SuperSuper: Alternative Superconducting Superlattices based on Covalent Organic Frameworks (COFs)

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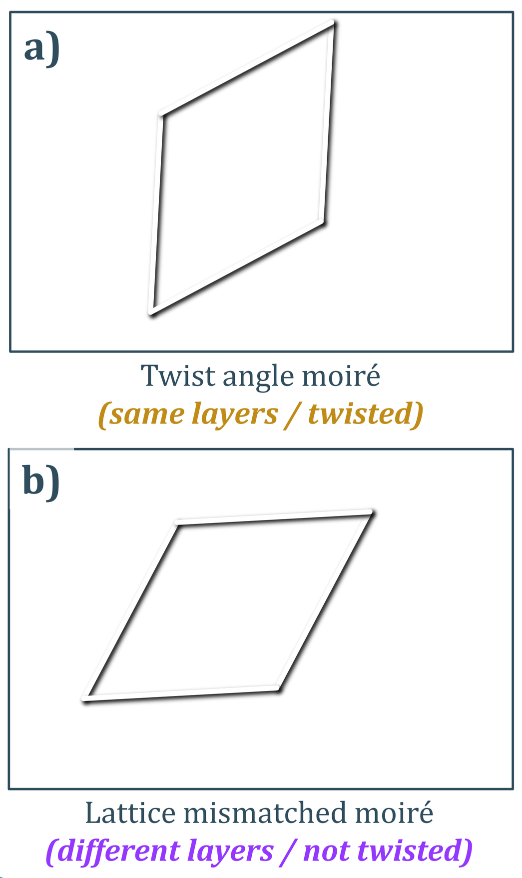
Moiré materials have emerged as a two-dimensional platform with a broad range of states between superconducting and insulating. Despite the remarkable scientific progress, an important bottleneck is the lack of a general and facile approach to producing moiré materials. SuperSuper is targeted at developing a disruptive straightforward bottom-up approach to prepare highly crystalline moiré materials, which will exceed the current challenges and limitations of existing materials and methods and establish Proof-of-Principle of their technological potential. Achieving this would represent an important step forward in the design of the next generation of superconductors.

Moiré Superlattices



A 2D moiré superlattice is formed when two (or more) atomically thin layers of materials, such as graphene or transition metal dichalcogenides (TMDs), are stacked with a relative twist angle or a slight mismatch in lattice constants. This superposition creates a moiré pattern with a period much larger than the lattice constants of the original 2D materials. Moiré superlattices exhibit new quantum properties and cooperative phenomena that are often markedly different from the original materials. Several exotic quantum phenomena have been observed in moiré superlattices, including flat electronic bands, the anomalous quantum Hall effect, unconventional superconductivity, and ferromagnetism. These discoveries highlight the immense potential of moiré materials to revolutionize future quantum technologies

Opportunities

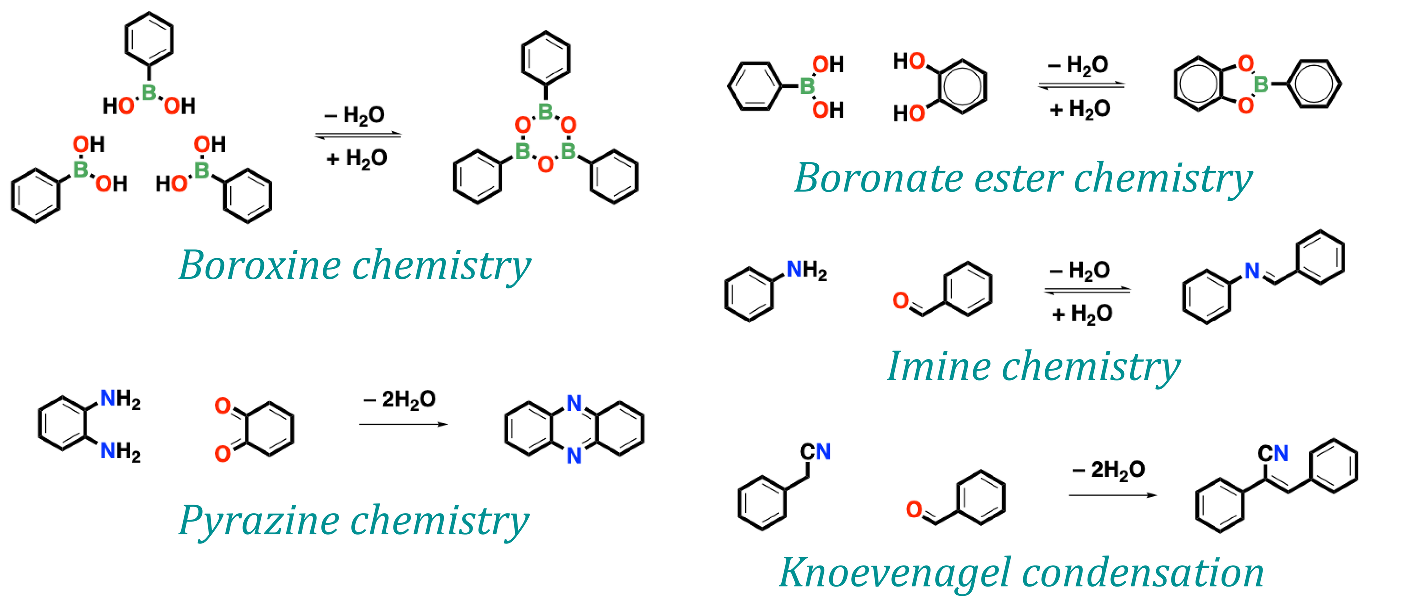
- Flat bands
- Superconductivity
- Ferroelectricity
- Quantum Hall effect
- Exotic physics

Challenges

- Angle dependence
- Angled domains
- Lattice relaxation
- Reconstruction
- Strain
- Reproducibility

2D Polymers and 2D-COFs

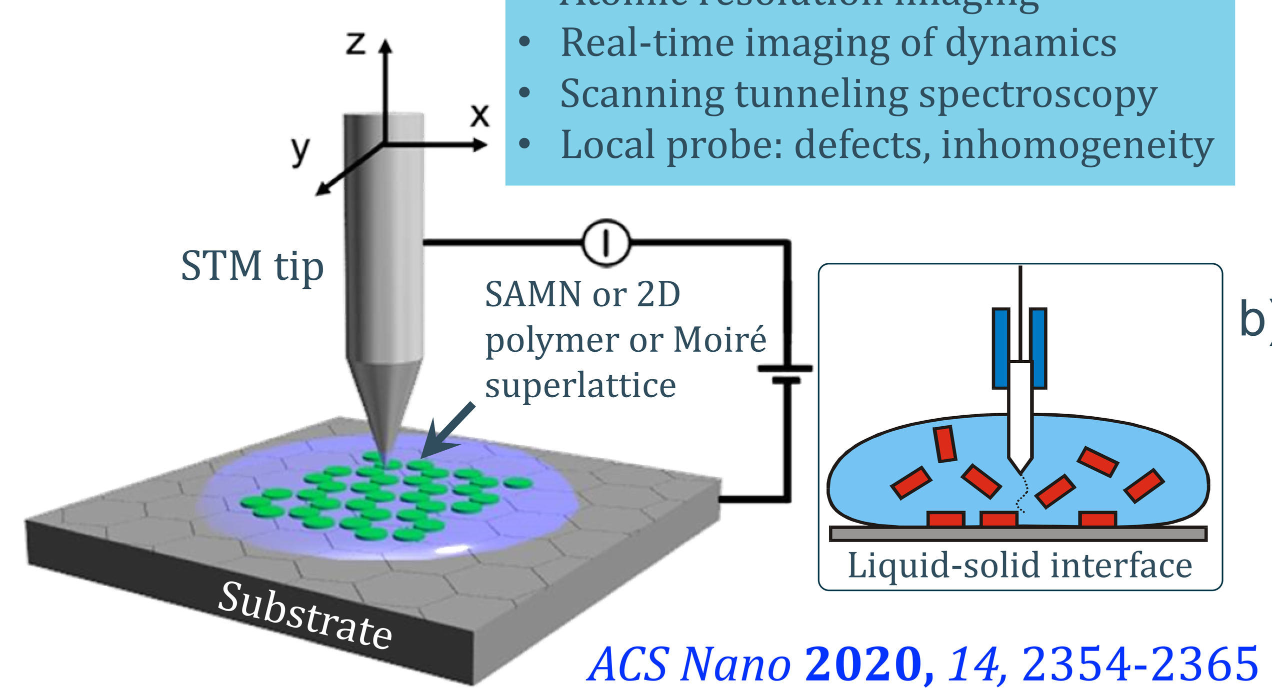
Synthetic 2D polymers offer a designer alternative to conventional 2D materials such as graphene and TMDs. They are composed of individual molecular subunits, called monomers, which are linked by covalent bonds to form atomically thin sheets with long-range order



Scanning tunneling microscopy (STM) of 2D Polymers: Structure, nucleation and growth kinetics

STM at the solution-solid interface

- **Scanning tunneling microscopy**
- Atomic resolution imaging
- Real-time imaging of dynamics
- Scanning tunneling spectroscopy
- Local probe: defects, inhomogeneity



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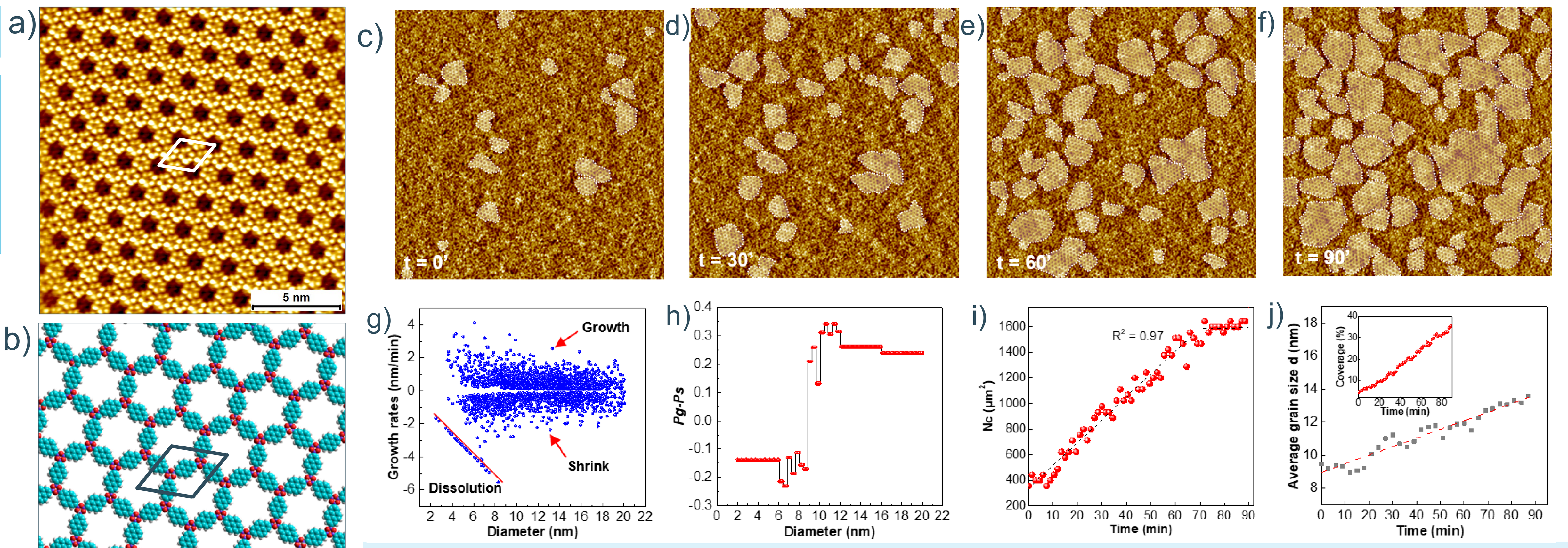
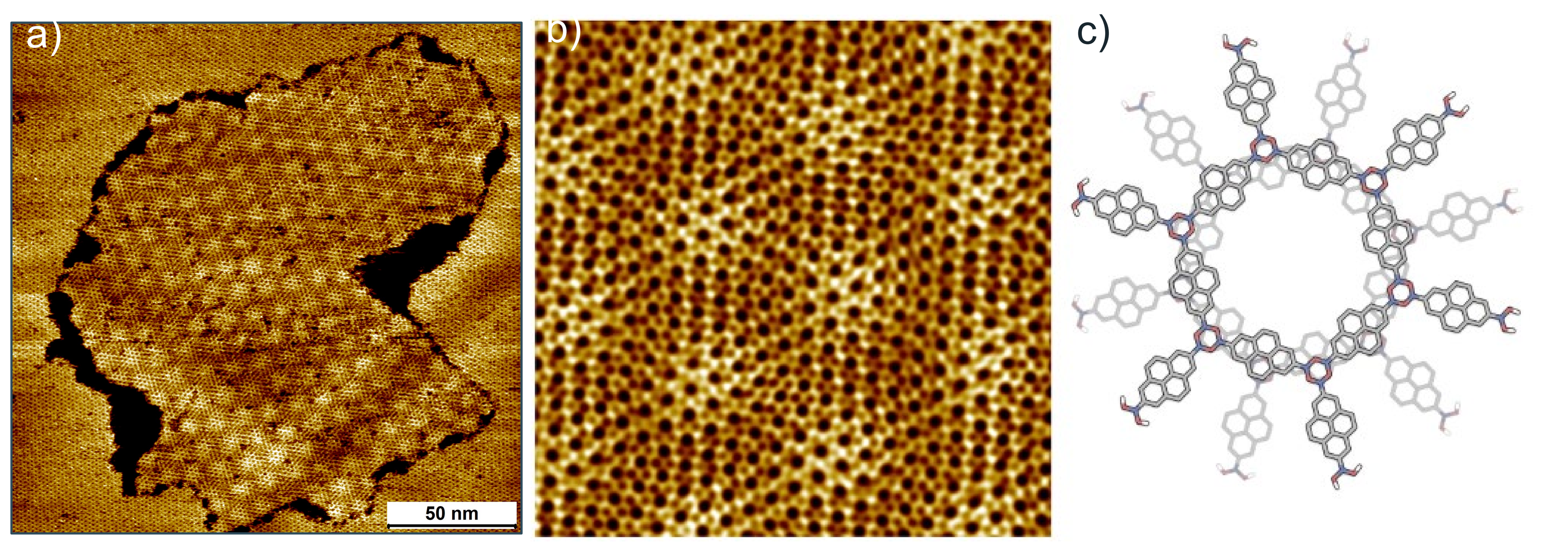


(a) High-resolution STM image of the pyrene-based 2DP. (b) Molecular model for the 2DP. (c-f) Sequential STM images showing the evolution of 2DP nuclei over time. (g-j) After analyzing 2268 individual nuclei, the growth rate (g), critical nucleation size (h), nucleation rate (i), and average growth rate (j) could be obtained.

2D Polymer-based Moiré Superlattices: The proof-of-concept



(a) Proof-of-concept results showing the moiré superlattice formation in the case of a synthetic 2D polymer. The STM image shows a moiré (AB stack) domain surrounded by a non-moiré (AA stack) domain formed at the liquid-solid interface. The molecular structure of the monomer is provided. (b) High-resolution STM image of the moiré superlattice. (c) Proposed molecular model for the moiré superlattice

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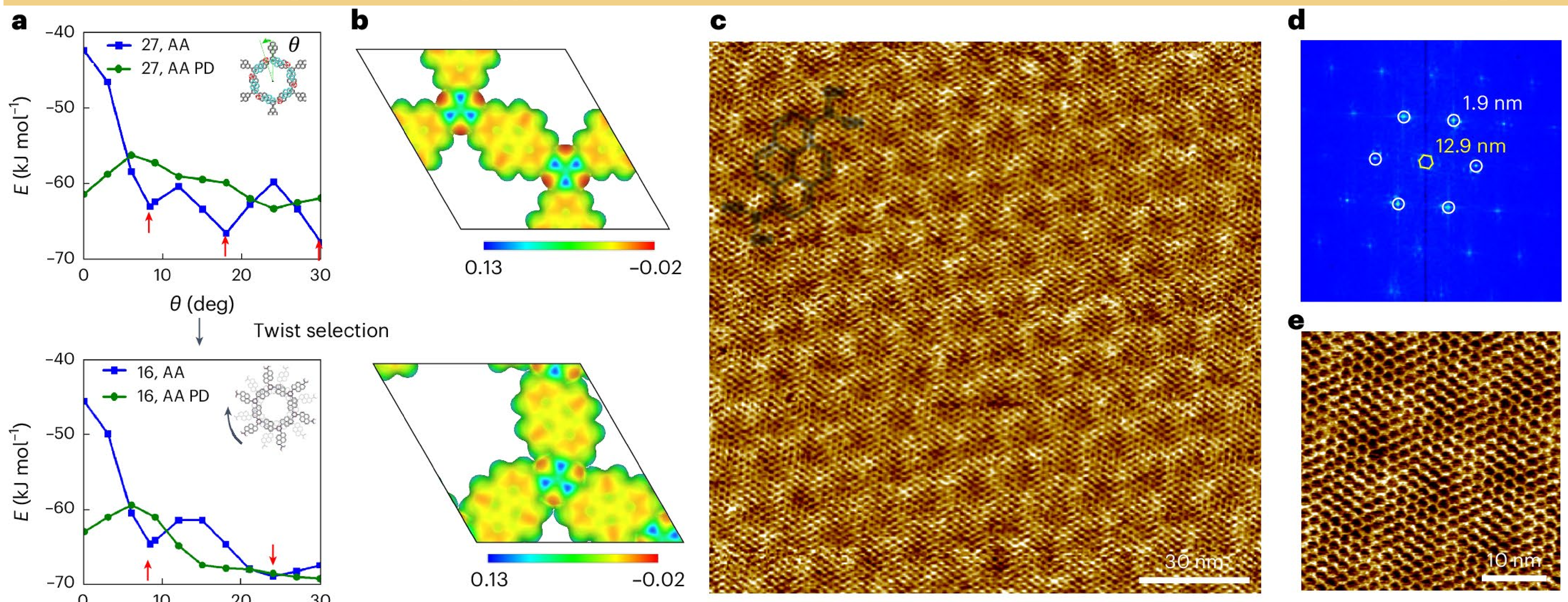
Bottom-up engineering of 2DP moiré superlattices: Deterministic twist angle



(a) Calculated system energy variation as a function of the twisted angle between top and bottom COF layers normalized by pyrene unit. (b) Electrostatic potential surface (Ha) for 27-(top) and 16-(bottom) PDBA periodic monolayers projected on the 0.05 eÅ^{-3} electron density iso-surface. (c) Large-scale STM image revealing the moiré superlattice formed by twisted bilayer 2D-COFs. (d) FFT of c. The hexagonal arranged white circle points out the COFs-2 lattice, while the inner yellow hexagon indicates the superlattice. (e) High-resolution STM image of the structure shown in c.

Summary:

- ✓ *Synthetic 2D polymers or 2D COFs offer an exciting and exotic alternative to moire superlattices formed by graphene and TMDs*
- ✓ *Scanning tunneling microscopy of synthetic 2D polymer bilayers obtained via on-surface synthesis supported by computer simulations is a promising approach to target new moiré superlattices via a bottom-up approach.*
- ✓ *Recently published proof-of-concept results clearly demonstrate the feasibility of this approach and highlight the rich and complex behavior of twisted 2D polymer bilayers.*
- ✓ *These initial results are expected to pave the way towards engineering of synthetic twisted materials with moiré superlattices with potential exotic properties.*

Acknowledgments

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