## **Supporting Information**

Synthesis of highly crystalline Black Phosphorus Thin Film on GaN

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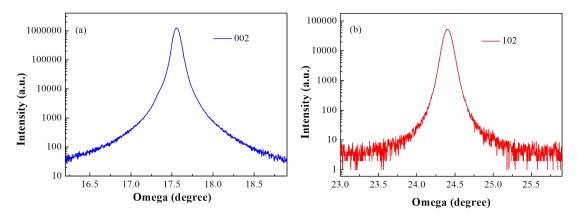
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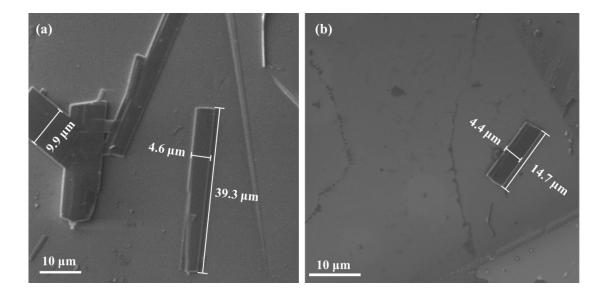
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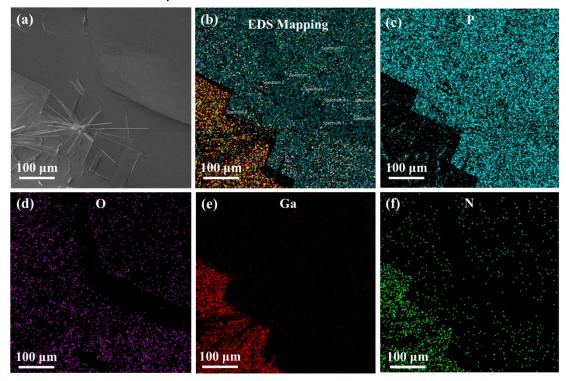
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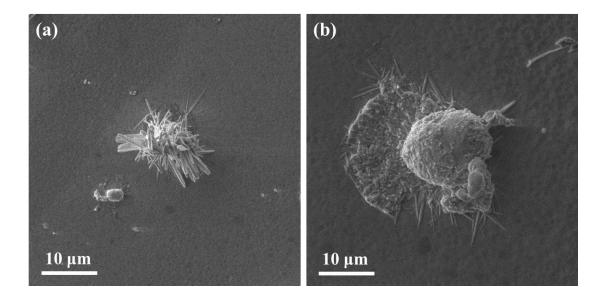
**Supplementary Figure 1** The rocking curves of (002) and (102) planes of GaN epitaxial layer.



Supplementary Figure 2 SEM images of thin-film BP with varied sizes. (a) The maximum length and width are 39.3  $\mu$ m and 9.9  $\mu$ m. (b) The thin-film BP with dimensions of 11 × 81.6  $\mu$ m.



**Supplementary Figure 3** (a)-(b) SEM image and EDS mapping of large area of layered BP on the GaN, (c)-(f) P, O, Ga and N atoms EDS mapping, respectively.



**Supplementary Figure 4** (a)-(b) SEM images of needle-like and other large area amorphous phosphorus on GaN. respectively.