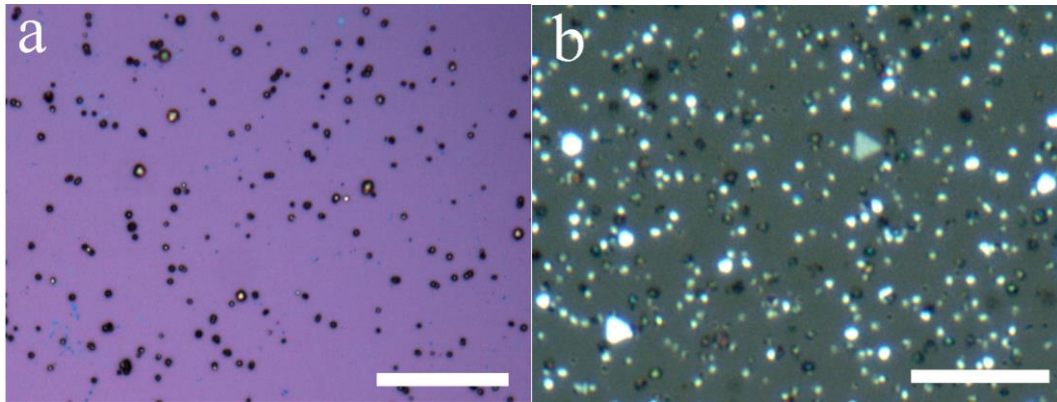
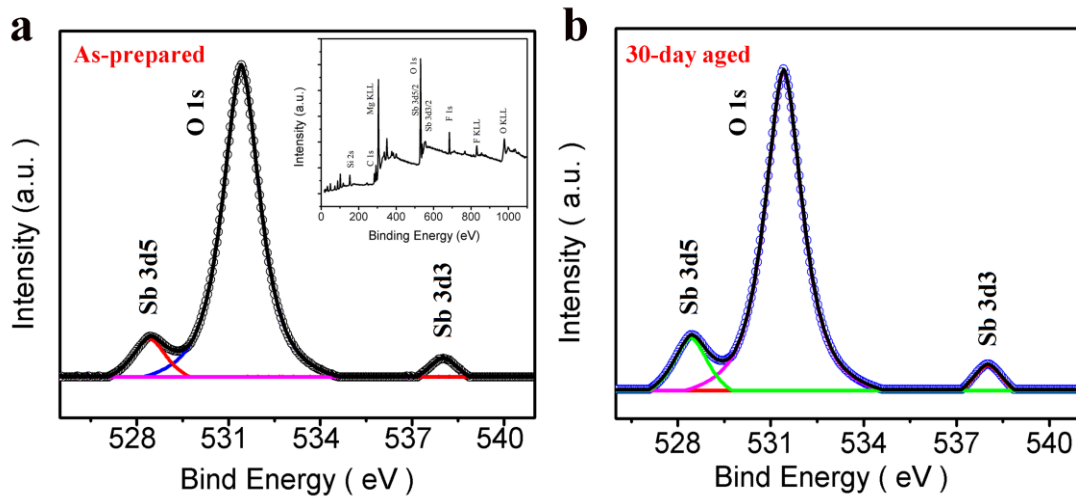


Supplementary Figures

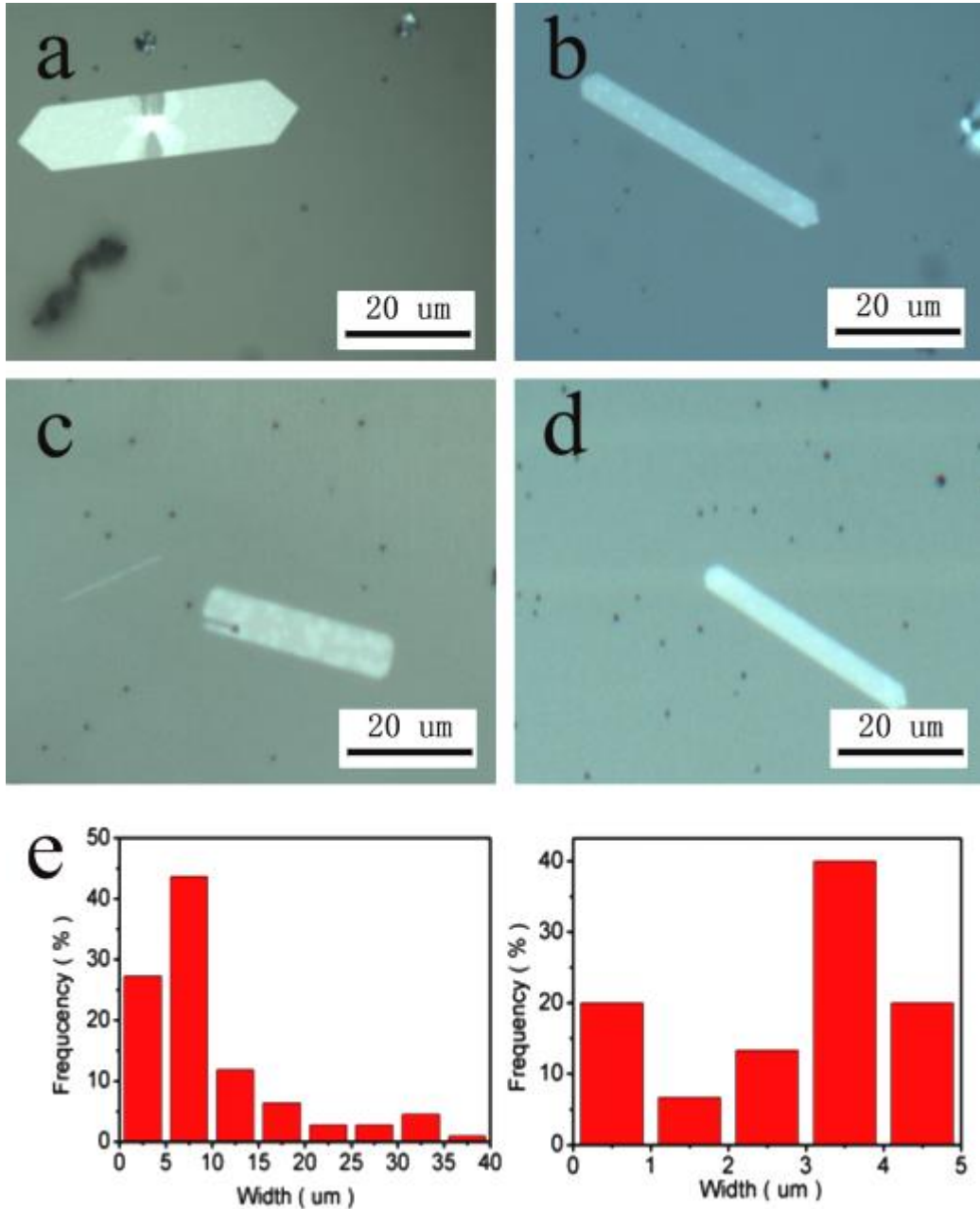


Supplementary Figure 1. Optical microscope images of antimonene polygons grown on (a) silicon substrate and (b) sapphire substrate. The scale bars are 5  $\mu\text{m}$ .



1

2 **Supplementary Figure 2. XPS survey spectra of Sb/mica sample.** (a) Sb 3d region  
 3 spectrum of one freshlt-made sample. In Sb 3d region, 3d3 and 3d5 peaks at 528.4 eV  
 4 and x538 eV, respectively. The O 1s peak is attributed to mica. (b) Sb 3d region  
 5 spectrum of one sample stored for 30 days. No obvious peak shift is observed. The  
 6 different intensity ratio of Sb 3d and O1s in Figure a and b is attributed to different  
 7 coverage in the test zone randomly selected.



1

2 **Supplementary Figure 3. Optical microscope image of several typical**

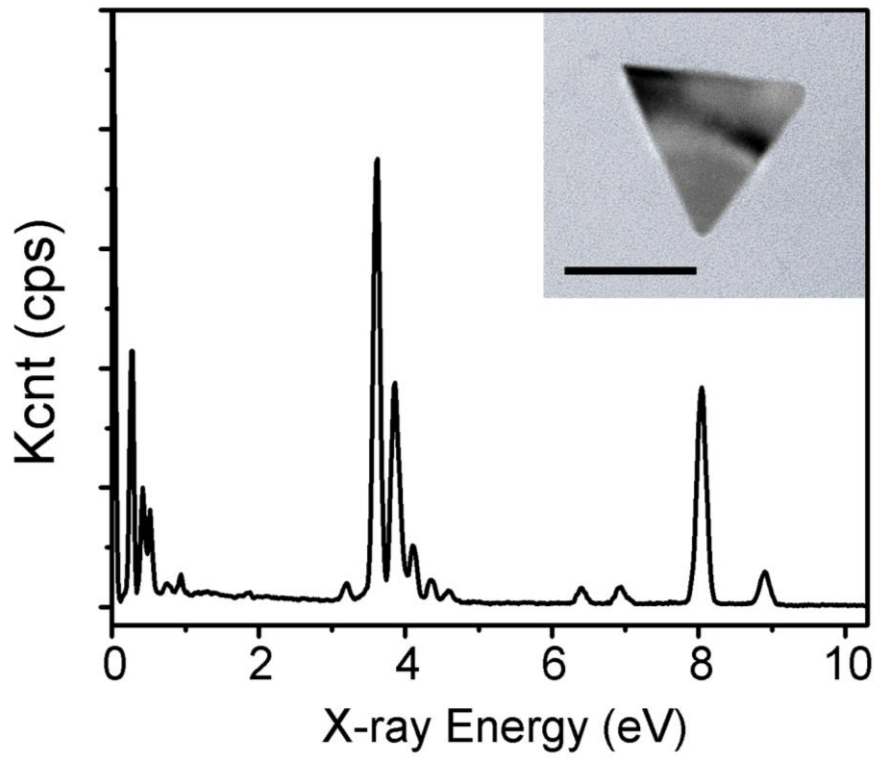
3 **antimonene sheets with stretched hexagonal shape.** (a-d) Similar stretched

4 hexagonal polygons could be mistaken as ribbons when the lateral sizes are small or

5 when the image magnification is low. (e) Dispersion of ribbons width ranging from 0

6 to 40 μm(left) and 5μm (right).

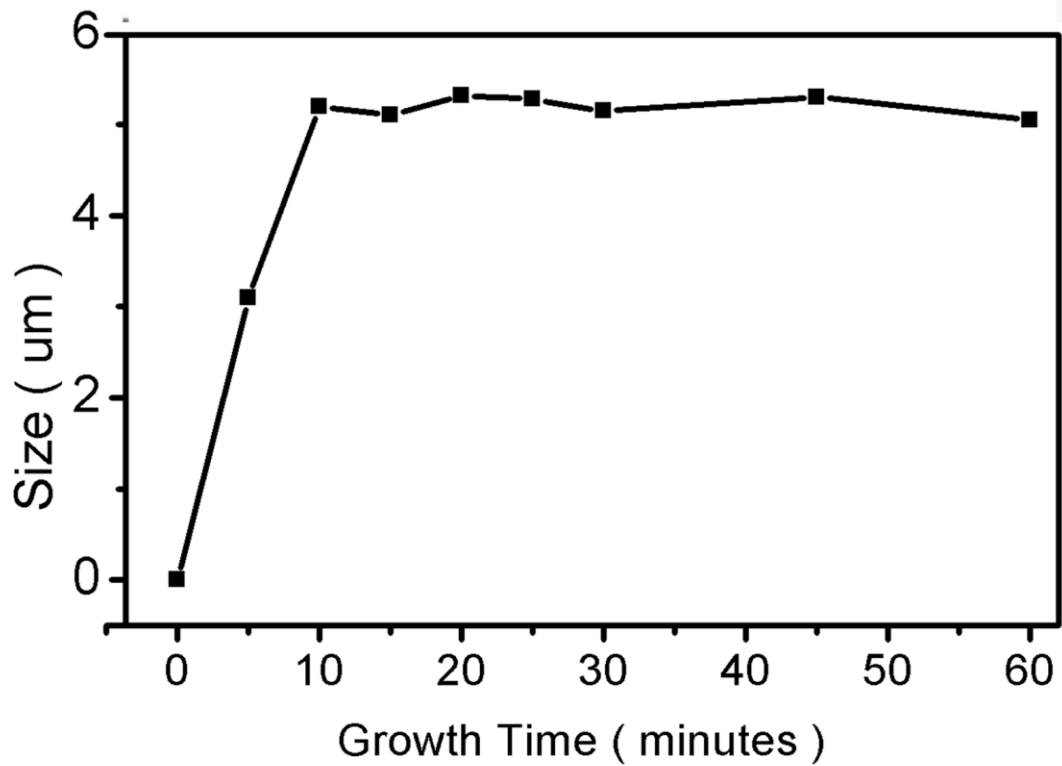
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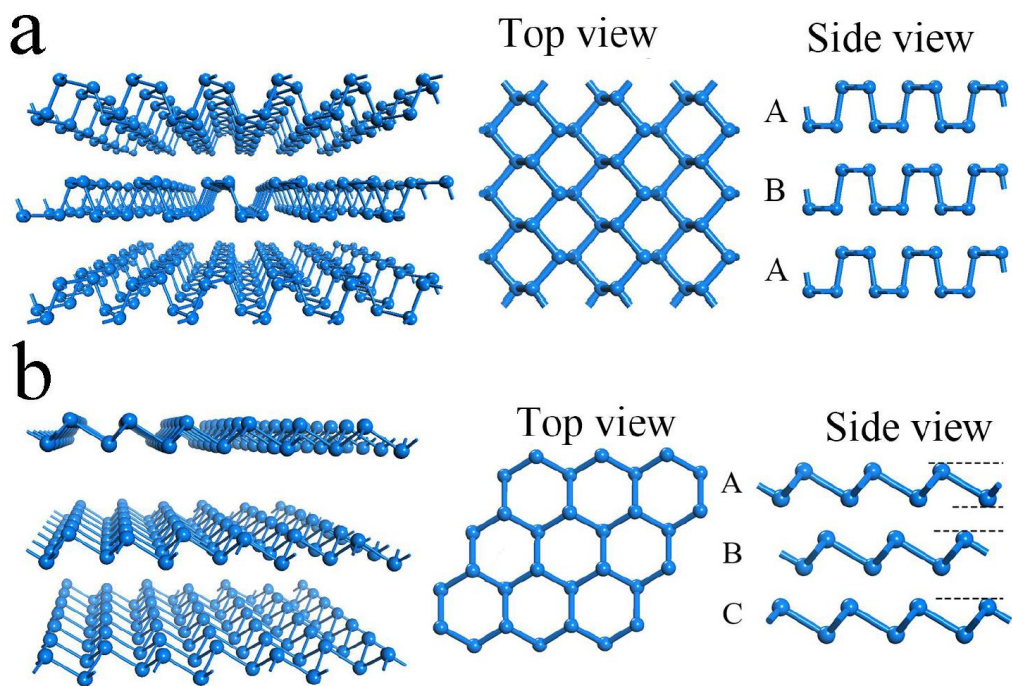
2 **Supplementary Figure 4. EDS spectrum of one tiny nanosheet.** The EDS spereum

3 indicates the tiny sample composed of antimony. The scale bar(inset) is 100 nm.

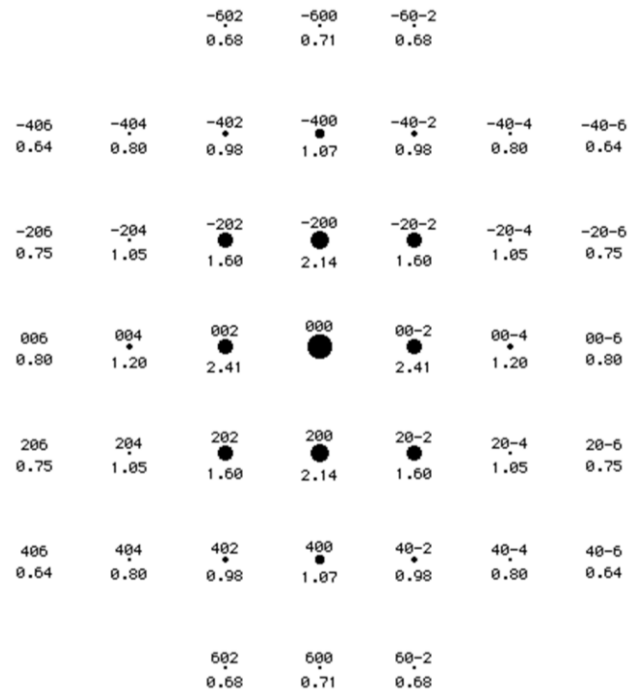


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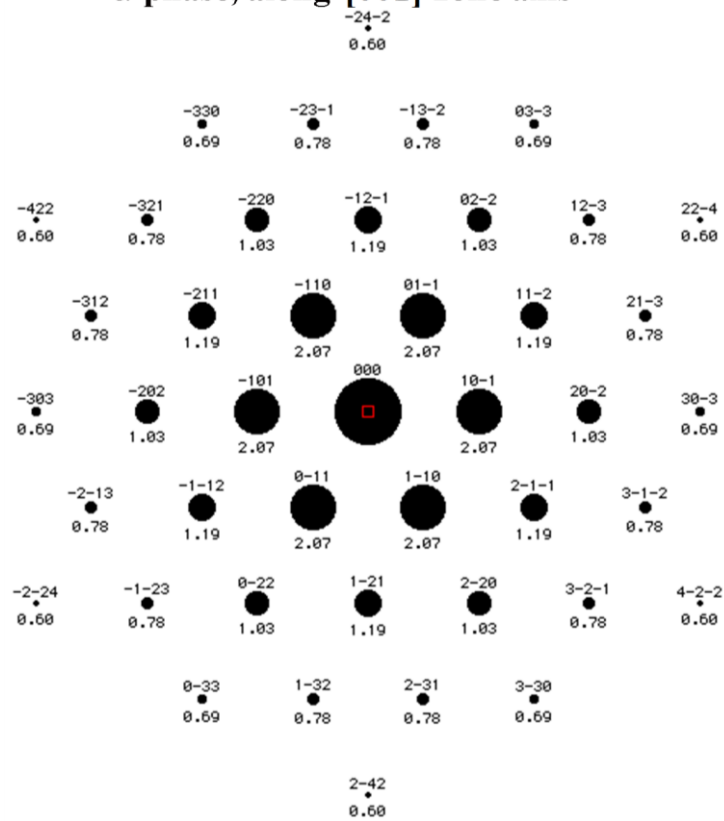
2 **Supplementary Figure 5. Lateral size as a function of growth time and AFM**  
 3 **images of nucleation and lateral growth.** Antimonene layers finished growth in the  
 4 lateral size, even for longer durations. The scale bars of the nucleation and  
 5 growth are 500 nm and 5  $\mu\text{m}$ , respectively.



1  
 2 **Supplementary Figure 6. The comparison of atomic structures of  $\alpha$ -phase (a) and**  
 3  **$\beta$ -phase (b) antimony.  $\alpha$ -phase antimony has orthorhombic structure and  $\beta$ -phase**  
 4 **antimony has rhombohedral structure.**



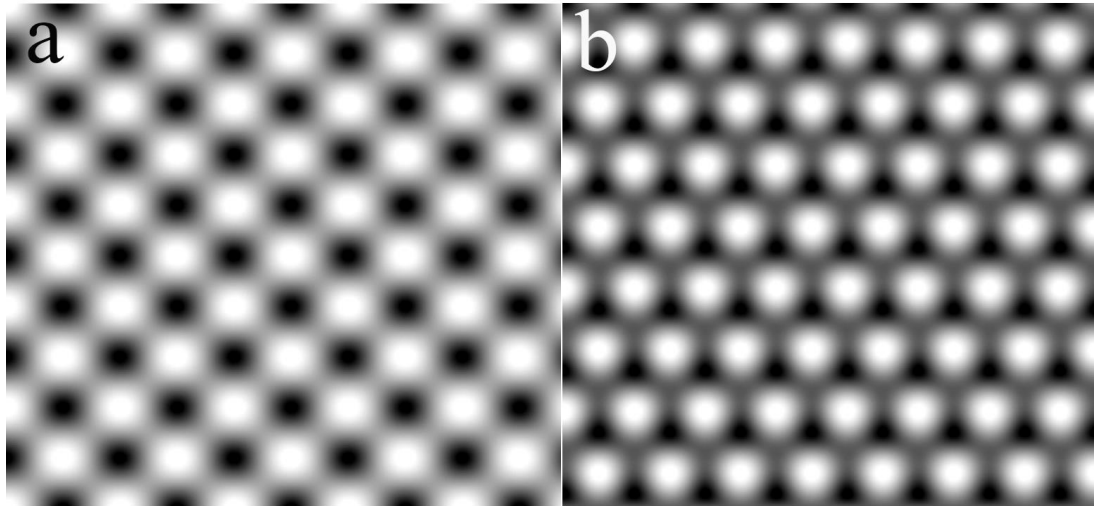
$\alpha$ -phase, along [001] zone axis



$\beta$ -phase, along [001] zone axis

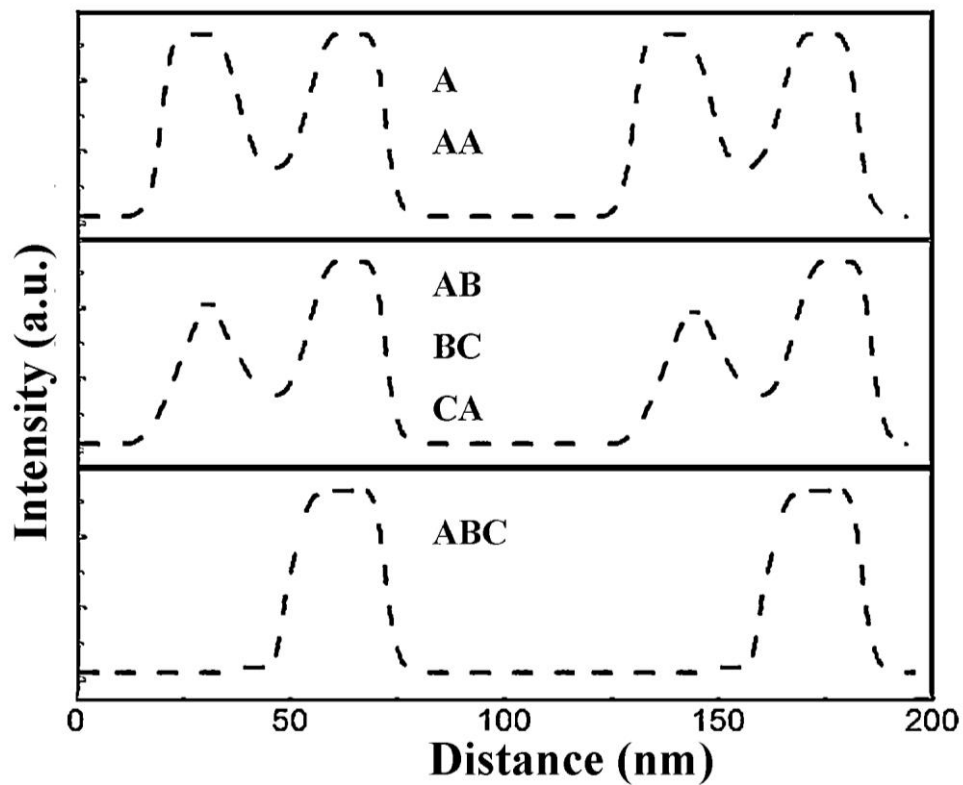
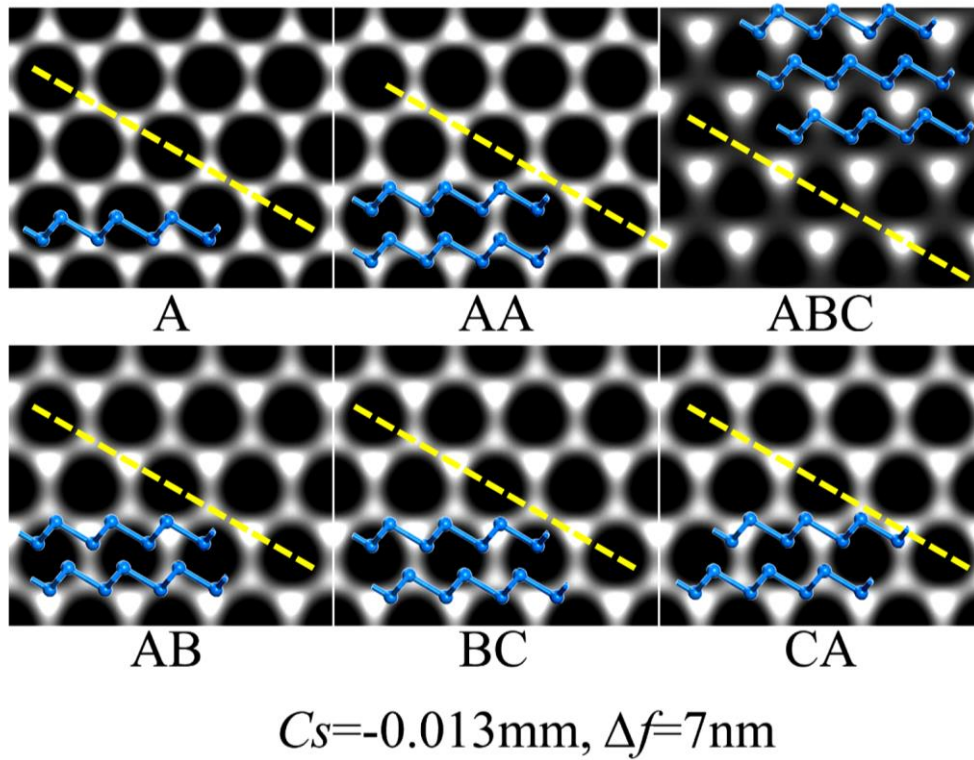
- 1
- 2 Supplementary Figure 7. The simulated kinematical SAED pattern of  $\alpha$ -phase
- 3 and  $\beta$ -phase .

4

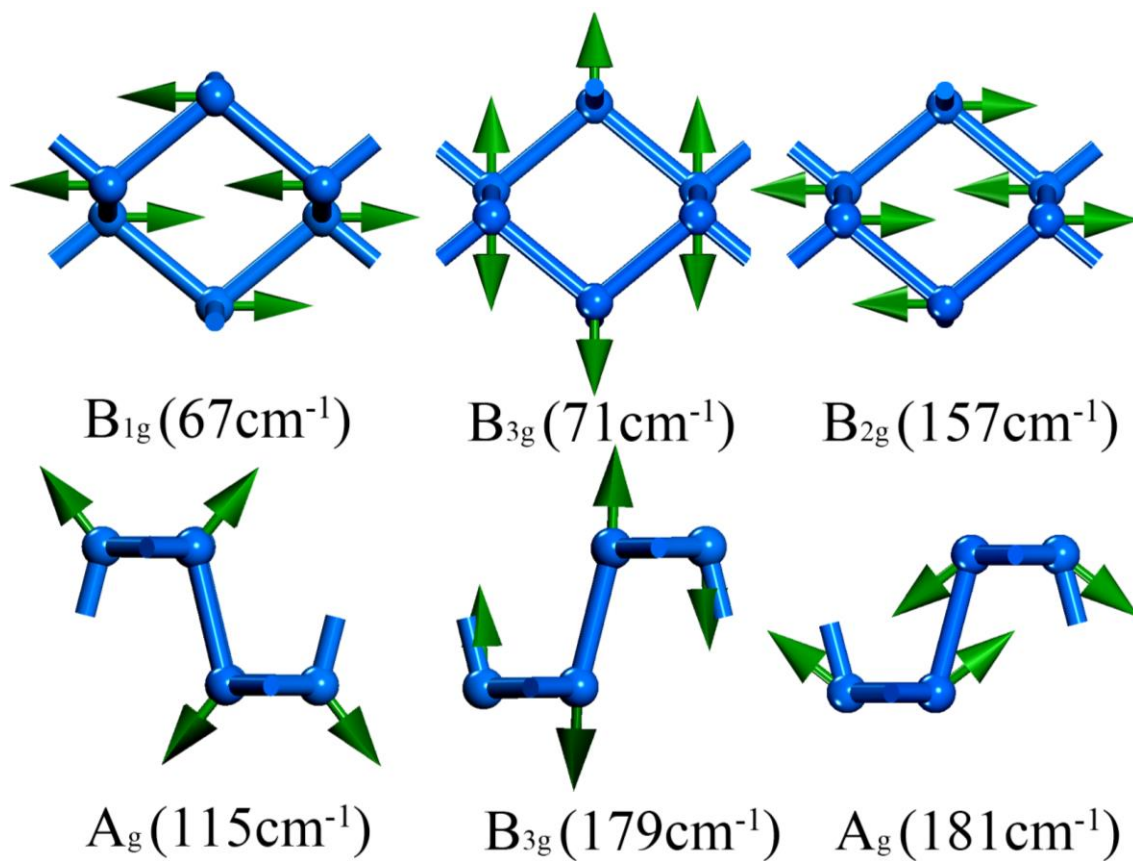


1  
2 **Supplementary Figure 8. Simulated HRTEM images of  $\alpha$ -phase (along [001] zone**  
3 **axis) and  $\beta$ -phase (along [001] zone axis) few-layer antimonenes using the same**  
4 **microscopy parameters.**





1  
 2 **Supplementary Figure 9. Simulated atomic images of antimonene with different**  
 3 **stacking under the the condition of  $C_s = -13 \mu\text{m}$  and  $\Delta Z = +7 \text{ nm}$  and dash**  
 4 **scanning files of different stacking order.**



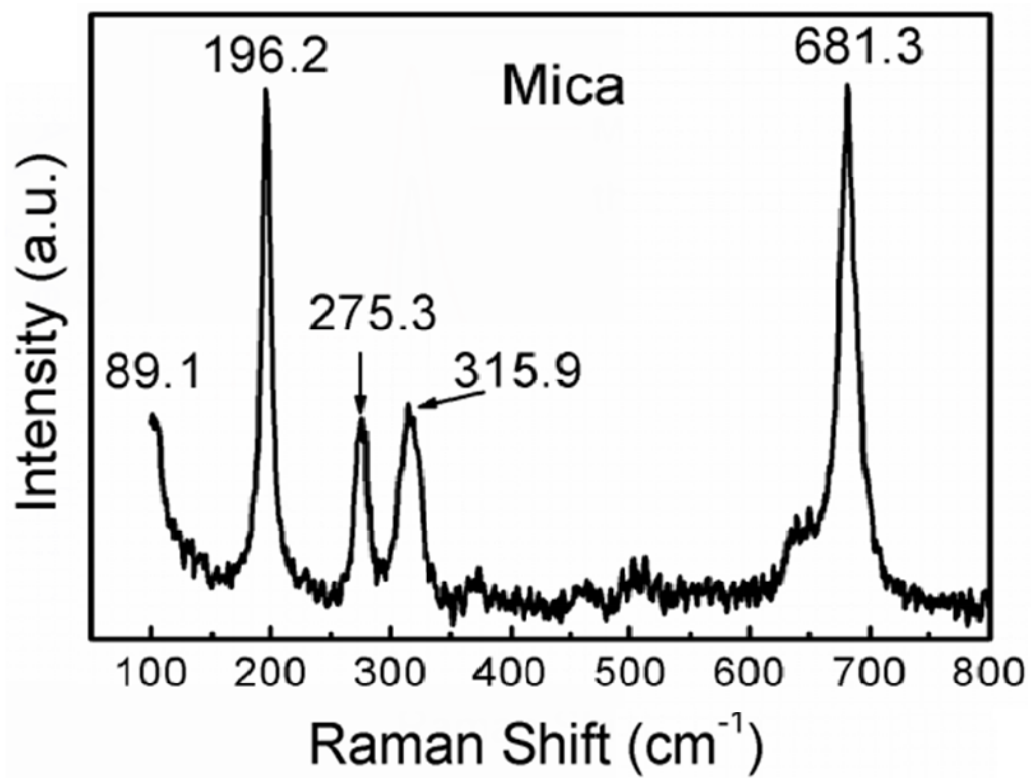
1

2 **Supplementary Figure 10. Raman active modes of  $\alpha$ -phase monolayer**

3 **antimonene.** The frequencies are obtained from the first-principle calculations with

4 the LDA method.

5



1

2 **Supplementary Figure 11.** Raman spectrum of mica substrate measured under the

3 same conditions as the Raman measurement of antimonene polygons.