
# *Structure Prediction of Molecular Interfaces*

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***Materials Science & Engineering***

**Carnegie Mellon University**

# Molecular Crystal/ Interface Structure Prediction Tools

**CSP software from Marom's group**

**Estimate molecular solid form volume with PyMoVE**

↓

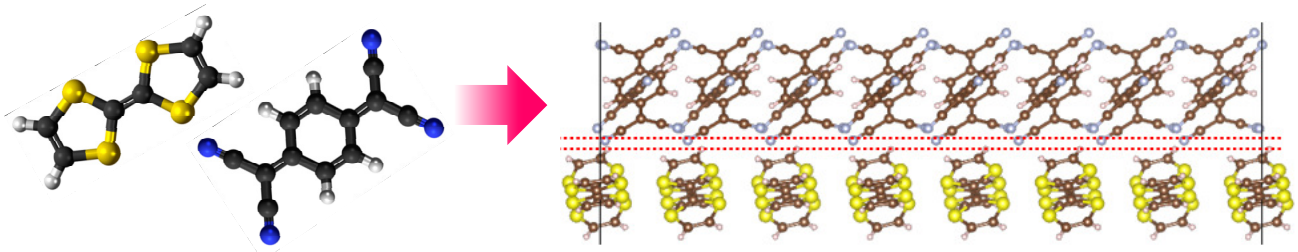
**Generate random structures with Genarris**

↓

**Genetic algorithm (GA) optimization with GAtor**

↓

**Surface energy, morphology, and interface structure prediction with Ogre**



I. Bier and N. Marom, *J. Phys. Chem. A* 124, 10330 (2020)

**Genarris 4.0 coming soon!**

Genarris

Genarris 3.0: Y. Yang *et al. JCTC* 21, 11318 (2025)

FastCSP: V. Gharakhanyan *et al. arXiv* 2508.02641 (2025)

Genarris Interfaces: H. Ni *et al. JCTC* 22, 4835 (2026)

**GAtor 2.0 coming soon!**

GAtor

F. Curtis *et al. JCTC* 14, 2246 (2018)

R. Tom *et al. Chem. Mater.* 35, 1373 (2023)

Ogre

**Ogre 2.0 coming soon!**

S. Yang *et al. J. Chem. Phys.*, 152, 244122 (2020)

S. Moayedpour *et al. J. Phys. Chem. C* 127, 10398 (2023)


# Structure Prediction of Epitaxial Organic Interfaces with Ogre

# Structure Prediction of Epitaxial Organic Interfaces

Interfaces between organic materials perform critical functions in organic electronic devices

In analogy with inorganic materials, ordered crystalline interfaces may lead to improved device performance

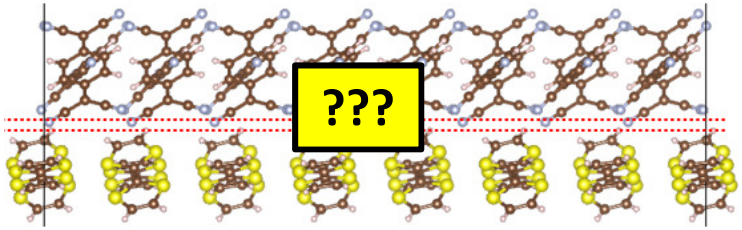
Ordered interfaces are more conducive to electronic and optical characterization

Epitaxial templating can be used to promote the growth of desired polymorphs

TTF-TCNQ = quintessential donor-acceptor pair

Epitaxial interface grown and studied by UPS without structural characterization

B. Kattel, T. Wang, T. R. Kafle, W.-L. Chan *Organic Electronics* 48, 371 (2017)




gre

S. Moayedpour, I. Bier, W. Wen, D. Dardzinski, O. Isayev, N. Marom *J. Phys. Chem. C* 127, 10398 (2023)

Select substrate facet(s) based on surface energy

Lattice matching with optional Miller index scan

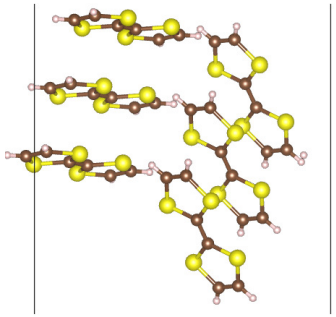
Surface matching for lowest misfit interfaces

Energy ranking

Electronic structure calculations for most stable interface(s)

# Calculating Surface Energies with Ogre

**Surface energy is used to select the most stable substrate facets**

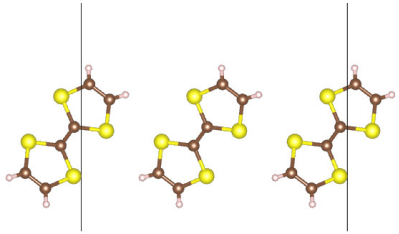


**The surface energy is given by:**

$$\gamma = \lim_{N\to\infty} \frac{1}{2A}(E_{slab} - NE_{bulk})$$

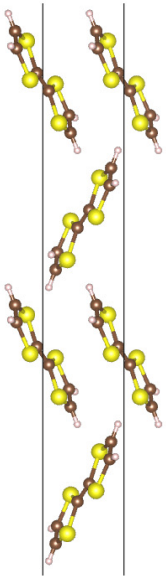
**The linear method:**

$$E_{slab} = NE_{bulk} + 2A\gamma$$

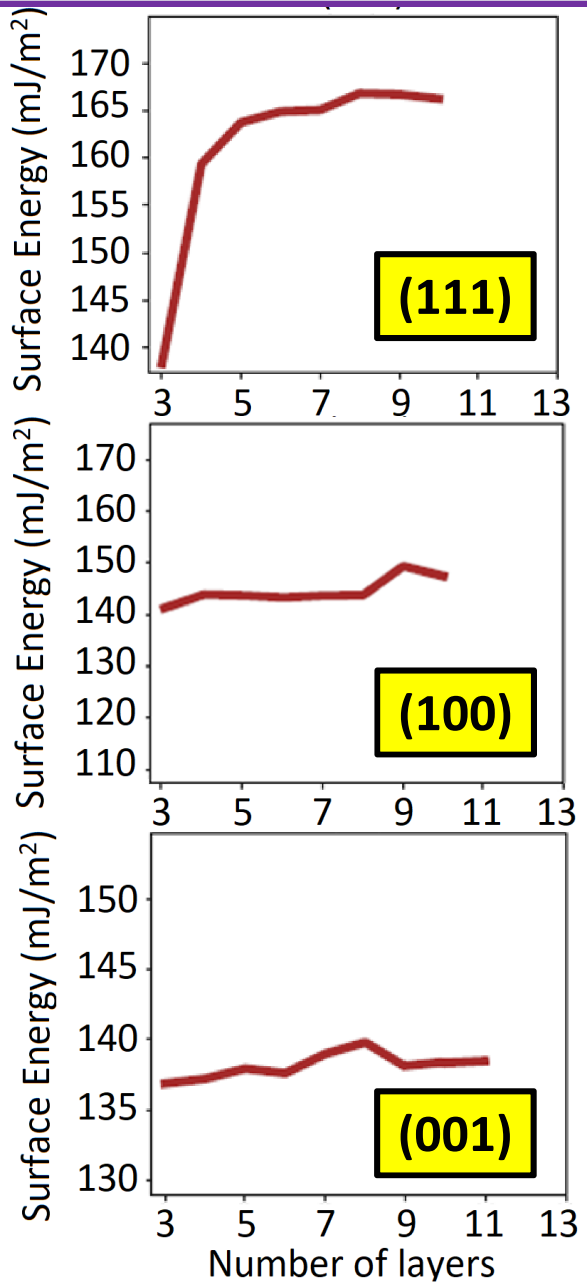


**Surface energy of molecular crystals depends on the strength of intermolecular interactions being cleaved and the chemical groups exposed on the surface**

**Most stable facets of TTF: (001),(100),(011) with H atoms on surface, not π-system**




S. Moayedpour, I. Bier, W. Wen, D. Dardzinski, O. Isayev, and N. Marom *J. Phys. Chem. C* 127, 10398 (2023)

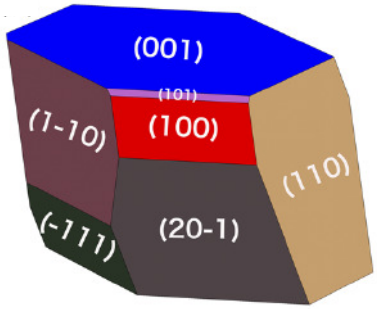



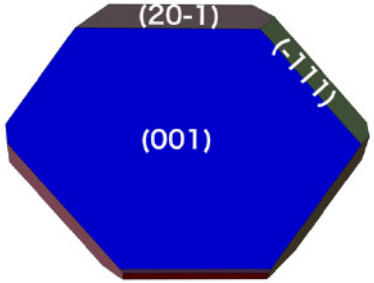
# Constructing Wulff Shapes with Ogre

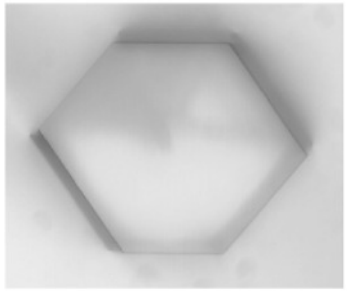
**The Wulff construction predicts the equilibrium shape of a crystal by minimizing the surface energy**

**The Wulff shapes produced by Ogre (PBE+MBD) are in good agreement with experiment for aspirin and tetracene**

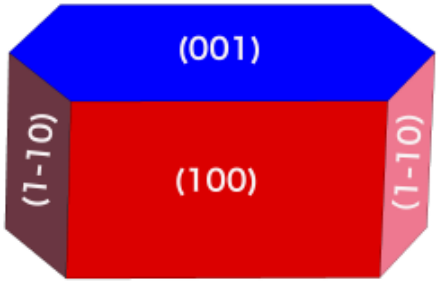
**Tetracene**

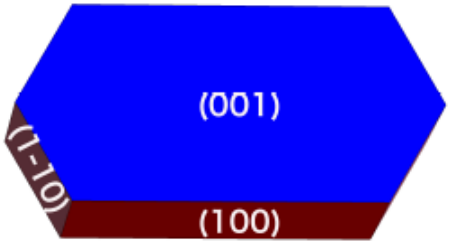


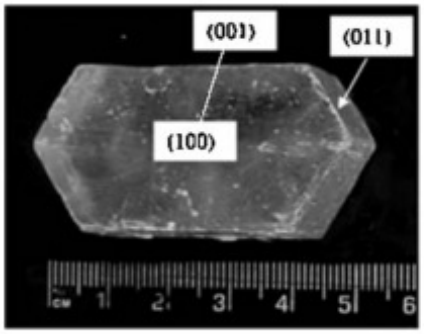



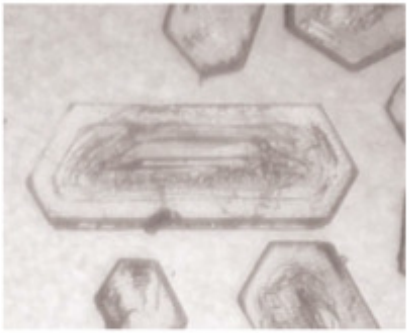



**Aspirin**



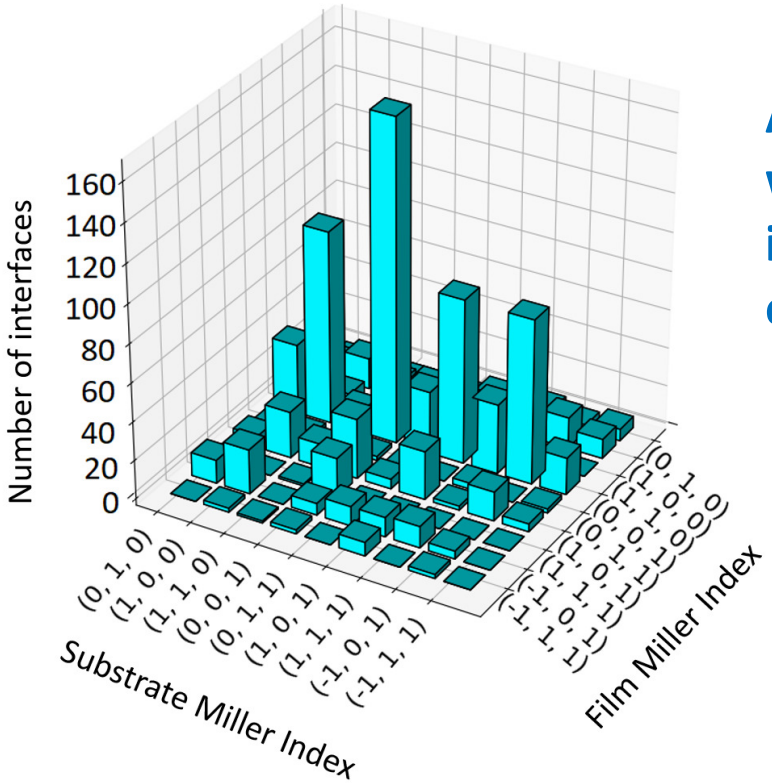



S. Yang, I. Bier, W. Wen, J. Zhan, S. Moayedpour, and N. Marom,
*J. Chem. Phys.* 152, 244122 (2020)

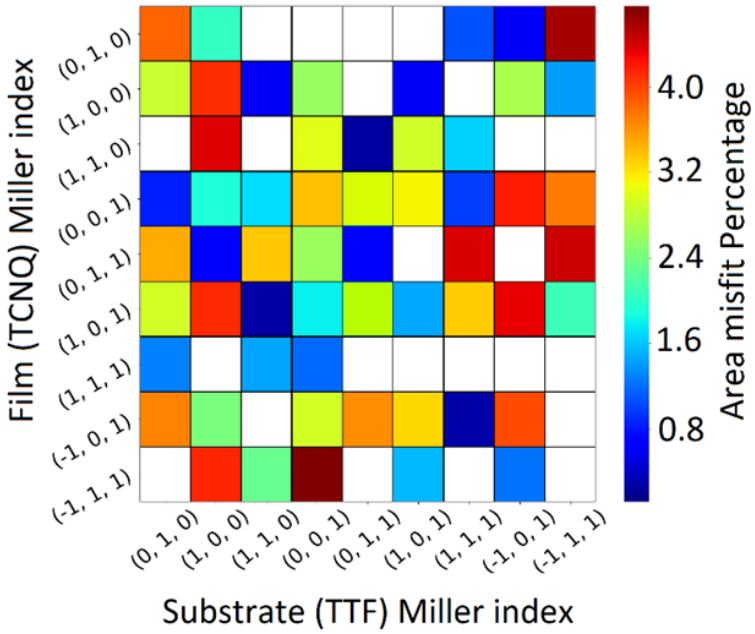
# Ogre: Lattice Matching

**The lattice matching step produces domain-matched interfaces, where commensurability is achieved with different integer multiples of the substrate and film unit cells within a given area and misfit tolerance**

A. Zur and T. C. McGill,
*J. Appl. Phys.* 55, 378 (1984)




**A Miller index scan for the TCNQ/TTF interface with a maximal Miller index of 1, maximum interface area of 1000 Å$^2$, and misfit tolerance of 5% produces 965 domain-matched structures**




**57 Miller index combinations with misfit < 5%**

**Narrow down to most stable TTF facets and misfit < 2.5%**

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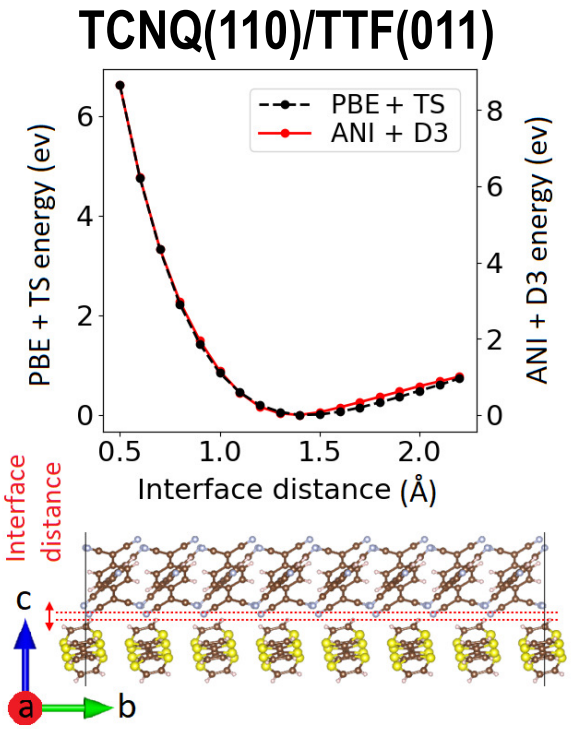
D. Dardzinski, O. Isayev, N. Marom *J. Phys. Chem. C* 127, 10398 (2023)

# Ogre: Surface Matching

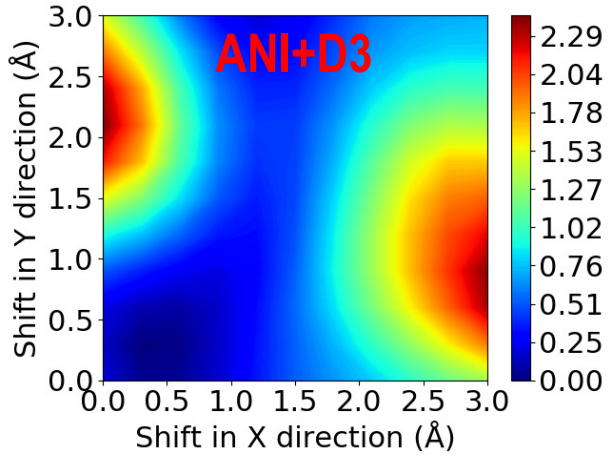


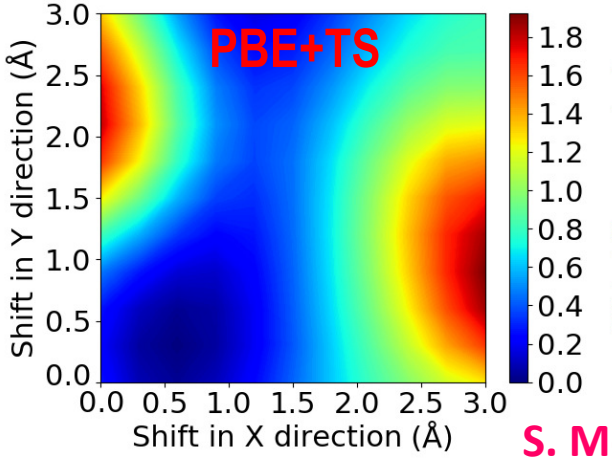

**The surface matching step finds the optimal position of the film on top of the substrate**

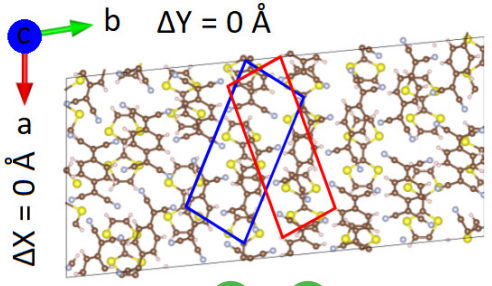
**Bayesian optimization is used with an objective function based on the ANI deep neural network interatomic potentials with the Grimme D3 dispersion correction**

ANI: J. S. Smith, O. Isayev, A. E. Roitberg *Chem. Sci.* 8, 3192 (2017)

**ANI+D3 is in good agreement with DFT for the optimal interfacial distance and in-plane registry**

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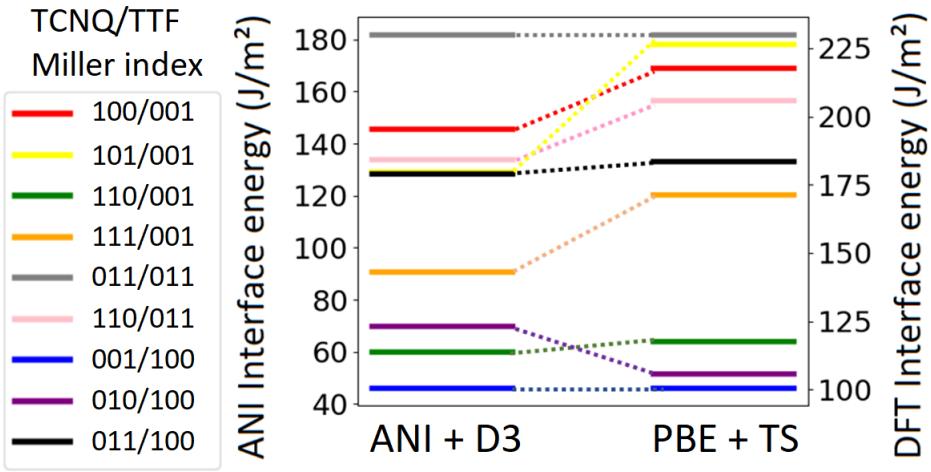
# Ogre: Ranking and Electronic Structure

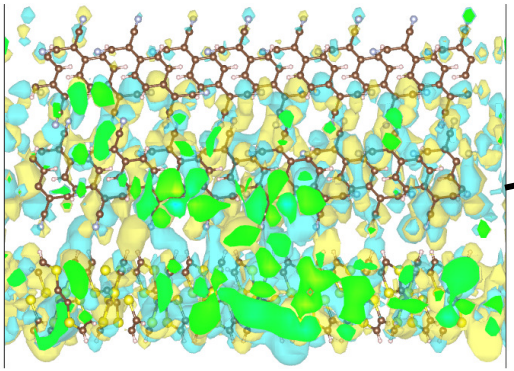
**Interface energy:** $\sigma = \gamma_{sub} + \gamma_{film} - W_{ad}$ $\quad W_{ad} = \frac{1}{A}(E_{sub} + E_{film} - E_{int})$

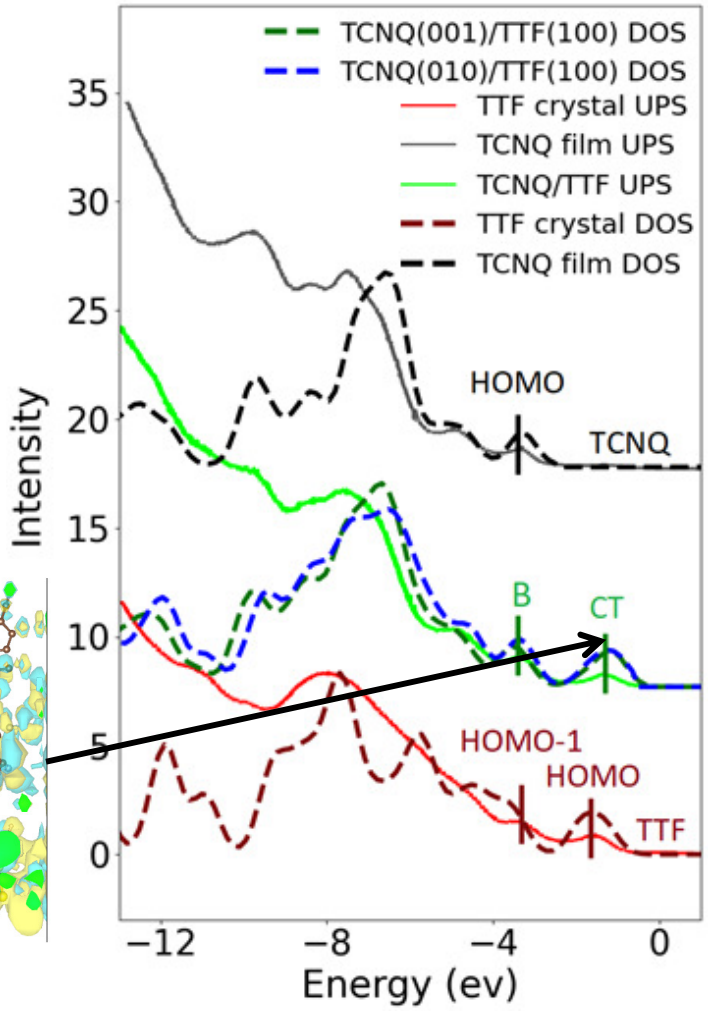
**ANI+D3 correctly identifies the most stable interface structures**




**The HSE DOS is in excellent agreement with UPS experiments**

**A charge transfer (CT) interface state is found in agreement with experiment**



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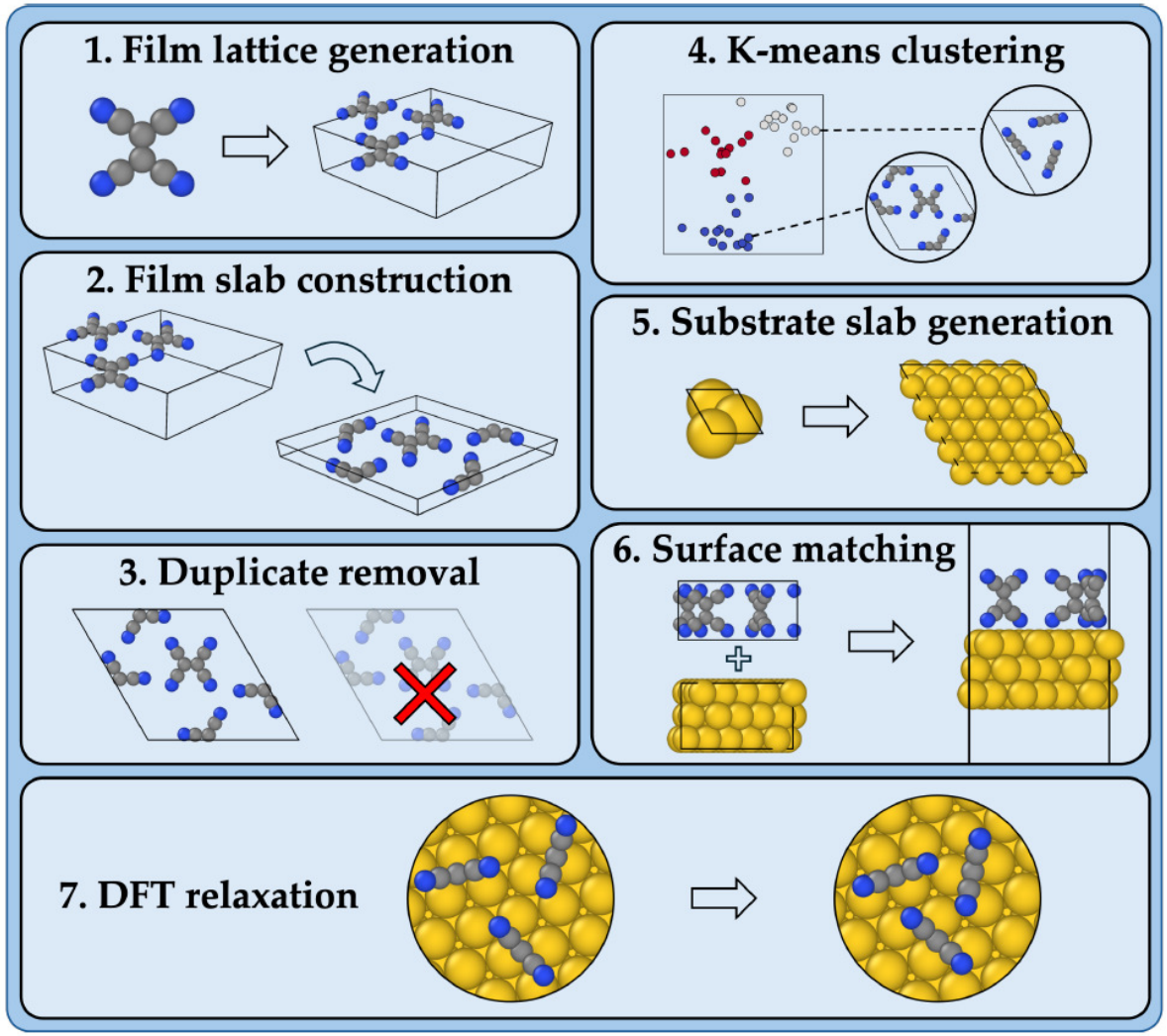
# Structure Prediction of Organic/Inorganic Interfaces with Genarris Interfaces

# Interface Structure Prediction with Genarris Interfaces




H. Ni, K. Larkin, W. Wen, S. Moayedpour, R. Tom, I. Bier, D. Dardzinski, and N. Marom *JCTC* 22, 4835 (2026)

Interfaces of organic active layers with inorganic electrodes are important for organic electronics

Interaction with substrate can induce formation of "surface polymorphs"

Molecular film structures are generated in all compatible layer groups

Epitaxy matrices are used to impose commensurism

No assumptions are made about the binding site

Films with varying surface coverage are generated

# Film Structure Generation with Genarris

**Inputs**

- Number of molecules per unit cell (Z)
- *x*-*y* lattice vectors of the substrate

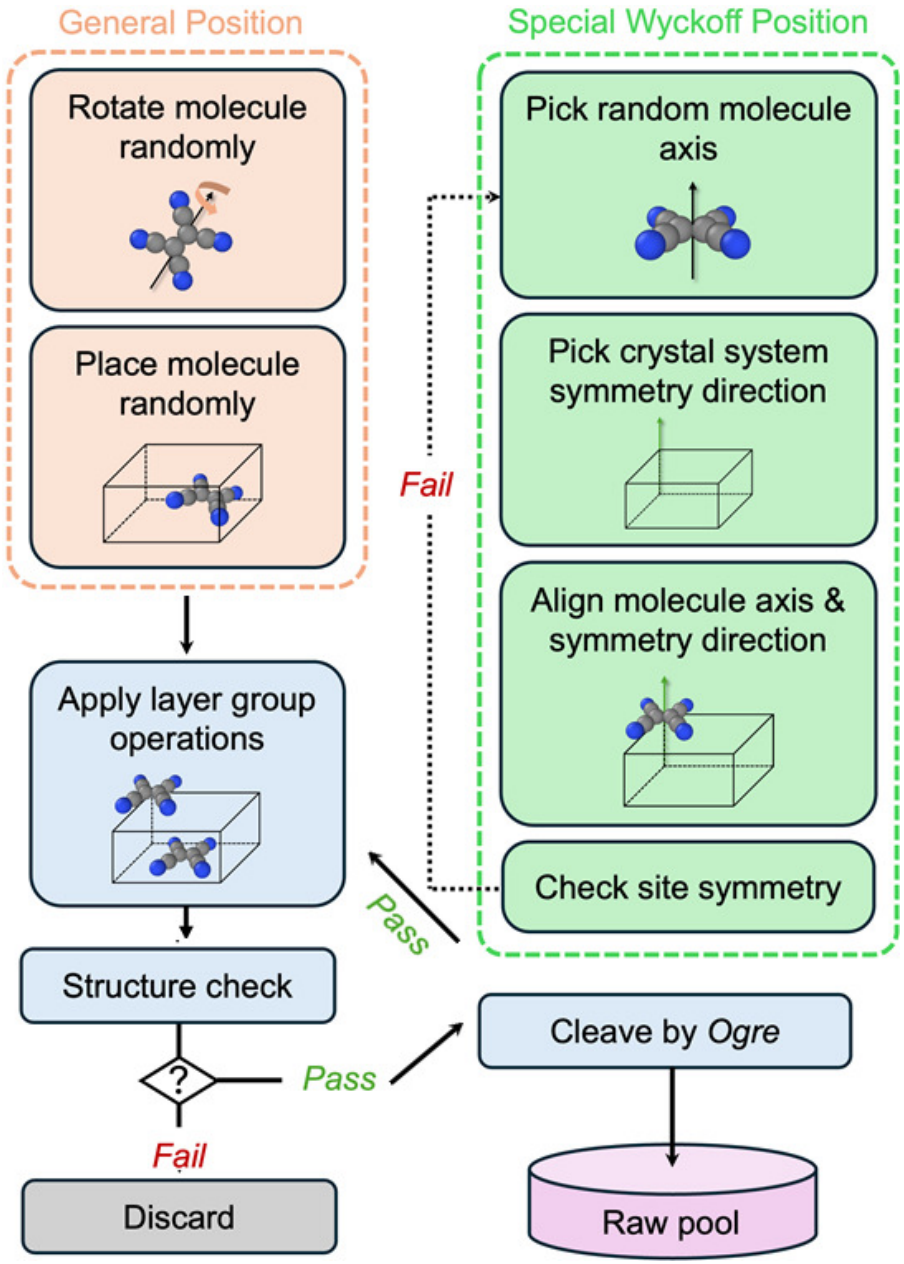
**Set Constraints**

- Estimate volume of the film unit cell
- Set maximum surface area
- Determine compatible layer groups

**Under the constraints, do:**

- Construct compatible epitaxy matrices
- Determine the *z* lattice vectors

Film unit cells

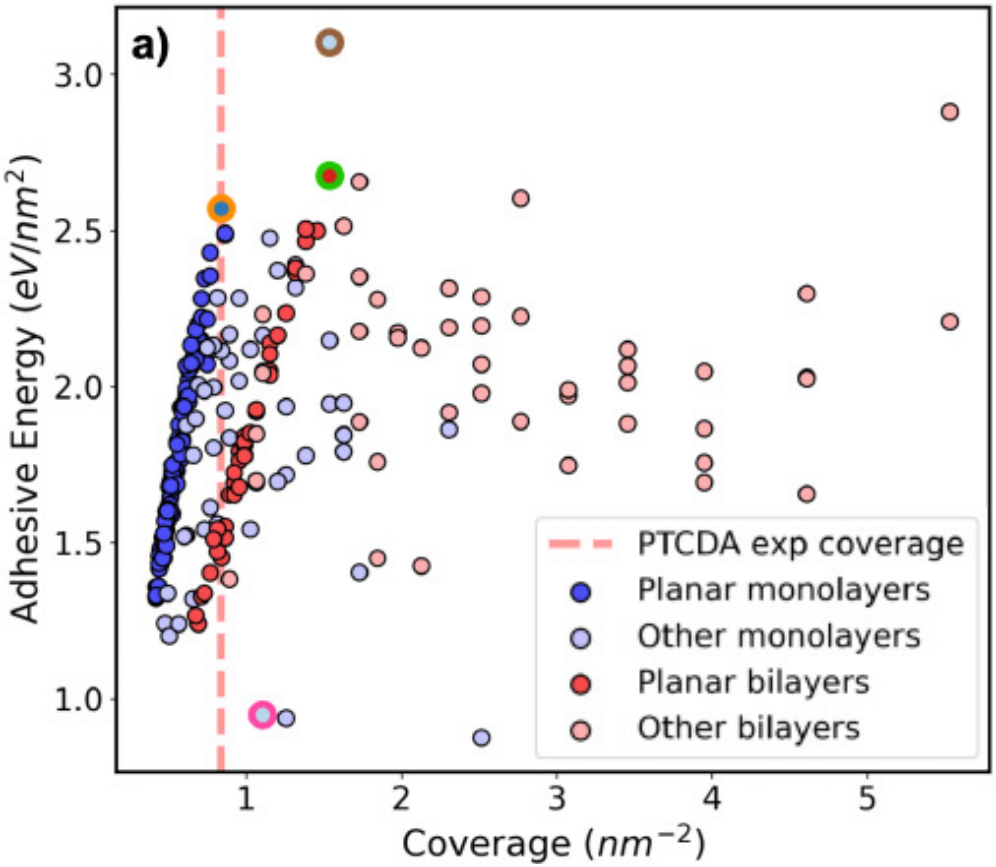


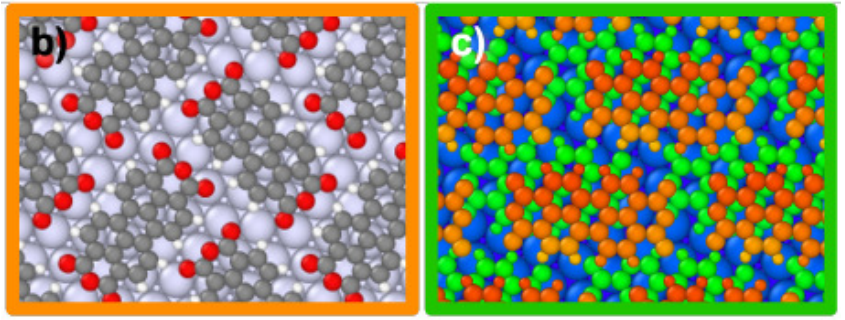

**Film structures are generated like molecular crystal structures but using layer groups instead of space groups**

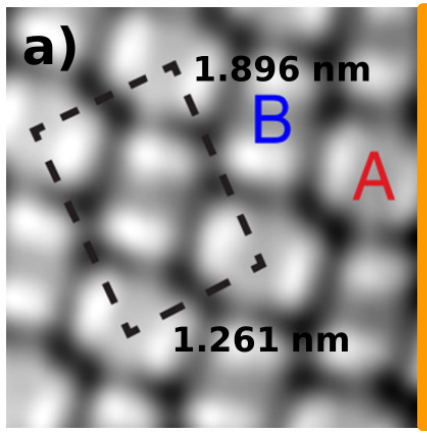
# Genarris Interfaces: PTCDA/Ag(111) Z=2

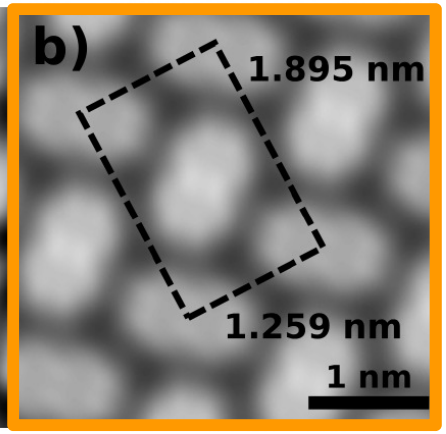
**Planar monolayer structures are favorable at low coverage**

**The best match to the experimental structure is the most stable for the corresponding coverage**



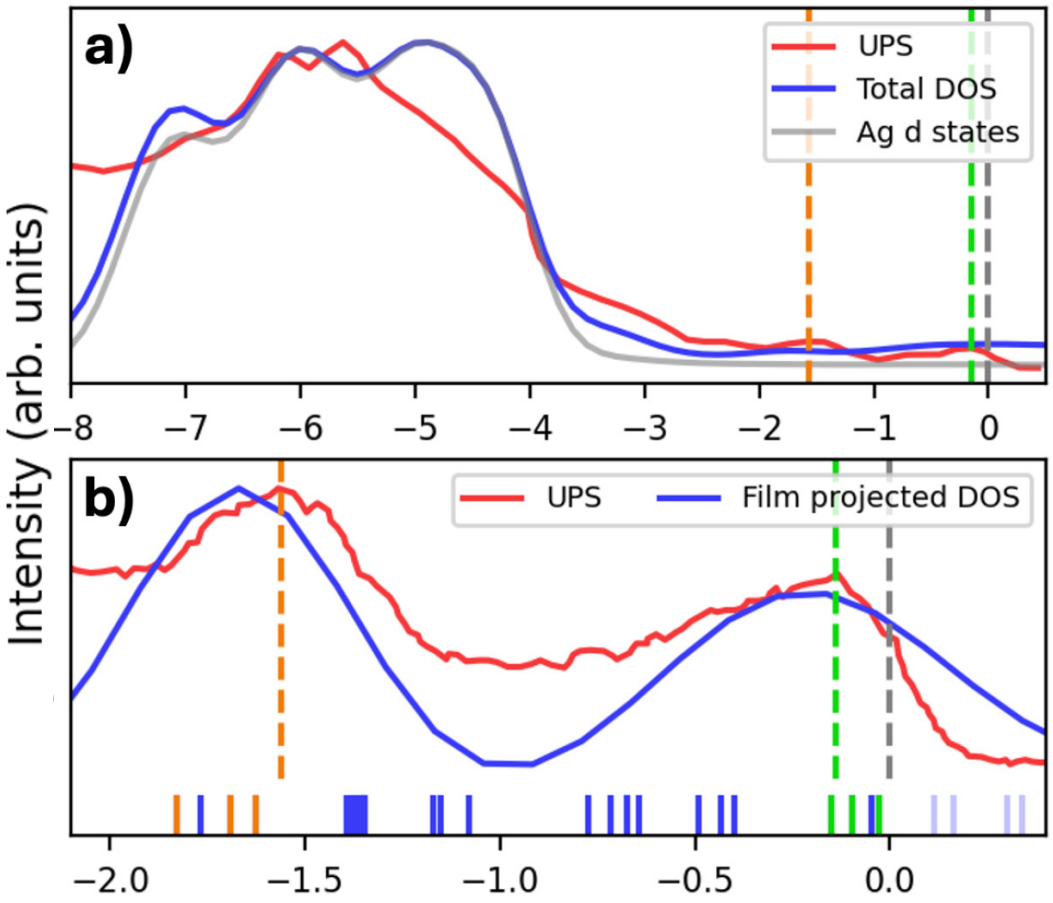



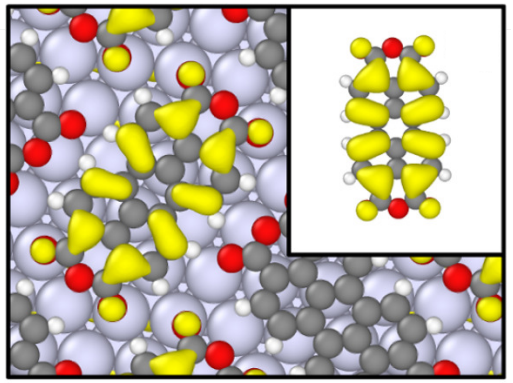

H. Ni, K. Larkin, W. Wen, S. Moayedpour, R. Tom, I. Bier, D. Dardzinski, and N. Marom *JCTC* 22, 4835 (2026)

# Electronic Structure of PTCDA/Ag(111)

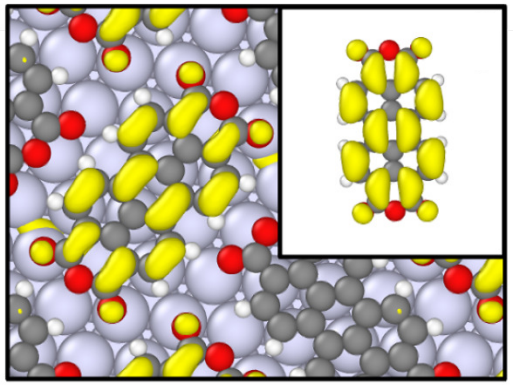
**The DOS of the interface structure generated by Genarris is in good agreement with UPS experiment**

**The LUMO-derived states become occupied upon adsorption**



**HOMO-derived state**

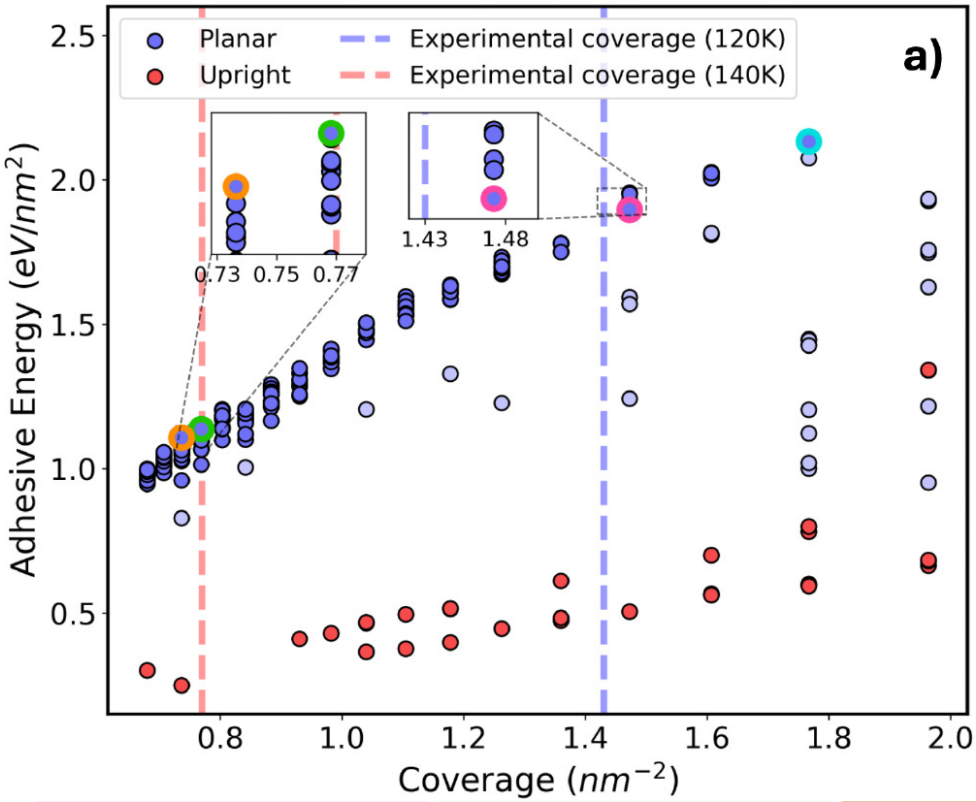


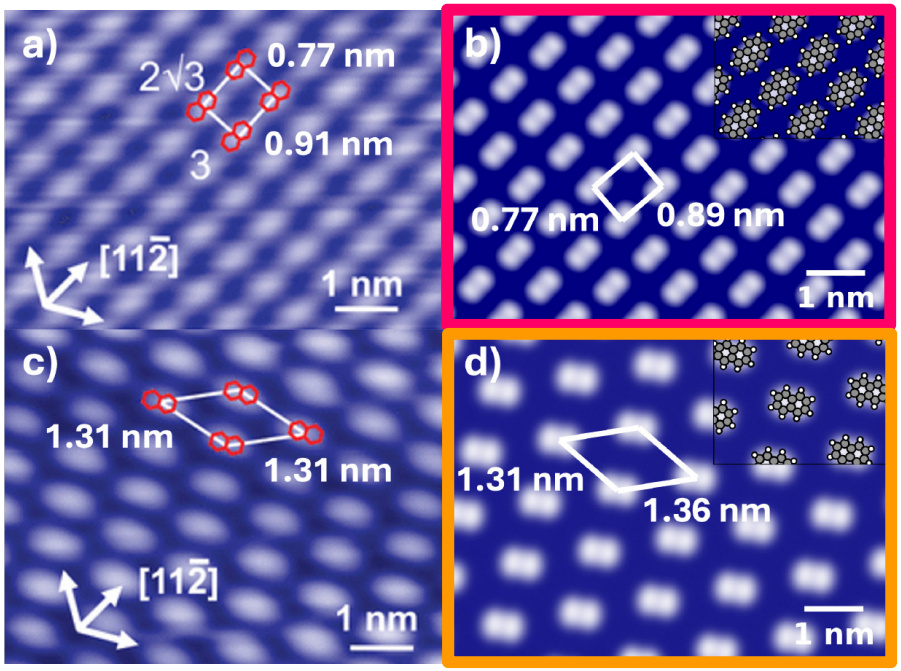
**LUMO-derived state**

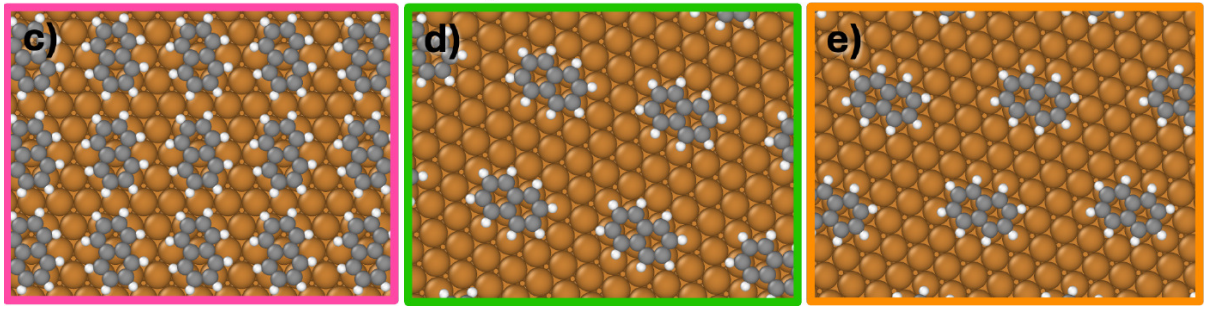
H. Ni, K. Larkin, W. Wen, S. Moayedpour, R. Tom, I. Bier, D. Dardzinski, and N. Marom *JCTC* 22, 4835 (2026)

# Genarris Interfaces: Naphthalene/Cu(111) Z=1

## Matches to both Z=1 polymorphs are generated close to the experimental coverage



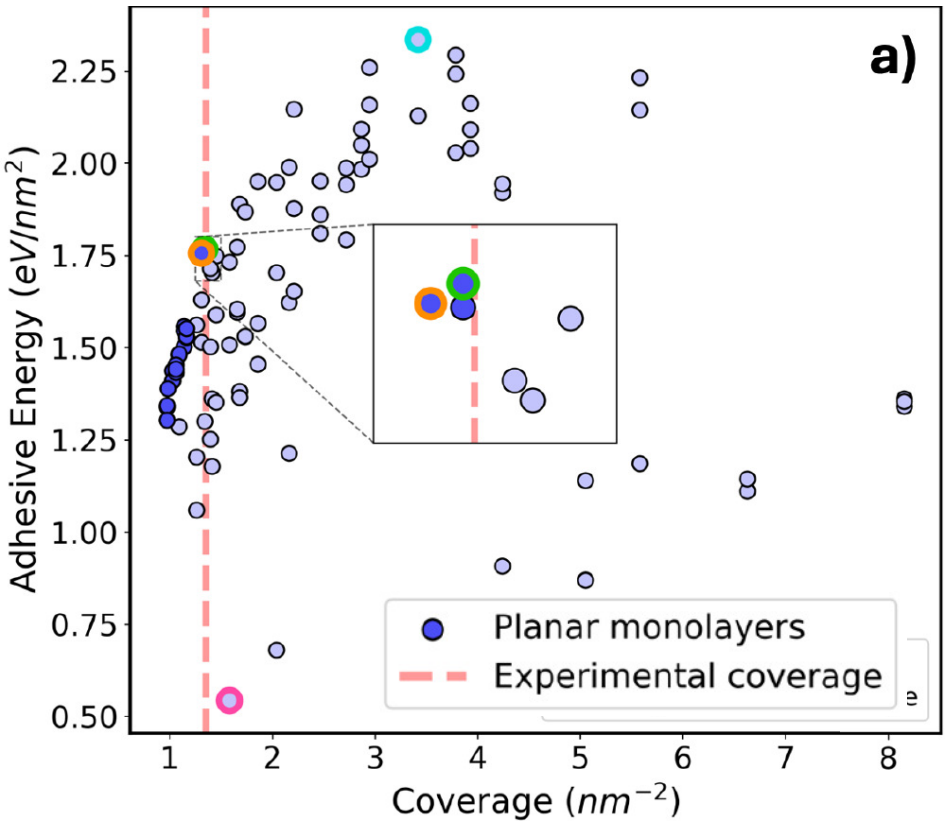



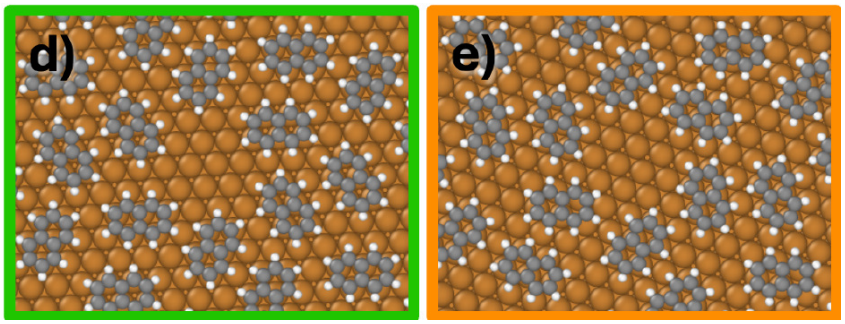



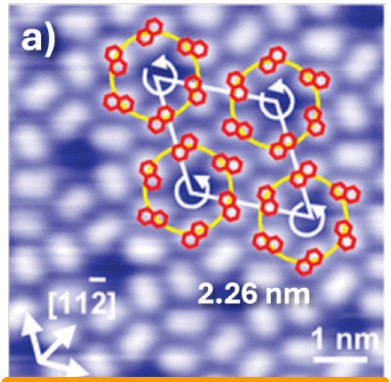


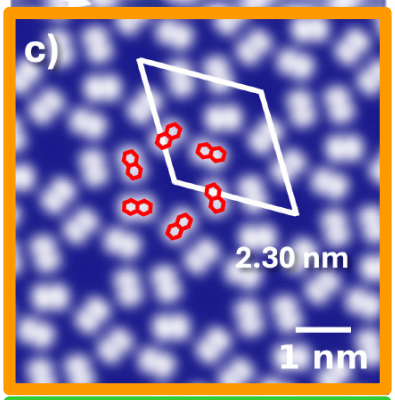
H. Ni, K. Larkin, W. Wen, S. Moayedpour, R. Tom, I. Bier, D. Dardzinski, and N. Marom *JCTC* 22, 4835 (2026)

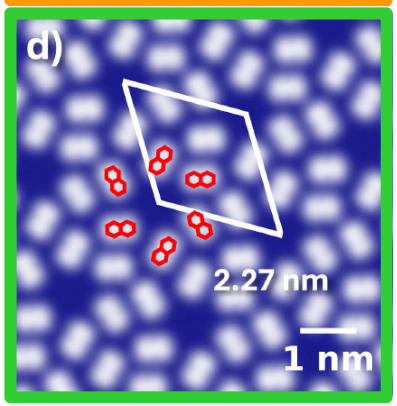
# Genarris Interfaces: Naphthalene/Cu(111) Z=6



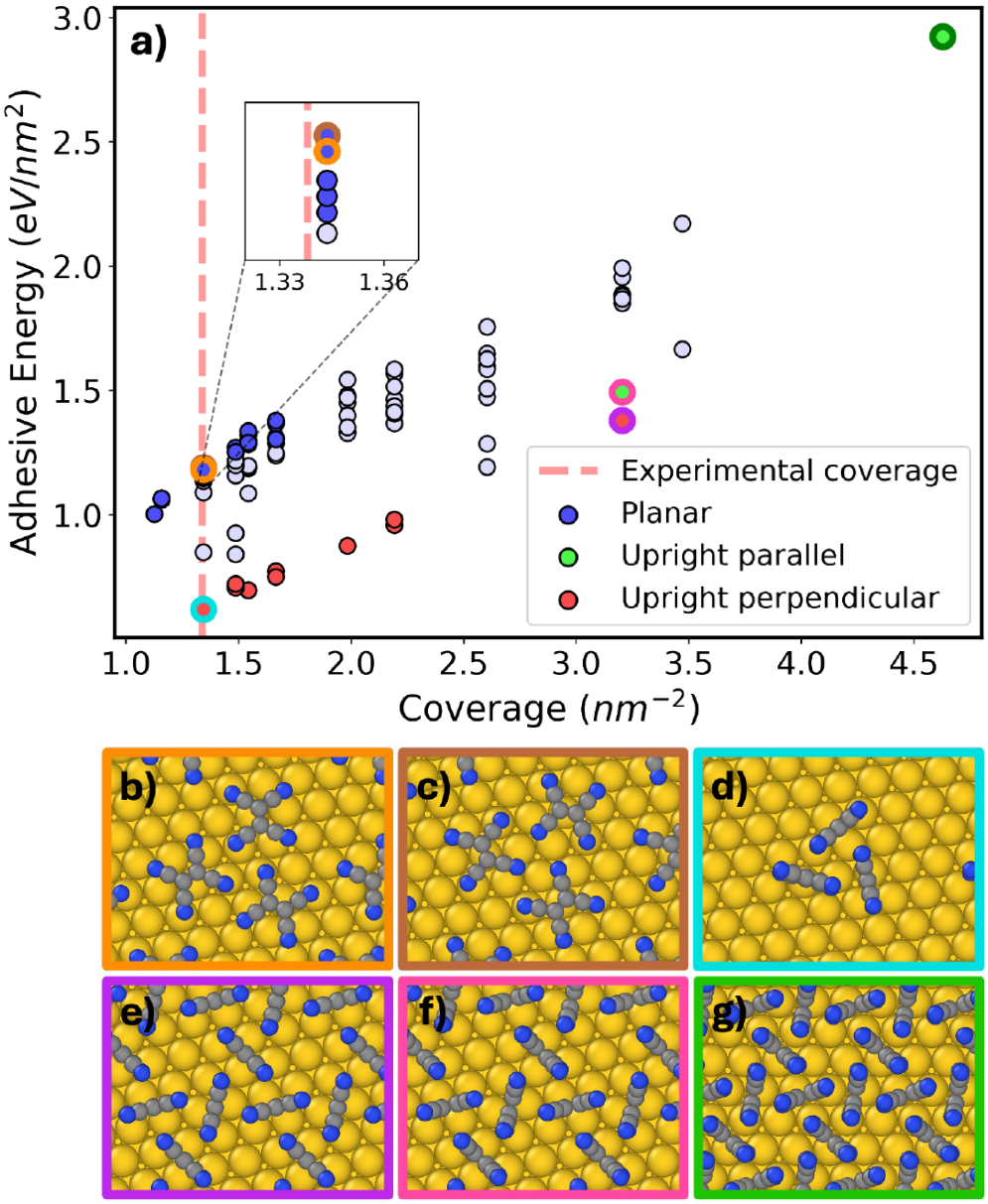



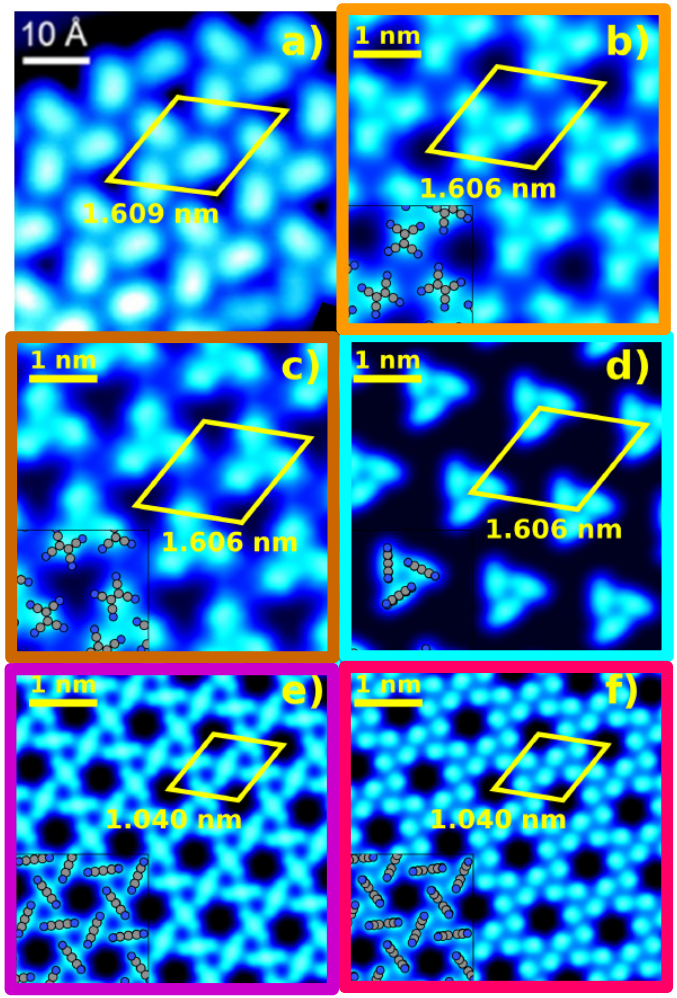

**Matches to the Z=6 polymorphs are generated at/close to the experimental coverage**

H. Ni, K. Larkin, W. Wen, S. Moayedpour, R. Tom, I. Bier, D. Dardzinski, and N. Marom *JCTC* 22, 4835 (2026)

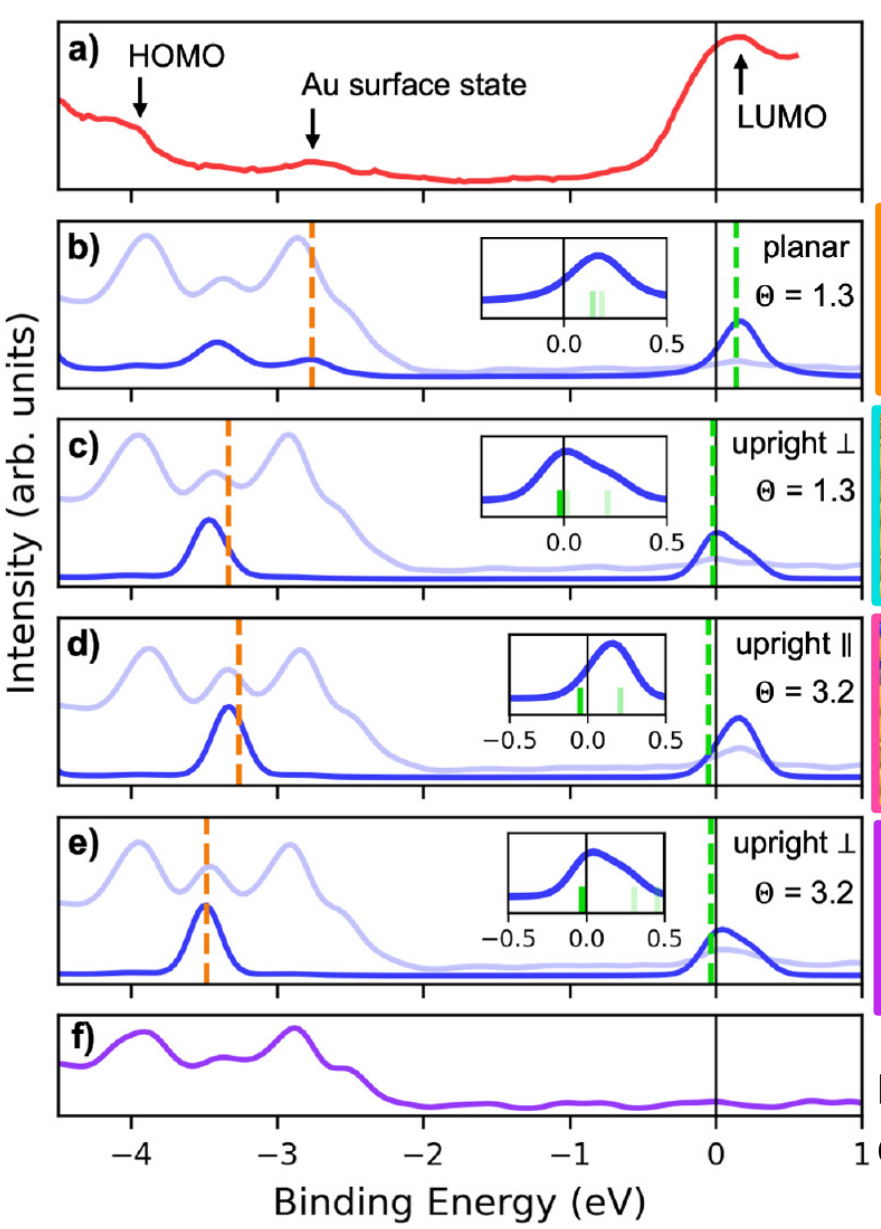
# Genarris Interfaces: TCNE/Au(111) Z=3

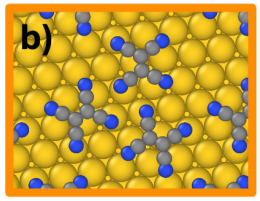


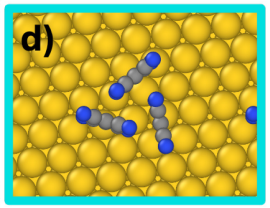

**Planar structures resemble experiment; Upright structures become stable only at high coverage**

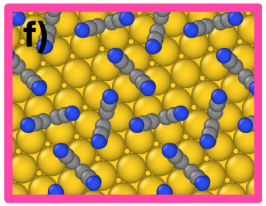


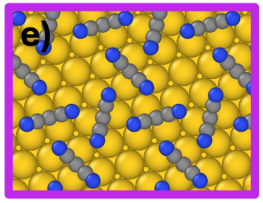
# Electronic Structure of TCNE/Au(111)



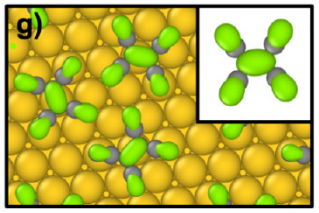



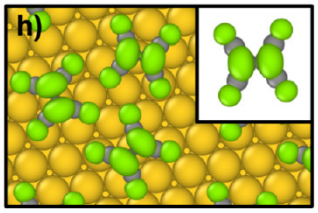




**It can be difficult to distinguish between different adsorption modes based on spectral signatures**

H. Ni, K. Larkin, W. Wen, S. Moayedpour, R. Tom, I. Bier, D. Dardzinski, and N. Marom *JCTC* 22, 4835 (2026)

**HOMO-derived state**




**LUMO-derived state**

# Conclusion & Acknowlegements

**Ogre can be used to predict the surface energy and morphology of molecular crystals**

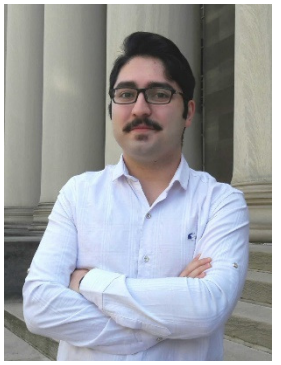
Ogre: S. Yang *et al.* *J. Chem. Phys.*, 152, 244122 (2020)

Shuyang Yang

**Ogre can be used to predict the structure of epitaxial organic or inorganic interfaces**

Ogre Organic: S. Moayedpour *et al*. *JPCC* 127, 10398 (2023)

Ogre Inorganic: D. Dardzinski *et al*. *ACS Nano* 127, 10398 (2025)



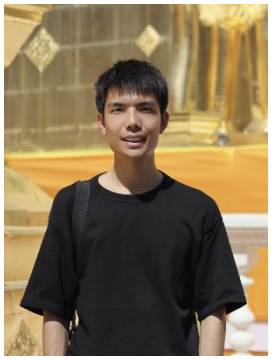
Saeed Moayedpour

**Ogre 2.0 coming soon!! Organic and inorganic interfaces in a unified code base+ MLIP support**

Derek Dardzinski

**Genarris Interfaces can be used to predict the structure of molecular films on surfaces**

Genarris Interfaces: H. Ni *et al*. *JCTC* 22, 4835 (2026)



Haoran Ni



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