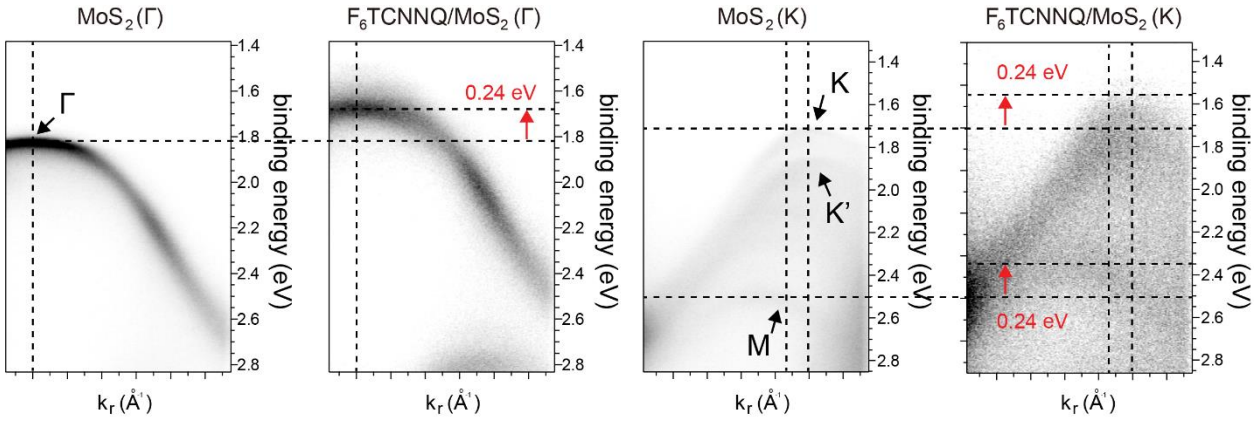
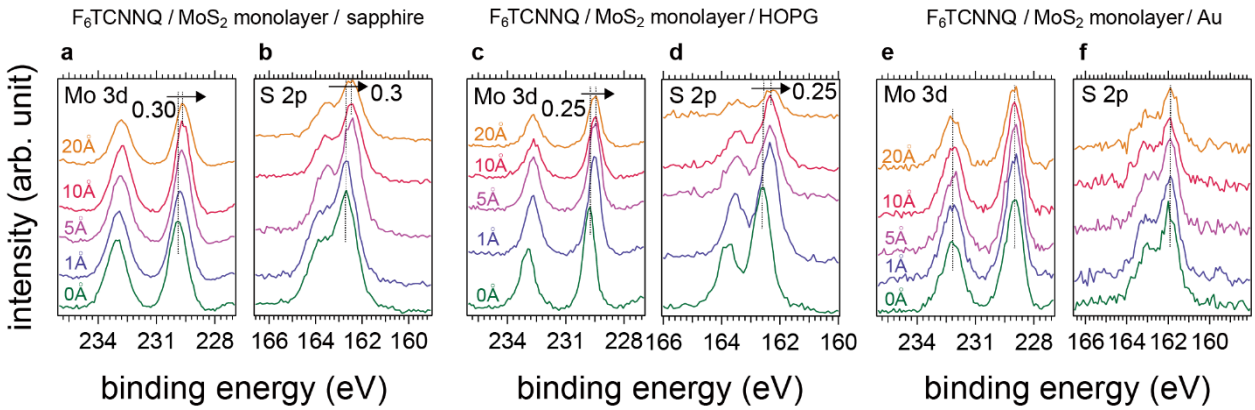
Supplementary Information

Supplementary Figure 1: 2D angle resolved ultraviolet photoelectron spectroscopy (ARUPS) spectra of MoS_2 on highly oriented pyrolytic graphite (HOPG) with/without F_6TCNNQ



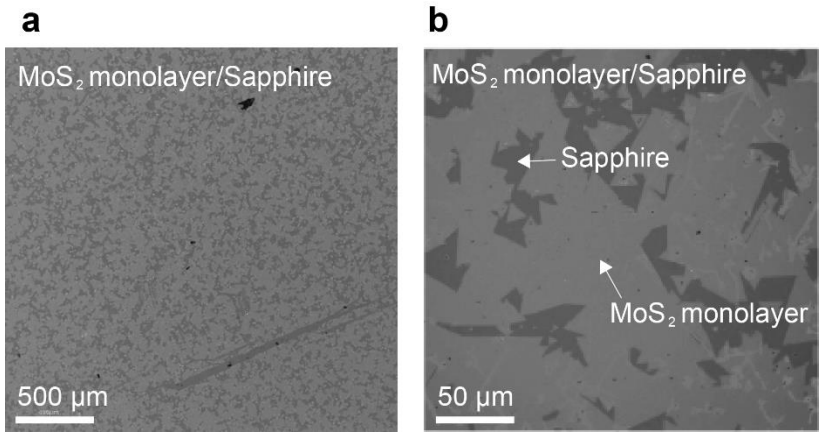
Supplementary Figure 1. Measured angle resolved ultraviolet photoelectron spectroscopy (ARUPS) spectra of MoS_2 monolayer on highly oriented pyrolytic graphite (HOPG) near the Γ and K points (as indicated in the figures), with/without 5Å F_6TCNNQ, respectively.

Supplementary Figure 2: X-ray photoelectron spectroscopy (XPS) spectra



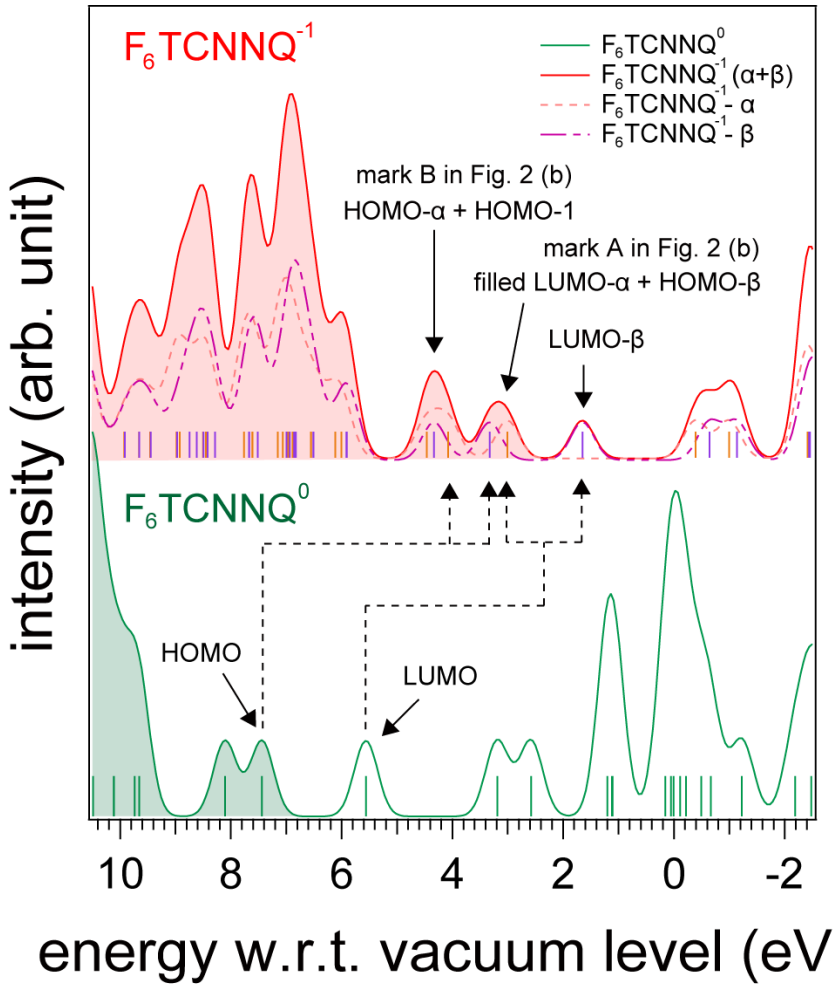
Supplementary Figure 2. Mo 3d and S 2p core level spectra for MoS_2/sapphire (a, b), MoS_2/ highly oriented pyrolytic graphite (HOPG) (c, d), and MoS_2/Au (e, f) heterostructures with different F_6TCNNQ coverage, given in angstrom (Å). The two different core levels shift in parallel, evidencing no site-selective interaction with the molecular acceptor.

Supplementary Figure 3: High-coverage and interconnected MoS_2 monolayer



Supplementary Figure 3. Optical microscopy image of a MoS_2 monolayer on sapphire with (a) low magnification and (b) high magnification. Dark and bright regions indicate the sapphire substrate and MoS_2 monolayer, respectively. Other samples used in this study had a very similar appearance.

Supplementary Note 1: Simulated density of states (DOS) of F_6TCNNQ



Supplementary Figure 4. Calculated density of states of anionic F_6TCNNQ (red curve) and neutral F_6TCNNQ (green curve).

To simulate the density of states of F_6TCNNQ, density functional theory (DFT) calculations were performed on anionic and neutral isolated molecules, using the Gaussian 09 software package.[1] Becke-style three-parameter exchange and Lee-Yang-Parr correlation hybrid functional (B3LYP) with a basis set of 6-311+g were used in geometry optimization and single point energy calculations.[2,3] The results are shown in Supplementary Figure 4.

Supplementary References

1. M. J. Frisch, G. W. Trucks, H. B. Schlegel, G. E. S. *et al.* Cite for Gaussian 09. *Gaussian 09, Revis. B.01, Gaussian Inc., Wallingford, CT* (2009).
2. Becke, A. D. Density-functional thermochemistry. III. The role of exact exchange. *J. Chem. Phys.* **98**, 5648–5652 (1993).
3. Lee, C., Yang, W. & Parr, R. G. Development of the Colle-Salvetti correlation-energy formula into a functional of the electron density. *Phys. Rev. B* **37**, 785–789 (1988).