

Supporting Information for

Graphene-based electrodes for Organic Thin Film Transistors: Graphene transfer, surface morphology and interface properties of Pentacene films on Graphene/SiO₂ surface

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Optical image

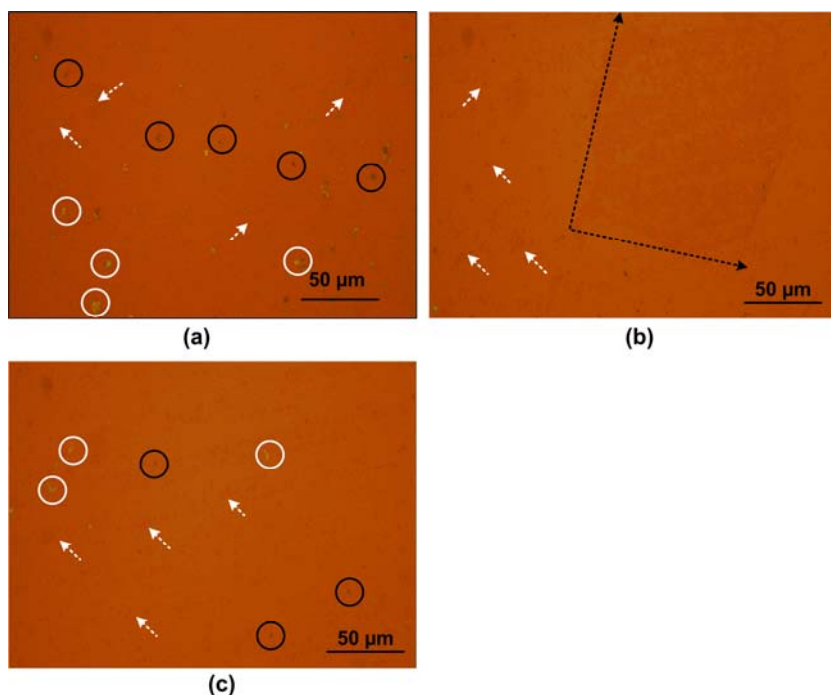


Fig. S1. Optical images of graphene film on SiO₂/Si substrate transferred by the conventional “PMMA-assisted” method. Residual metal particles (white circles), small holes (black circles), multilayer graphene films (white dotted arrows), and PMMA residues (black dotted lines) were observed on transferred graphene films.

SEM image:

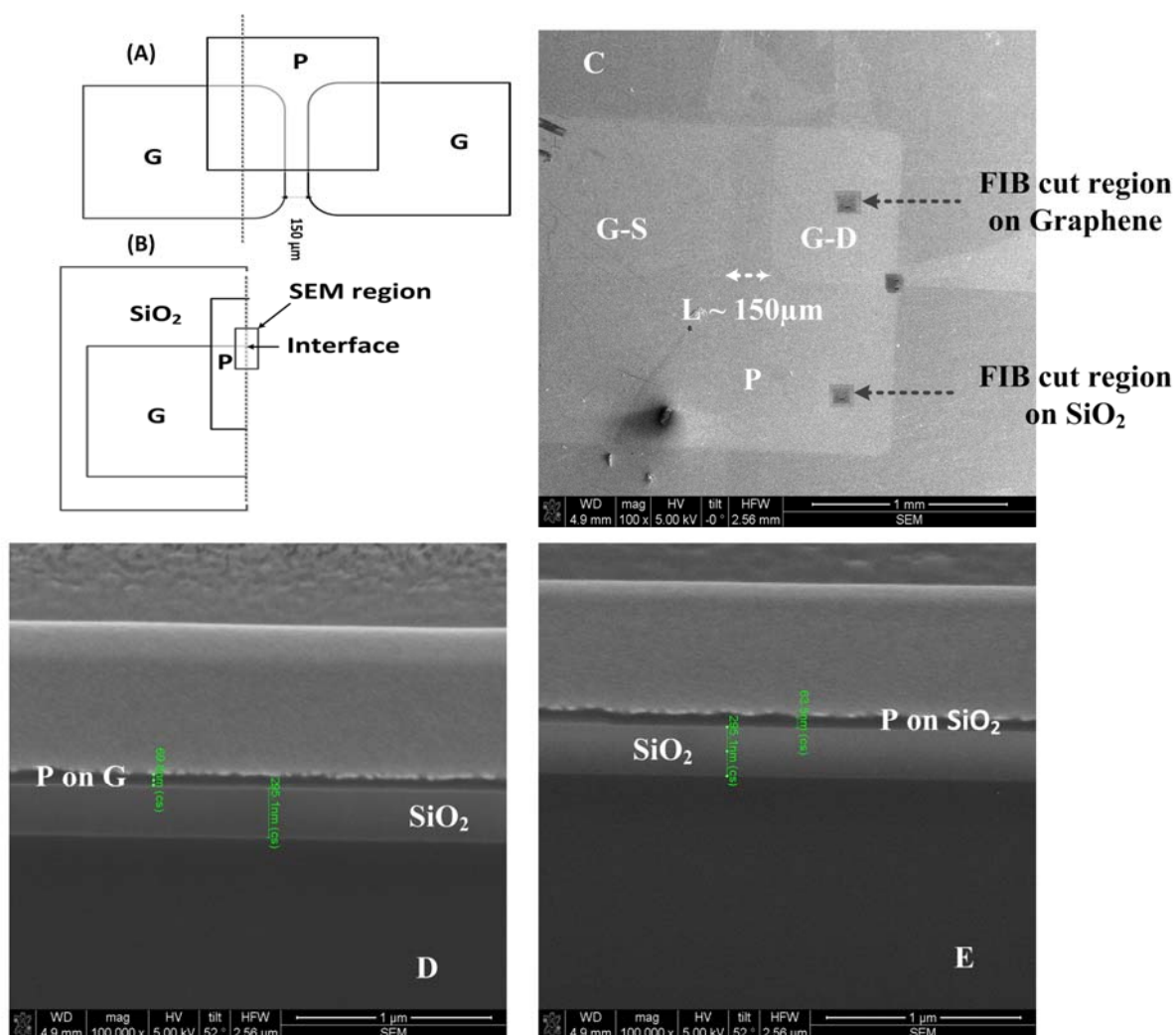


Fig. S2. (A) Shows one OTFT device. P ~ pentacene, G ~ graphene, channel length ~ 150 μm, (B) Sample is prepared for study of the Cross-sectional SEM image. The sample is cut through dotted line to observe the interface clearly. The interface demonstrates the pentacene grown on graphene electrode/ SiO₂ surface. (C) Dual-beam focused ion beam, DB-FIB (FEI Nova-200 nanolab compatible) system utilized a finely focused beam of gallium ions allows the milling of small holes in the sample at well localized sites, so that cross-sectional SEM images of the structure can be obtained, (D) pentacene layer deposited on a graphene surface, and (E) Pentacene layer deposited on SiO₂ surface showing continuous growth of pentacene films.

FIB-TEM image

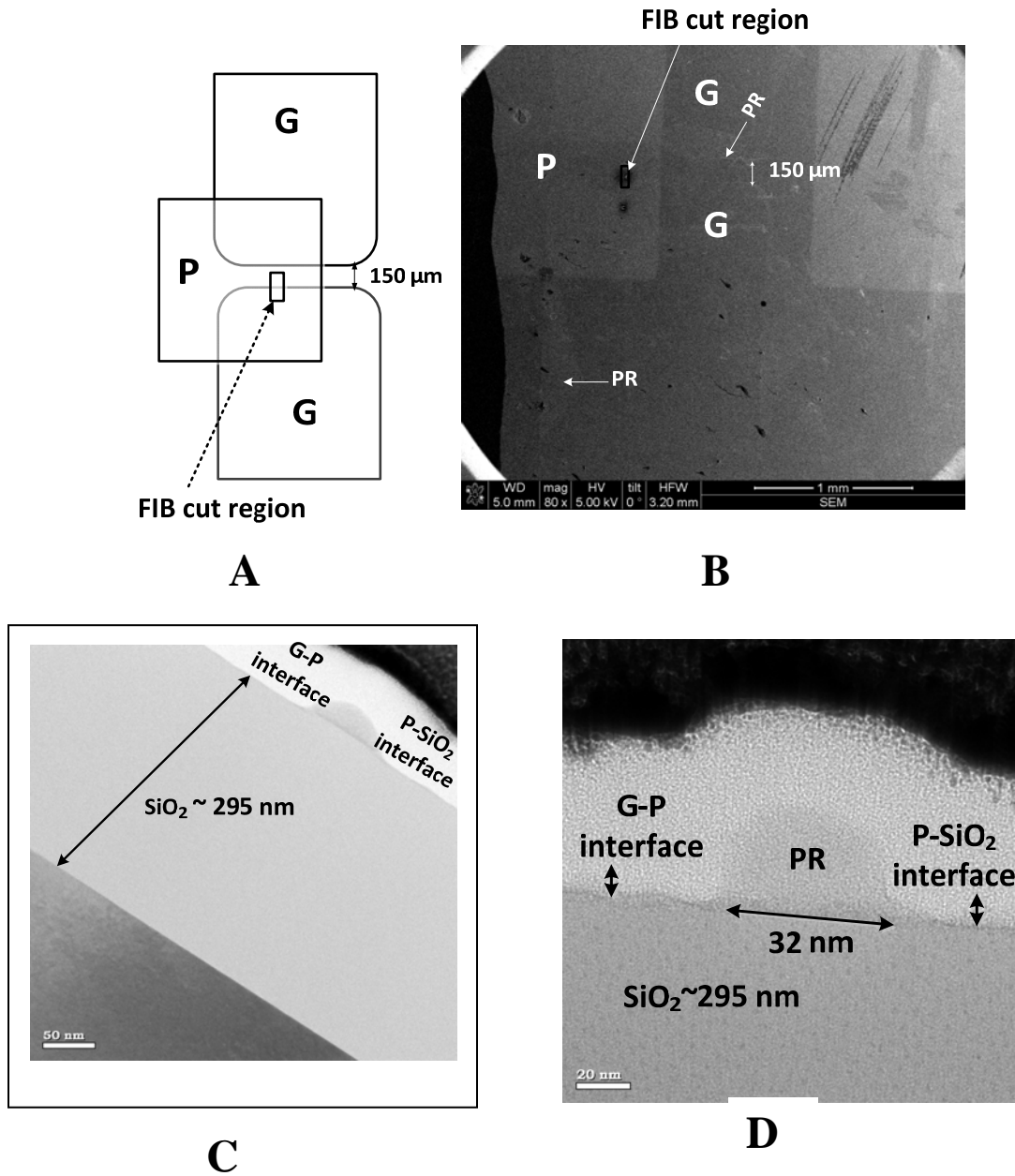


Fig. S3. (A) and (B) shows the clear view of FIB cut region. (C) and (D) illustrates the HR-TEM images of interface between pentacene layer grown on graphene/bare SiO₂ surface. A 32 nm dimple region indicates the existence of photo-resist (PR) between the interfaces during lift-off technique by acetone. This photo-resist residue is clearly observed in AFM images of Fig. 3(m) in original manuscript after patterning the graphene electrode.

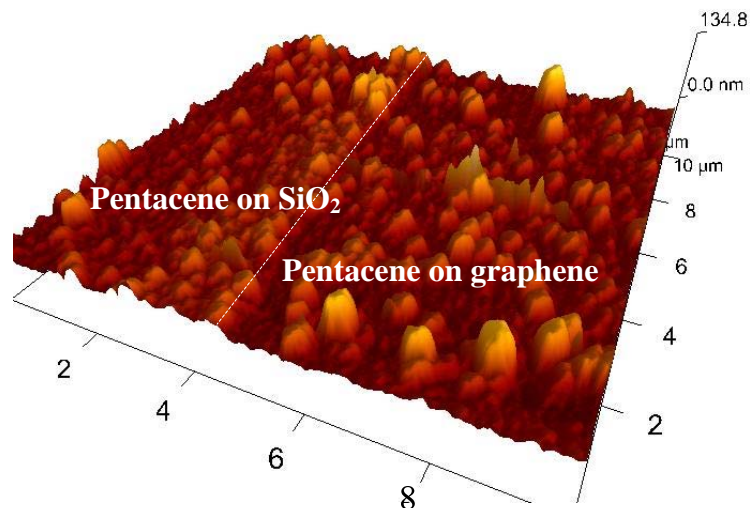


Fig. S4. 3D AFM images ($10\mu\text{m} \times 10\mu\text{m}$) of the 65 nm-thick pentacene films at the boundary between the bare SiO_2 surface and graphene S-D electrode.

Pentacene molecules “lie horizontally” on the Π -conjugated monolayer carbon surfaces. Because most organic semiconductors consist of Π -conjugated planar acenes, these molecules can interact with the underlying graphitic film surface via Π - Π interactions and are arranged according to the aquasi-epitaxial growth mode. On the other hand, pentacene molecules were arranged in a herringbone packing structures “standing nearly vertically” on the SiO_2 gate dielectric, clearly demonstrated from 2D-GIXD patterns studies reported by Lee et al.¹ However some PMMA and photoresist residues exist on patterned graphene electrode. Therefore, pentacene molecule grown on clean graphene surface differs from pentacene grown on transferred and patterned graphene surface. The growth characteristics mainly depend on residue which covers the patterned graphene electrode. To justify this reason, we have added 2D and 3D tapping mode AFM study in our original manuscript. If the scanning position varies on the graphene S-D region, we can clearly observe from Figs. 3(r) and 3(t) (original manuscript) that the grain sizes and rms values of the deposited pentacene film are not constant throughout the graphene S-D region because of the presence of PMMA residues. Polymer residues were unavoidably physisorbed on the graphene surface, as shown in Fig. 3(a). So we can conclude that growth characteristics and surface morphology of pentacene on patterned graphene

electrodes was varied due to residues situated on graphene surface, rather than π - π interaction with graphene. Fig.4 shows that pentacene films grew continuously at the interface between the channel and electrode. These transition regions indicated by white dotted line which strongly suggest better interfaces facilitating charge injection from graphene to semiconductor. The orientational homogeneity of pentacene in channel and electrode regions can allow the continuous grain growth at the interface. The measured work functions of Au and graphene electrodes were 4.85 eV and 4.7eV, respectively. The hole injection barrier was smaller in the graphene electrode (0.5 eV) relative to that in the Au electrode (0.7 eV). Oxygen atoms in PMMA and photoresists residues contribute p-doping in graphene.^{2,3}

The organic-metal interface is dominated by charge exchange, chemical reaction, and/or interface electronic effects such as adsorbate-induced modification of the metal surface. With improved interface properties of pentacene/graphene facilitates efficient carrier injection and transport through graphene to pentacene region.

Small electrostatic dipole was observed between the pentacene/graphene interfaces demonstrating a small charge transfer occurs through the interfaces.⁴ Therefore, the hole injection barrier is small, instead of the low work function of the graphene electrode. The patterned graphene S-D monolayer thickness is also less than 1 nm and this value is less than enough that of an Au metal electrode. The growth of pentacene grain was not restricted due to the less thickness of graphene electrode and its lower surface roughness value (< 0.9 nm). As a result, the pentacene grown on a graphene electrode/channel interface moves continuously toward the same direction although the morphology and grain sizes are not identical on electrode and channel region.

References

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