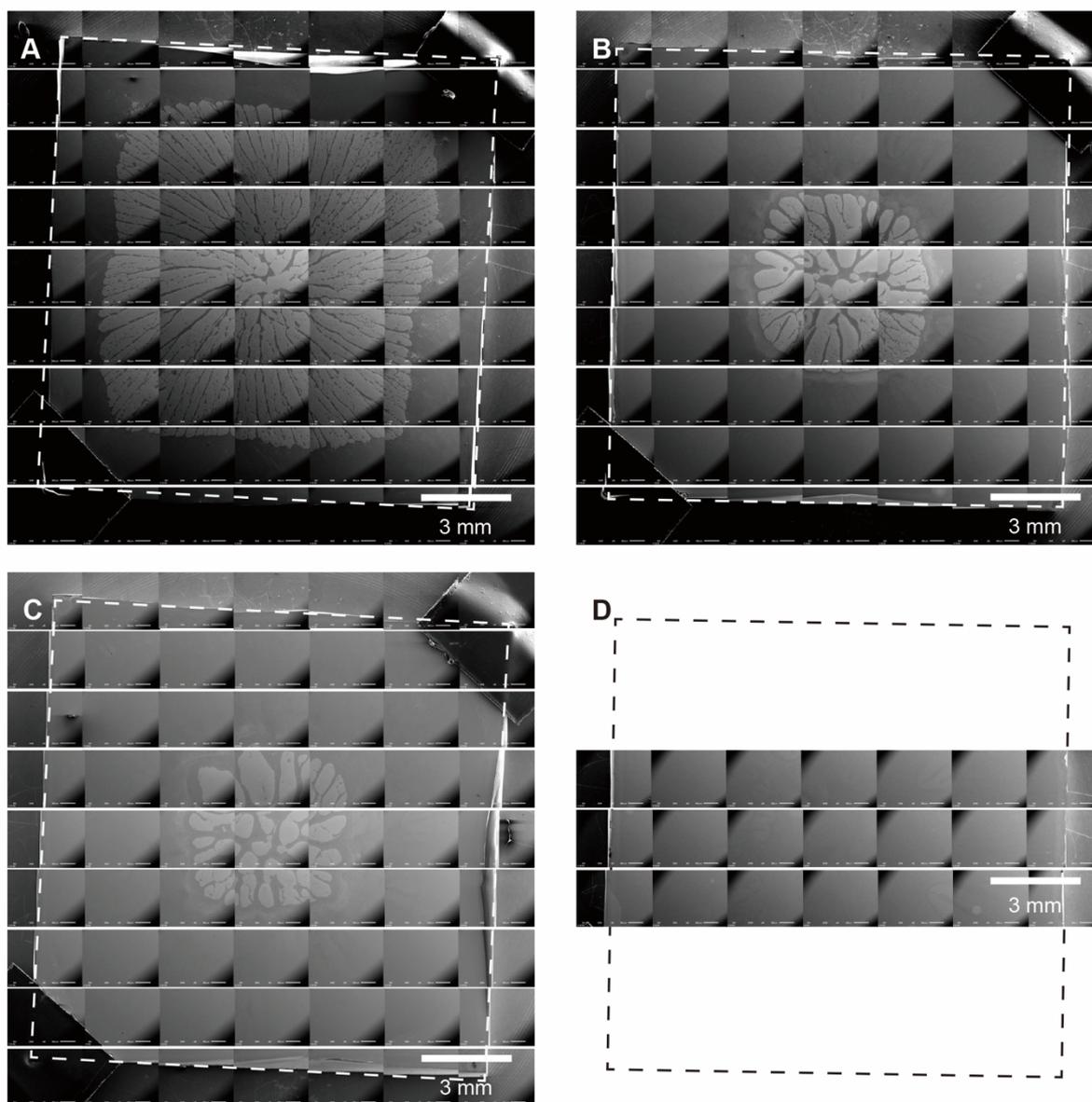
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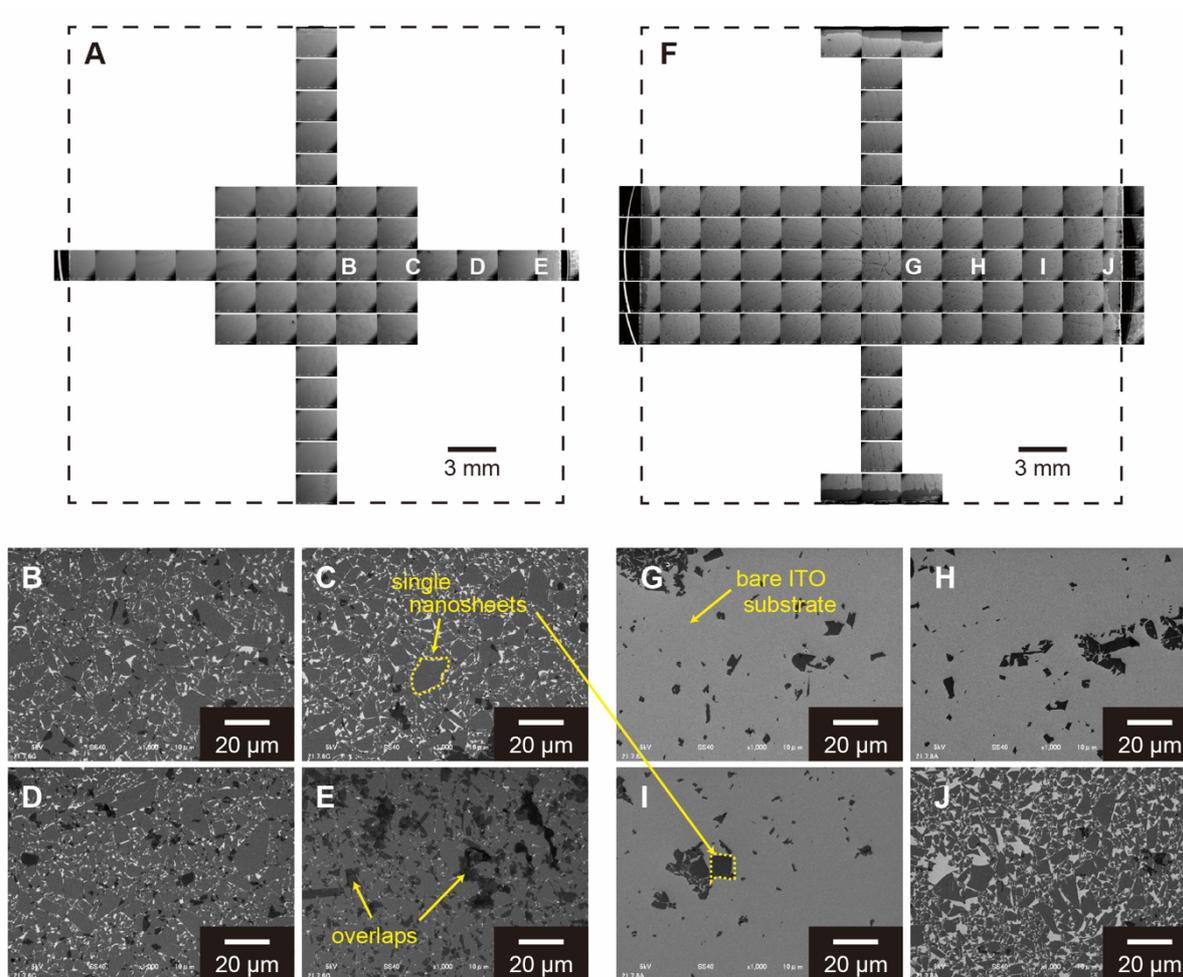
\* Corresponding author. E-mail: sakai.nobuyuki@nims.go.jp



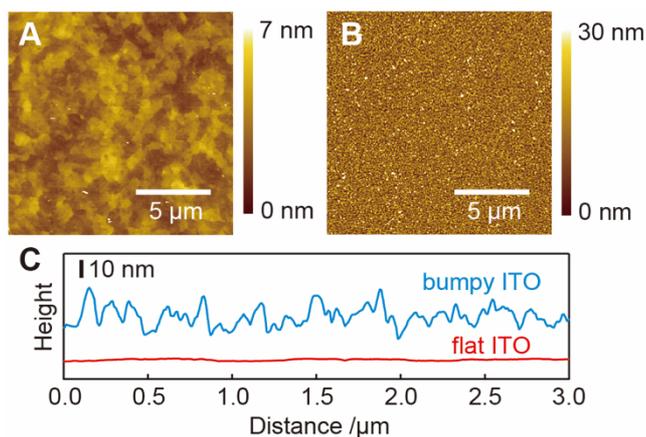
**Figure S1.** UV-vis absorption spectra and pictures of aqueous suspensions of  $\text{Ti}_{0.87}\text{O}_2^{0.52-}$  nanosheets before and after being centrifuged at 5000 rpm (2300g) or 10000 rpm (9300g) for 10 min. The top solutions were diluted 1000 times with water for acquiring their spectra. The nanosheets settled when centrifuged at 10000 rpm, whereas they did not settle at 5000 rpm.



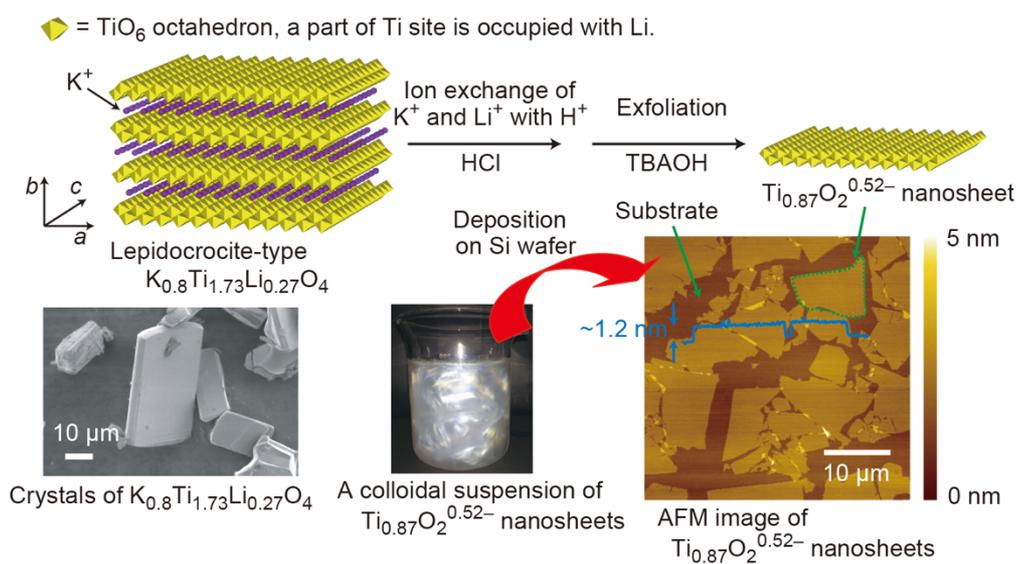
**Figure S2.** Assembly of SEM images of  $\text{Ti}_{0.87}\text{O}_2^{0.52-}$  nanosheets on a flat ITO substrate ( $15 \text{ mm} \times 15 \text{ mm}$ ) spin-coated at (A) 4500, (B) 2600, (C) 2250, and (D) 2000 rpm. The concentration of  $\text{Ti}_{0.87}\text{O}_2^{0.52-}$  nanosheet suspension used for the spin-coating is 0.1 wt %. The square with a dashed line indicates the position of the substrate edge.



**Figure S3.** (A, F) Assembly of SEM images of  $\text{Ti}_{0.87}\text{O}_2^{0.52-}$  nanosheets on a flat ITO substrate ( $30\text{ mm} \times 30\text{ mm}$ ) spin-coated at (A–E) 1050 and (F–J) 8000 rpm. The square with a dashed line indicates the position of the substrate edge. The concentration of  $\text{Ti}_{0.87}\text{O}_2^{0.52-}$  nanosheet suspension used for the spin-coating is 0.1 wt %. Magnified images were collected at (B, G) 2 mm, (C, H) 6 mm, (D, I) 10 mm, and (E, J) 14 mm apart from the center of the substrate, the locations of which are indicated in the images (A, F).



**Figure S4.** AFM images of bare ITO substrates with (A) flat and (B) bumpy surfaces and (C) their cross-sectional profiles.



**Figure S5.** Synthesis procedure of  $\text{Ti}_{0.87}\text{O}_2^{0.52-}$  nanosheets via exfoliation of the layered titanate. The nanosheets are obtained as a colloidal suspension. The thickness of the nanosheets is  $\sim 1.2$  nm.