

## Supporting Information for

### Semiconductor monolayer assemblies with oriented crystal faces

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Supplementary Methods  
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## Supplementary Methods

### ***Commercialized products of TiO<sub>2</sub> (rutile), SrTiO<sub>3</sub> and ZnO.***

TiO<sub>2</sub> (Rutile) is from Wako Pure Chemical Industries, Ltd. with a purity of 99%.

SrTiO<sub>3</sub> is from Kojundo Chemical Laboratory Co., Ltd. with a purity of 99%.

ZnO is from Kanto Chemical Co., Inc. with a purity of 99%.

### ***Synthesis of BaTaO<sub>2</sub>N powder.***

BaTaO<sub>2</sub>N powder was prepared by a conventional nitridation method. BaCO<sub>3</sub> and Ta<sub>2</sub>O<sub>5</sub> powders purchased from Kanto Chemical Co. and High Purity Chemical Co., respectively, were mixed by grinding in an agate mortar with a molar ratio of 2:1. The mixture was loaded into an alumina boat and subsequently inserted into the center of a horizontally placed alumina tube furnace with an inner diameter of 24 mm. Then, the sample was heated under dry NH<sub>3</sub> flow at 200 mL min<sup>-1</sup> at 1173 K for 20 h with intermediate grinding. The obtained sample will be referred to as BaTaO<sub>2</sub>N(SSR). Post-treatment of BaTaO<sub>2</sub>N(SSR) was carried out as the next step. A flux of NaCl was added to BaTaO<sub>2</sub>N(SSR), and then mixed by grinding in an agate mortar. Then, the mixture was subjected to heat-treatment under dry NH<sub>3</sub> flow at 100 mL min<sup>-1</sup> in a tubular furnace at 1073 K. The final products were washed with distilled water to remove residual flux.

### ***Synthesis of small-particle LaTiO<sub>2</sub>N powder.***

All of the reagents were analytical grade and used without further purification. The oxide precursor of  $\text{La}_2\text{Ti}_2\text{O}_7$  was prepared by a polymerized complex (PC) method<sup>2</sup>, and then  $\text{LaTiO}_2\text{N}$  was obtained by heating  $\text{La}_2\text{Ti}_2\text{O}_7$  under  $\text{NH}_3$  flow ( $200 \text{ mL min}^{-1}$ ) at  $950^\circ\text{C}$  for 15 h.

#### ***Synthesis of large-particle $\text{LaTiO}_2\text{N}$ powder.***

All of the reagents were analytical grade and used without further purification. The oxide precursor of  $\text{La}_2\text{Ti}_2\text{O}_7$  was prepared by molten salt<sup>3</sup>. In a typical preparation procedure,  $\text{La}_2\text{O}_3$  and  $\text{TiO}_2$  were mixed in a molar ratio of 1:2 and a salt of composition 50 mol% NaCl and 50 mol% KCl was then added, constituting 50 wt% of the total reaction mixture. The mixture was then heated up to  $1150^\circ\text{C}$  at a rate of  $10^\circ\text{C}/\text{min}$  and maintained at  $1150^\circ\text{C}$  for 5 hours, the temperature was cooled down to  $800^\circ\text{C}$  at  $10^\circ\text{C}/\text{min}$  and then cooled down to room temperature naturally. The calcined mixture was added into water to dissolve the salt. Crystallized  $\text{La}_2\text{Ti}_2\text{O}_7$  powder was obtained by filtrating the above aqueous solution and then dried at  $200^\circ\text{C}$  for using. To prepare  $\text{LaTiO}_2\text{N}$ , the  $\text{La}_2\text{Ti}_2\text{O}_7$  precursor was nitrided at  $950^\circ\text{C}$  for 15 h under a  $\text{NH}_3$  flow.

#### ***Characterization of products***

The as-prepared samples were characterized by X-ray powder diffraction (XRD, Geiger-flex RAD-B, Rigaku; Cu  $\text{K}\alpha$ ) and field-emission scanning electron microscopy (FE-SEM; S-4700, Hitachi).

**References:**

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**Supplementary table 1:** Calculated ratio of relative peak intensity of the XRD showed in fig.2

	$I_{101}:I_{110}$	$I_{111}:I_{110}$	$I_{211}:I_{110}$	$I_{220}:I_{110}$	$I_{301}:I_{110}$
TiO <sub>2</sub> powder	0.379	0.181	0.356	0.102	0.082
TiO <sub>2</sub> /GP	0	0	0	0.051	0
	$I_{100}:I_{110}$	$I_{111}:I_{110}$	$I_{200}:I_{110}$	$I_{211}:I_{110}$	$I_{220}:I_{110}$
SrTiO <sub>3</sub> powder	0.037	0.185	0.311	0.204	0.087
SrTiO <sub>3</sub> /GP	3.23	0.467	15.3	0	0
	$I_{002}:I_{100}$	$I_{101}:I_{100}$	$I_{102}:I_{100}$	$I_{110}:I_{100}$	$I_{103}:I_{100}$
ZnO powder	0.671	1.57	0.285	0.388	0.288
ZnO/GP	0.041	0	0	0	0
	$I_{100}:I_{200}$	$I_{110}:I_{200}$	$I_{211}:I_{200}$	$I_{220}:I_{200}$	$I_{310}:I_{200}$
BaTaO <sub>2</sub> N powder	0.266	5.07	1.02	0.311	0.218
BaTaO <sub>2</sub> N/GP	0.417	0	0	0	0

**Supplementary table 2:** Calculated ratio of relative peak intensity of the XRD showed in fig.4

	$I_{002}:I_{112}$	$I_{202}:I_{112}$	$I_{004}:I_{112}$	$I_{114}:I_{112}$	$I_{204}:I_{112}$
LaTiO <sub>2</sub> N powder-LP	0.274	0.162	0.174	0.047	0.145
LaTiO <sub>2</sub> N/GP-LP	0.012	0	0.008	0	0
LaTiO <sub>2</sub> N powder-SP	0.186	0.165	0.177	0.034	0.150
LaTiO <sub>2</sub> N/GP-SP	319	8.02	201	0	0