Electronic Supplementary Information

Configuration-Dependent Anti-Ambipolar Van der Waals p-n Heterostructures Based on Pentacene Single Crystal and MoS_2

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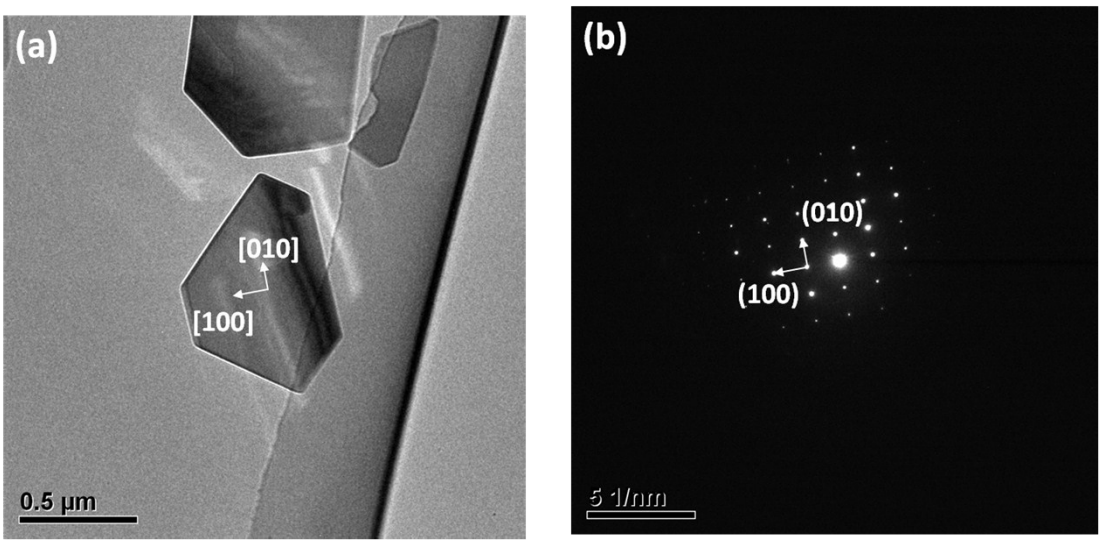

Figure S1 (a) TEM image and (b) the SAED pattern of thin pentacene single crystals.

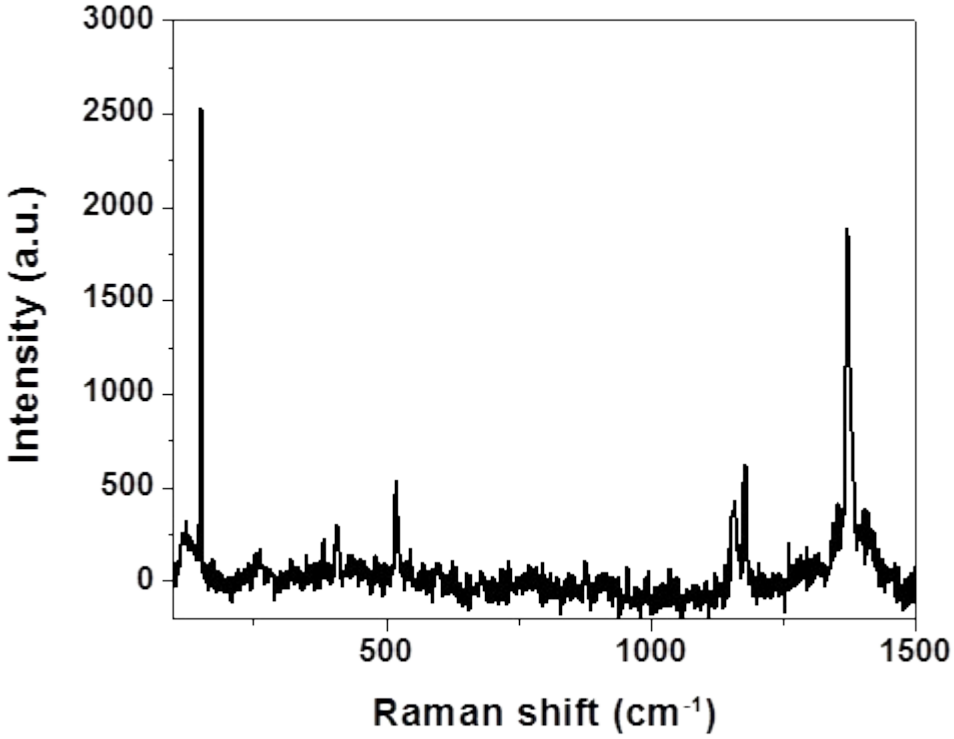

Fig. S2 Raman spectra of the position at the black dot in Fig. 1g.

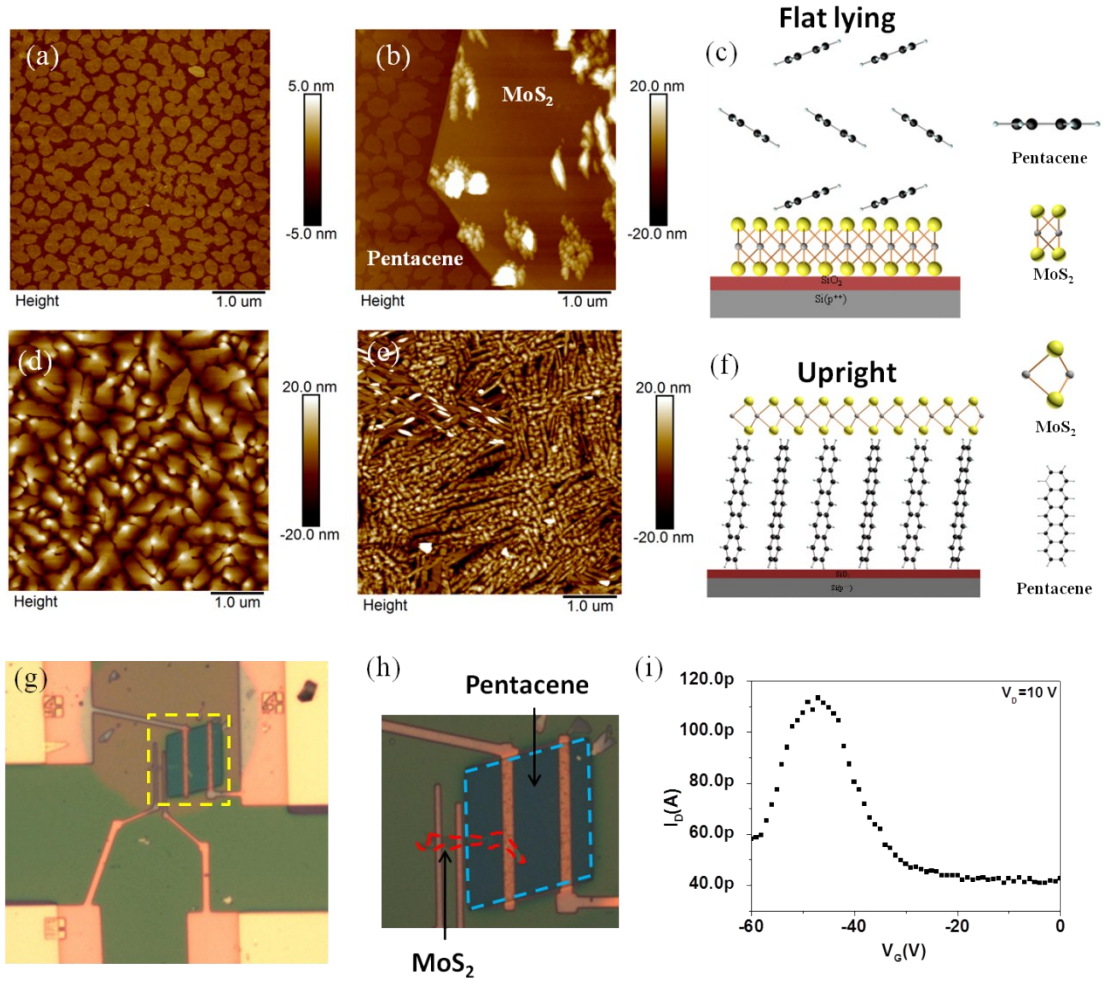

Fig. S3 (a) the AFM topography of pentacene monolayer on SiO_2 (b) the AFM topography of pentacene monolayer on SiO_2 and MoS_2, (c) the schematic of pentacene alignment on MoS_2, (d) the AFM topography of pentacene film on SiO_2, (e) the AFM topography of pentacene film on MoS_2, (f) the schematic of alignment within the heterostructure, (g) the optical graph of the PSC/MoS_2 vdWH, (h) the detail of the yellow square in (g), (i) the corresponding transfer curve of the device in (g).

The topographies of pentacene monolayer on SiO_2 and MoS_2 are shown in Fig. S3a and b. It is obvious that the pentacene monolayer grown on separate material has a marked difference on the morphology, as shown in Fig. S3b. This is due to the diverse pentacene molecule alignments. It is well known that the pentacene molecules' long axes roughly oriented upright with respect to the SiO_2 substrate as

schematically shown in Fig. S3f and it has an island-like shape for monolayer. However, some reports have shown that the pentacene molecules long axes align parallel to the graphene and the hexagonal boron nitride (h-BN) as schematically shown in Fig. S3c.[1] The rod-like morphology in Fig. S3b resembles that of pentacene monolayer grown on the graphene. We indicate the pentacene molecules flat lying on MoS_2, too. The pentacene molecules with flat lying arrangement on MoS_2 are schematically shown in Fig. S3c. We further deposited 40nm thin film on SiO_2 and MoS_2 as shown in Fig. S3d and e. The pentacene thin film on SiO_2 with typical herring bone arrangement is visible in Fig. S3d. While, the pentacene thin film showing with a rod-like morphology on MoS_2 probably inherits from the monolayer arrangement on MoS_2. It is reported that the pentacene film with lying down arrangement on the 2D materials is insulated.[2] Hence, the reported performance of anti-ambipolar for the pentacene film/MoS_2 vdWH device is poor due to the alteration of the pentacene arrangement on MoS_2.[3] We also fabricate the same device with poor performance as shown in S3g, h and i. The molecule alignment in Device I and Device II is shown in Fig. S3f. The typical herring bone molecule alignment with high mobility is maintained. Therefore, our performance is greatly enhanced.

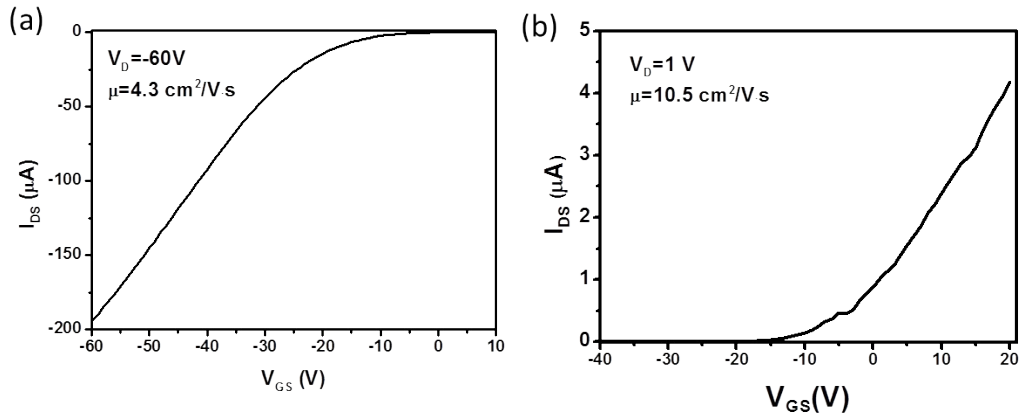

Fig. S4 (a) the transfer curve of a representative PSC device, (b) the transfer curve of a representative MoS_2 device

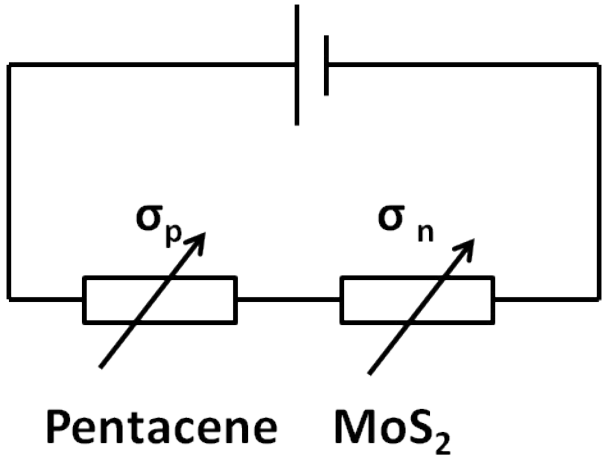

Fig. S5 (a) the schematic circuits of the Device I

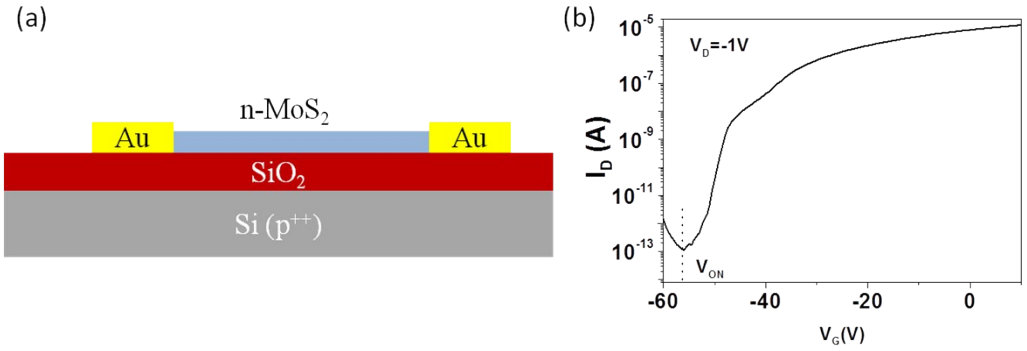

Fig. S6 (a) the schematic of the traditional MoS_2 device. (b) the representative transfer curve of MoS_2.

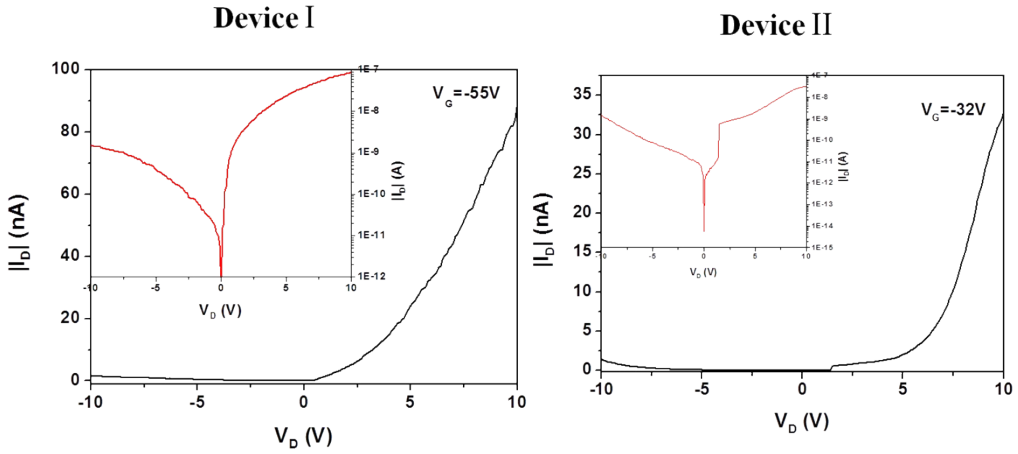

Fig. S7 the output curve of Device I and DeviceII.

Notes and references

1. N. N. Nguyen, S. B. Jo, S. K. Lee, D. H. Sin, B. Kang, H. H. Kim, H. Lee and K. Cho, *Nano Lett.*, 2015, **15**, 2474-2484.
2. A. D. Liao, J. Z. Wu, X. R. Wang, K. Tahy, D. Jena, H. J. Dai and E. Pop, *Phys. Rev. Lett.*, 2011, **106**.
3. D. Jariwala, S. L. Howell, K. S. Chen, J. M. Kang, V. K. Sangwan, S. A. Filippone, R. Turrisi, T. J. Marks, L. J. Lauhon and M. C. Hersam, *Nano Lett.*, 2016, **16**, 497-503.