# Energy level alignment at $C_{60}F_{48}/WSe_2$/Graphite interface

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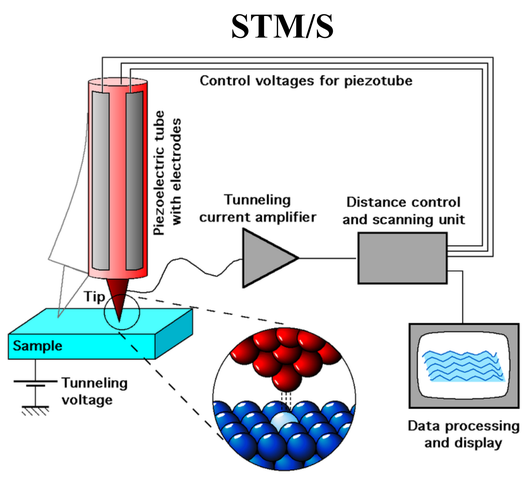
## Introduction

- Recent progress in 2D materials has demonstrated fascinating potential applications in next-generation optical/electrical devices. To achieve controllable tune the carrier type and Fermi level in the 2D materials, the direct deposition of organic molecular layers on the 2D materials is a promising approach to avoid structural damage, thereby retaining their highly desired optical/electrical properties.
- In particular, the interfacial properties, including charge transfer and energy level alignment (ELA) and change in the electronic properties of 2D interlayer upon molecular deposition is still not well understood.
- In this study, organic molecules $C_{60}F_{48}$ was chose due to their higher electron affinity as a potential electron acceptor. The electronic properties of a single-layer $WSe_2$ sandwiched between a graphite substrate and organic molecules $C_{60}F_{48}$ are investigated.

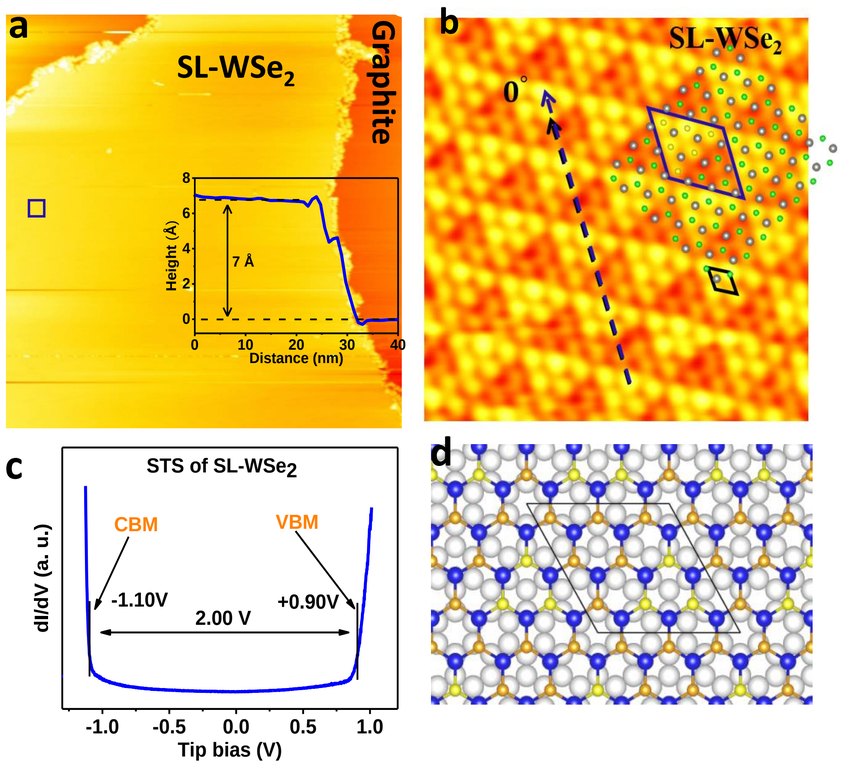
## Methodology




- First-principles method based density functional theory.

## Results

*Monolayer $WSe_2$ on graphite grown by CVD method*

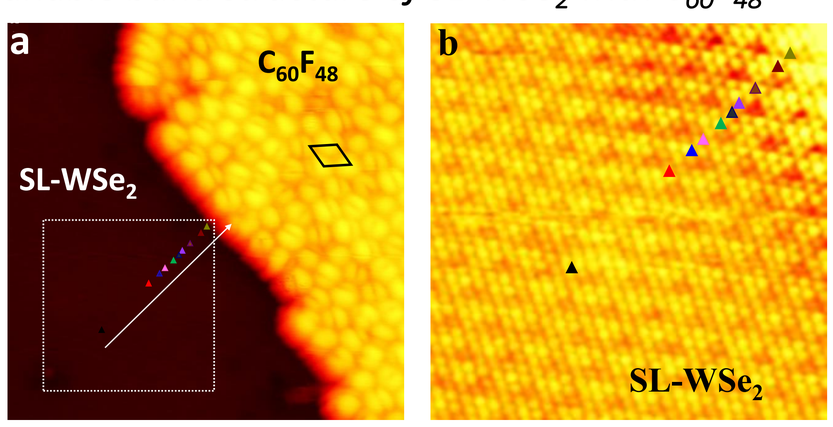



Inserted lateral profile reveals a thickness of ~7 Å for the $WSe_2$ film, which corresponds to the height of a monolayer $WSe_2$. The lateral size of the $WSe_2$ flakes is over hundreds nm to several μm, and bare graphite surface is visible in the large-scale images.

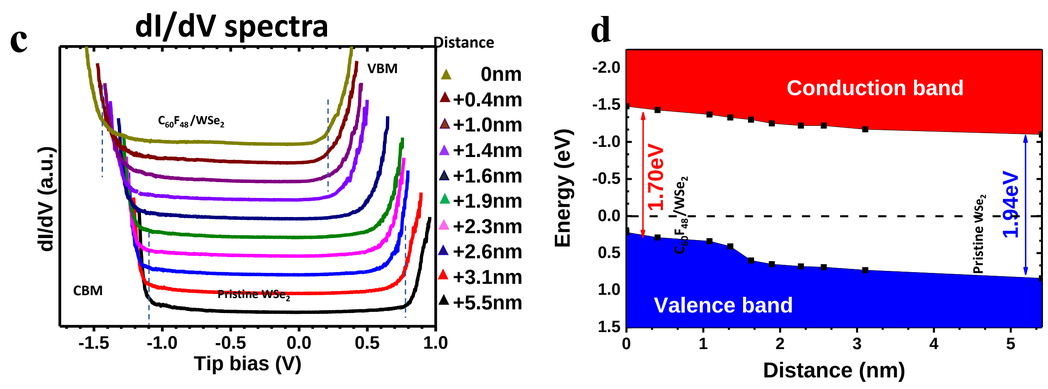
As revealed by the atomically resolved STM image. triangle crystal lattices are assigned to the top-layer Se atoms. The $WSe_2$ unit cell with a lattice constant of 3.4 ± 0.1 Å. Furthermore, an electronic bandgap of 1.95 ± 0.05 eV is obtained from the dI/dV curve.

A typical moiré superstructure arising from the lattice mismatch between graphite and $WSe_2$ is visible. As a proposed structural model, the Se atoms are seated on top of the underlying C atoms (gold color) for bright half and the hollow sites for the dim half.

*Tunable band structure of SL-$WSe_2$ with $C_{60}F_{48}$*



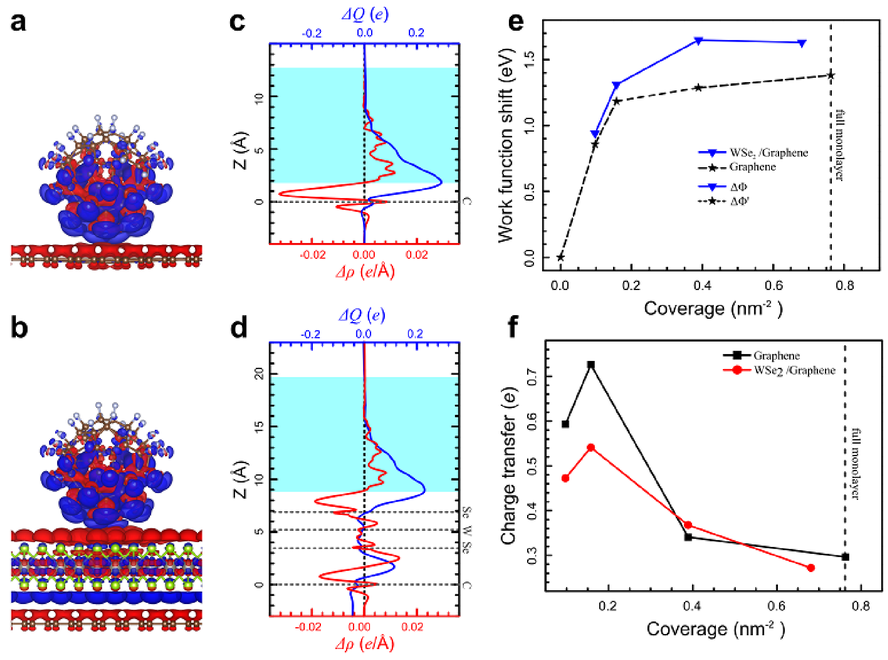

## Results (con't)




In contrast to the uniform $WSe_2$ surface observed far away from the molecular island (or before molecular deposition), a contrast variation is visible close to the island (the upper right corner Fig 2b) indicating that the electronic properties of the underlying $WSe_2$ layer are modified.

To measure the electronic structure changes, a series of STS spectra was taken as a function of distance to the $C_{60}F_{48}$ molecular island edge. Results reveal an upward shift of the VBM relative to the Fermi level is 0.62 eV, while the corresponding shift of the CBM is 0.38 eV, implying a bandgap decrease of 0.24 eV.

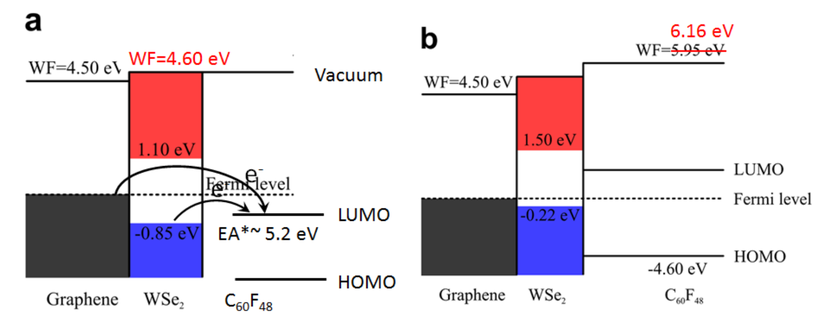
*Charge transfer and work function shifts on different substrates based on DFT calculation.*




The DCD plots reveal that the molecule is negatively charged accumulated at the lower half and the graphite is positively charged for both cases. For the $WSe_2$ interlayer, the top Se layer facing the molecule is hole-accumulated, while the bottom facing graphite is electron-accumulated.

The total amount of charge transfer per molecule ($\Delta Q_{total}$) from bare graphite to $C_{60}F_{48}$ reaches -0.40 *e*, and reduces slightly to -0.37 *e* with the SL-$WSe_2$ interlayer. , The contribution from the $WSe_2$ interlayer is 0.12*e*, which is only half of that from the graphite substrate. In contrast, the corresponding molecular dipole moments are 1.7 *e*·Å for the former and 2.6 *e*·Å for the later.

## Conclusion




(**a**) Energy levels for free graphite covered with a SL-$WSe_2$ and free $C_{60}F_{48}$. (**b**) Energy level realignments after contact. VBM and CBM are determined by STS.