Supplementary Information

Layer-controlled thinning of black phosphorus by Ar ion beam

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Fig. S1 (a~h) AFM images and the thickness profile of BP layers for different thinning time (thinning time = thinning number \times 2 min) and (i) BP thickness and BP layers as a function of thinning number.

Fig. S1 shows the AFM images and the thickness profile of BP layers thinned from 17.1 nm to 7 nm before obtaining 5.8 nm (10-layer) thick BP used in this experiment. The BP layer thinning experiment was conducted after forming 17.1 nm thick (~ 30-layer) non-oxidized BP layer on the SiO₂/Si substrate which was obtained after the removal of the oxidized and excessive BP layers on SiO₂/Si substrates using the Ar⁺ ions and by the measurement of the remaining BP thickness using AFM. Fig. S1(a) shows the AFM image of the BP film of 17.1 nm (reference) on SiO₂/Si after the

removal of oxidized and excessive BP. Fig. S1(b~h) show the BP layers after the thinning for (b) 2 min (thinning 1), (c) 4 min (thinning 2), (d) 6 min (thinning 3), (e) 8 min (thinning 4), (f) 10 min (thinning 5), (g) 12 min (thinning 6), and (h) 14 min (thinning 7). In Fig. S1(i), the thickness of the remaining BP layers and number of BP layers measured before and after the Ar⁺ ion beam thinning using AFM are shown. As shown in the fig. S1, the thickness of the BP layers after the thinning for 2, 4, 6, 8, 10, 12, and 14 min was ~14.7, ~13.5, ~12.1, ~10.8, ~9.6, ~8.3, and ~7 nm which correspond to 26, 24, 22, 20, 18, 16, and 14 layers of BP, respectively.



Fig. S2 (a~h) Optical images of thick BP layers thinned layers for different thinning time (thinning time = thinning number \times 2 min)

Fig. S2 shows the optical images of BP layers thinned from ~ 30 layers to ~14 layers on the SiO₂/Si substrate before obtaining 10-layer BP (5.8 nm) used in this experiment. Fig. S2(a) shows the optical images of the ~ 30-layer BP film (reference) on SiO₂/Si after the removal of oxidized and excessive BP. Fig. S2(b~h) show the BP layers after thinning for (b) thinning 1 (26 layers), (c) thinning 2 (24 layers), (d) thinning 3 (22 layers), (e) thinning 4 (20 layers), (f) thinning 5 (18 layers), (g) thinning 6 (16 layers), and (h) thinning 7 (14 layers). Actual thinning time corresponds to thinning number × 2 min.



Fig. S3 Ar⁺ ion energy distribution measured as a function of 2nd grid voltage from -50 to -250 V

Fig. S3 shows the ion energy distribution of Ar^+ ion from the Ar^+ ion beam source for different 2^{nd} grid voltages from -100 V to -250 V measured using a retarding grid ion energy analyzer during the Ar^+ ion beam thinning. The ICP power of the ion gun was maintained at 200 W and the process chamber pressure was 3.0 mTorr Ar. The 1^{st} grid voltage to the ICP ion gun was maintained at +30 V and the third grid voltage was grounded. A negative voltage to the 2^{nd} grid is to focus the Ar^+ ion beam and to control the flux of the ions extracted from the plasma source. As shown, at a given 2^{nd} grid voltage, the flux of the ions was increased with increase 2^{nd} grid voltage while the ion energy is slightly increased. (44 ~ 45 eV for -50 V, 46 ~ 48 eV for -100 V, 48 ~ 50 eV for -150 V, 49 ~ 51 eV for -200 V, and 50 ~ 52 eV for -250 V of 2^{nd} grid voltage.) Therefore, the ion beam

flux was generally increased with the increase of 2^{nd} grid voltage without significant energy increase unlike the increase of 1^{st} grid voltage.