## Supplementary information

## Langmuir-Blodgett Fabrication of Large-Area Black Phosphorus-C<sub>60</sub> Thin Films and Heterojunction Photodetectors

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**Fig. S1** AFM images of BP LB films on Si. a) Height and d) phase images of BP in NMP (9k), b) Height and e) phase images of BP in CHCl<sub>3</sub> (9-13k), c) Height and f) phase images BP-C<sub>60</sub> in CHCl<sub>3</sub> (9-13k) with 40  $\mu$ g/mL C<sub>60</sub>.



Fig. S2 Transfer ratio (indicated in green line) of BP-C<sub>60</sub> (9-13k) in CHCl<sub>3</sub> with 40  $\mu$ g/mL C<sub>60</sub> at 40 ± 1 mN/m.



Fig. S3 SEM images of BP LB films (9-13k) from a) BP-C<sub>60</sub> with 30  $\mu$ g/mL C<sub>60</sub> and b) BP-C<sub>60</sub> with 50  $\mu$ g/mL C<sub>60</sub>.



**Fig. S4** AFM images of BP LB films (40  $\mu$ g/mL C<sub>60</sub>) on Si. a) Height and d) phase images of BP at 5-9k, b) Height and e) phase images of BP at 2-5k, and c) Height and f) phase images of BP at 1-5k.



**Fig. S5** EDS spectrum of degraded BP nanosheets with clear oxygen signal at ~0.5 keV. The oxidation occurred during liquid exfoliation due to poor sealing of the glass vial containing BP.



**Fig. S6**. a) Structure and b) optical photograph of BP photodetector. Red rectangular indicates a working cell.



Fig. S7 Photoresponse of BP (5-9k rpm)- $C_{60}$  photodetectors under laser excitation at 660 nm and 808 nm.



Fig. S8 Photoresponse of pure  $C_{60}$  photodetectors under laser excitation at 660 nm and 808 nm.



**Fig. S9** Photoresponse of the device with a structure of ITO/ZnO/TAPC/MoO<sub>3</sub>/Ag under laser excitation at 660 nm and 808 nm.



Fig. S10 a) Temporal and b) frequency response of photodetector with BP film after toluene wash under laser excitation at 660 nm. The rise and fall times from the square wave modulation are approximately  $70 \pm 20 \,\mu$ s, but near the resolution limit attainable from the mechanical chopper itself. The frequency domain measurement shows that the bandwidth is 8.8 kHz.



**Fig. S11** a) Frequency-dependent measurement of output spectral density of photodetector under excitation of modulated laser at 660 nm (20 Hz), b) noise equivalent power (NEP) extrapolated

from frequency-dependent measurement. The signal noise ratio (SNR) was obtained through dividing output intensity (at 20 Hz) at each excitation power by noise floor. The extrapolated NEP was  $34 \pm 8$  nW and the detectivity was  $6.2 \pm 1.5 \times 10^6$  cm.Hz<sup>1/2</sup>.W<sup>-1</sup>.