[Supporting Information]

Impacts of Molecular Orientation on the Hole Injection Barrier Reduction: CuPc/HAT-CN/Graphene

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1. The work function saturation of a HAT-CN layer on graphene

Prior to CuPc deposition, we checked the thickness at which the work function of HAT-CN is saturated. Although the nominal thickness is slightly different from the previous report,[1] the overall curve shape and the saturation value coincides well.

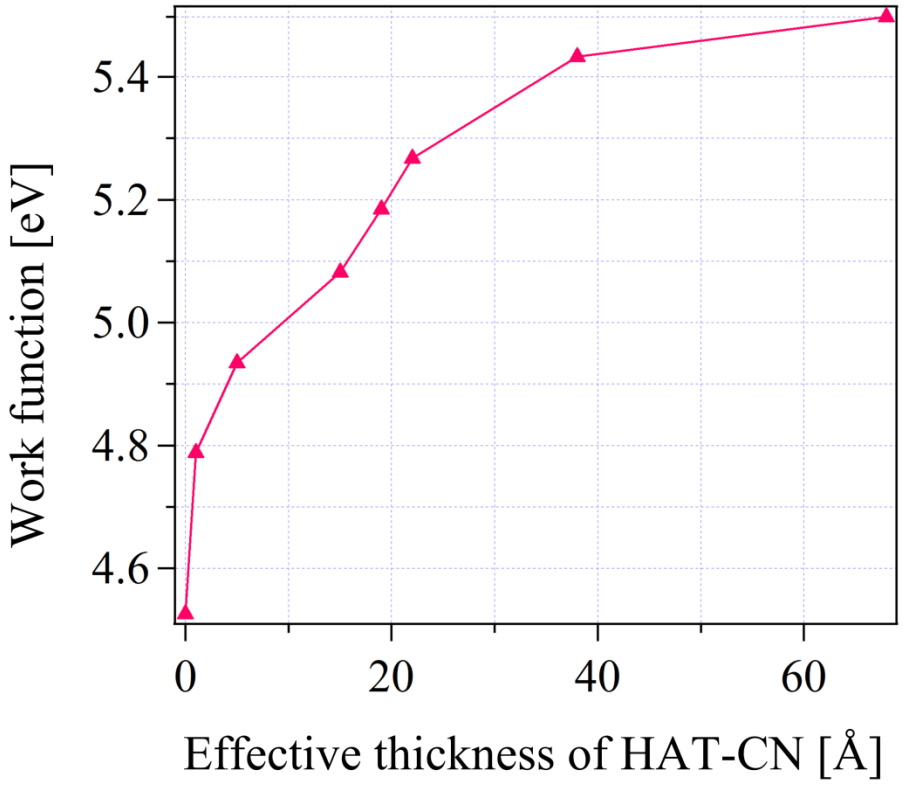

Figure S1. The work function with the different thicknesses of a HAT-CN layer

2. The Raman spectra of HAT-CN/graphene and graphene substrates

To extract the CuPc characteristics from the measured Raman spectra (Figure 1), we also measured the Raman spectra of each substrate. The characteristic peaks from graphene and Si wafer are observed in both spectra, while any remarkable peak from HAT-CN does not exist. To eliminate the orientation-dependent scattering of the substrate, the Raman spectra was taken with circularly-polarized source.

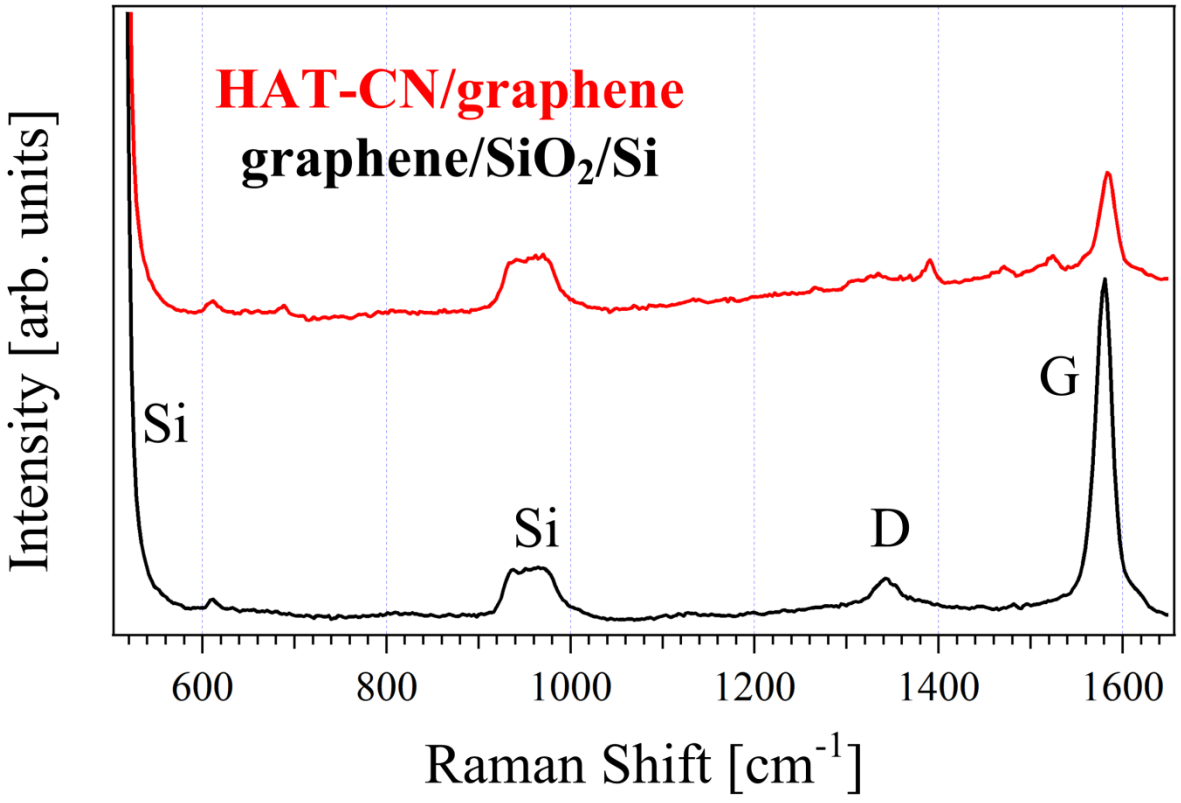

Figure S2. The Raman spectra of HAT-CN/graphene and graphene substrates.

3. Surface morphology conditions

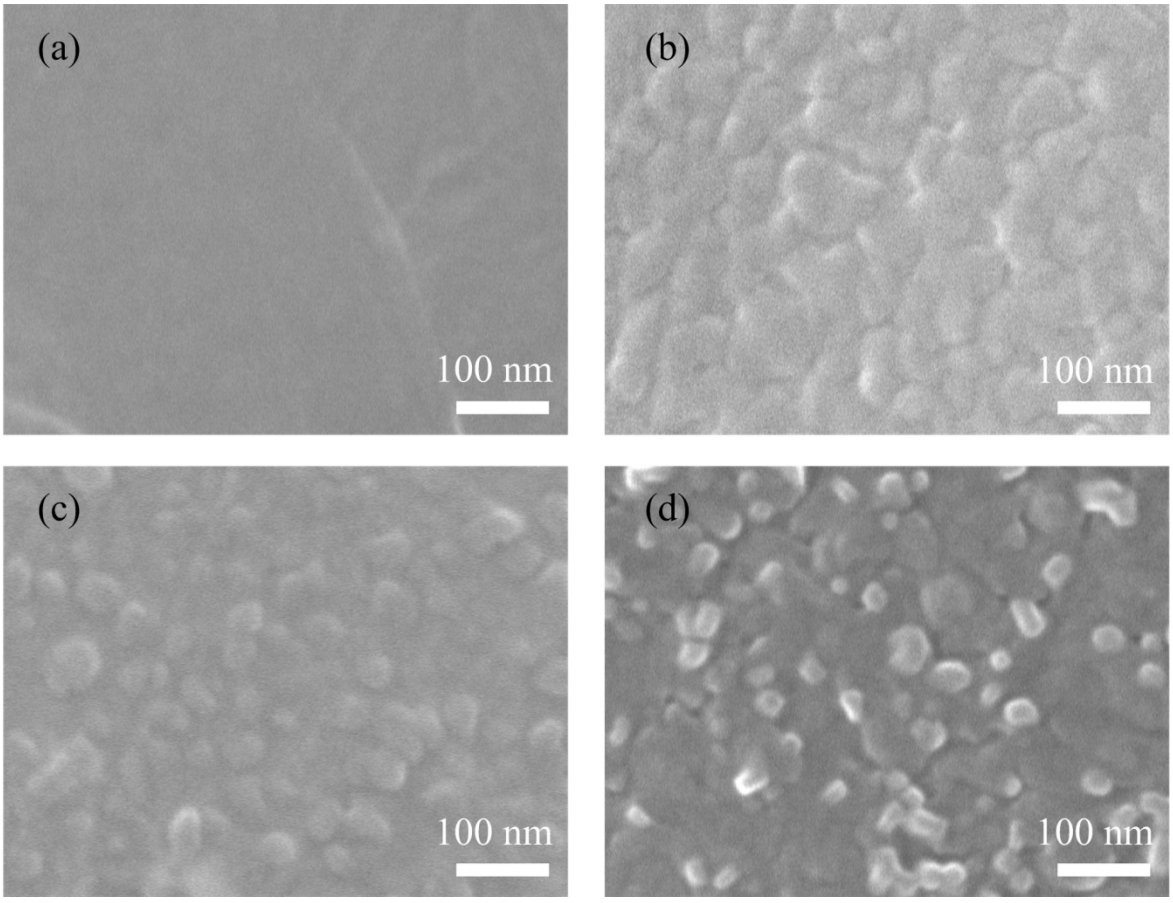

Figure S3. The SEM images of (a) graphene, (b) HAT-CN (68 Å)/graphene, (c) CuPc (30 Å)/graphene, and (d) CuPc (100 Å)/HAT-CN (68 Å)/graphene. (Scale bar: 100 nm)

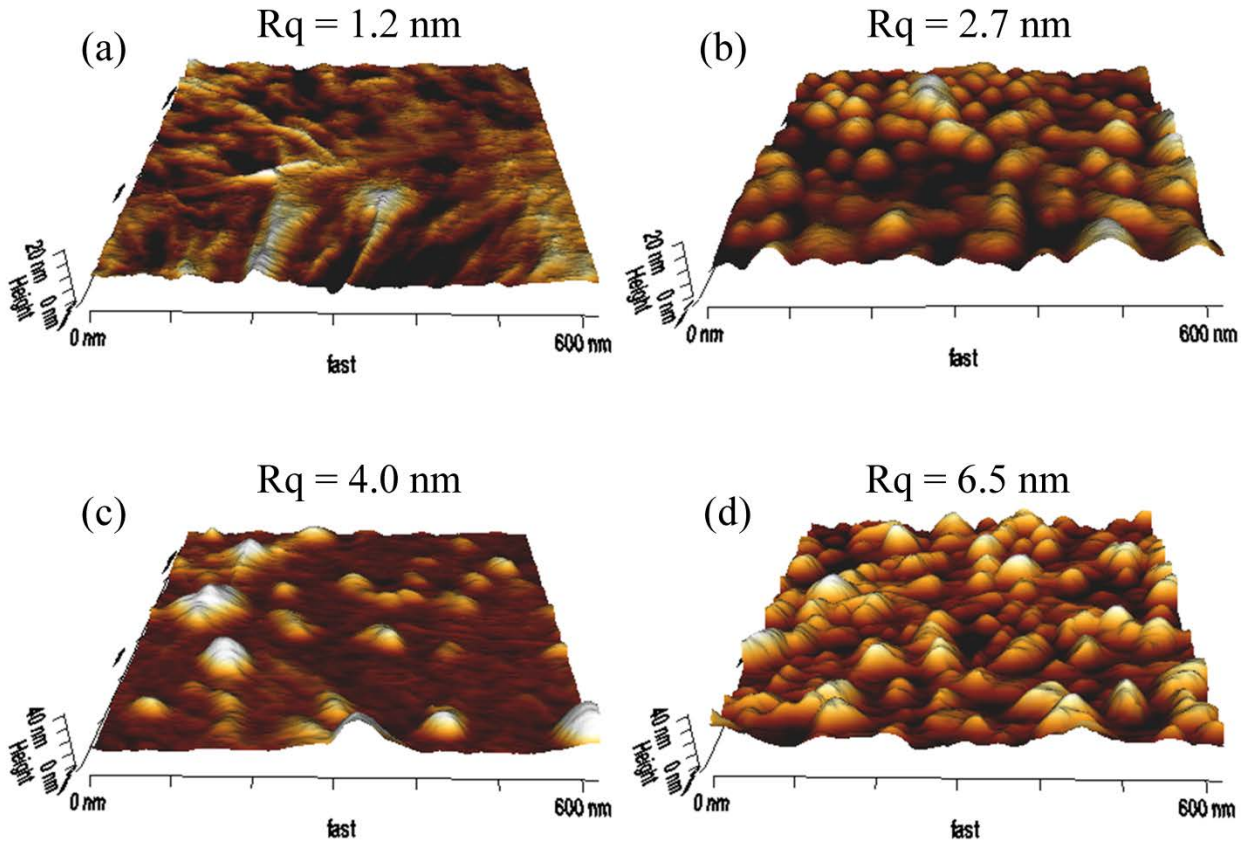

Figure S4 The AFM images of (a) graphene, (b) HAT-CN (68 Å)/graphene, (c) CuPc (30 Å)/graphene, and (d) CuPc (100 Å)/HAT-CN (68 Å)/graphene.

Bare graphene is flat in comparison with HAT-CN/graphene, as shown in Figure S3 and S4. The rough surface of a HAT-CN layer weakens the interaction between graphene and CuPc, leading to the mixed orientation of CuPc molecules.

Reference

(1) Christodoulou, C.; Giannakopoulos, A.; Nardi, M. V.; Ligorio, G.; Oehzelt, M.; Chen, L.; Pasquali, L.; Timpel, M.; Giglia, A.; Nannarone, S.; Norman, P.; Linares, M.; Parvez, K.; Müllen, K.; Beljonne, D.; Koch, N. *J. Phys. Chem. C* **2014**, *118*, 4784–4790.