

Alternative Superconducting Superlattices

based on Covalent Organic Frameworks (COFs).

Veniero Lenzi¹, Brecht Koek², Vipin Mishra², Karol Strutynski¹, Joaquín Almarza³, Kunal S. Mali², Aurelio Mateo-Alonso^{3,4}, Steven De Feyter² and Manuel Melle-Franco^{1*}

¹ CICECO – Aveiro Institute of Materials, Department of Chemistry, University of Aveiro, 3819-010 Aveiro, Portugal

² Department of Chemistry, Division of Molecular Imaging and Photonics, KU Leuven, Leuven, Belgium-3001

³ POLYMAT, University of the Basque Country UPV/EHU, Donostia-San Sebastian, Spain

⁴ Ikerbasque, Basque Foundation for Science, Bilbao, Spain

* manuelmelle@ua.pt

Moiré materials have emerged as a novel two-dimensional platform showing unexpected exotic properties such as superconductivity. Despite the remarkable scientific progress, an important bottleneck has been the lack of a general and facile approach to the production of these materials. *SuperSuper* aims to develop a chemical bottom-up approach to prepare highly-crystalline moiré materials.

In 2022, we reported the real-time, molecular resolution STM characterization of the nucleation and growth of a pyrene-based crystalline 2D covalent organic framework (2D-COF) on graphite. Furthermore, the synthesized material was found to be twisted in consistently reproducible angles with respect to graphite[1]. Recently, we took another step forward, by reporting the exciting experimental observation of moiré superlattices on these organic materials[2].

Despite our best efforts, why the material is twisted was not yet quantitatively understood. Aiming at filling this gap, we use computer modelling to study the growth of 2D-COFs at different twist angles revealing the origin of the experimental findings [3].

We will address the emergence of these synthetic twisted organic materials[1-2] as well as: 1) the physical reasons behind the twisting and 2) how this might change their properties. Overall, these results reveal, for the first time, the complex and rich behaviour of twisted 2D-COFs on graphene and should pave the way towards engineering synthetic twisted materials with moiré lattices and potential exotic properties.

References

[1] G. Zhan, *et al.*, [**Observing polymerization in 2D dynamic covalent polymers**](#), *Nature* **603** (2022), 835-840.

[2] G. Zhan *et al.*, [**Moiré two-dimensional covalent organic framework superlattices**](#), *Nature Chemistry*, in-press, (2025).

[3] V. Lenzi, *et al.*, **Why covalent organic frameworks grow twisted on graphite**, submitted (2025).

Acknowledgements

S.D.F. acknowledges support from KU Leuven (C1 C14/23/090) and FWO (FWO G0A4120N, G0K9822N). S.D.F., M.M.F. and A.M.-A. acknowledge funding by the European Union under the Horizon Europe grant 101046231 (FantastiCOF) and M-ERA.NET 2021 (SuperSuper). M.M.F. acknowledges support from the project CICECO-Aveiro Institute of Materials, UIDB/50011/2020, UIDP/50011/2020 and LA/P/0006/2020, financed by national funds through the FCT/MEC (PIDDAC). K.S. acknowledges funding from the Scientific Employment Stimulus Program (2022.07534.CEECIND). V.M. acknowledge funding from the European Union's Horizon 2020 research and innovation programme under the Marie Skłodowska-Curie grant agreement No 101154831(RAPID-AFM). A.M.-A. acknowledges support from the Basque Science Foundation for Science (Ikerbasque), POLYMAT, the University of the Basque Country, Diputación de Guipúzcoa, Gobierno Vasco (PIBA_2024_1_0030 and BEREC programme) and Agencia Estatal de Investigación (Projects PID2021-124484OB-I00, PCI2022-132921, CEX2020-001067-M and María de Maeztu Excellence Unit CEX2023-001303-M financed by MCIN/AEI/10.13039/501100011033). This Project has received funding from the European Research Council (ERC) under the European Union's Horizon 2020 Research and Innovation Programme (Grant Agreement No. 722951). Technical and human support provided by SGIker of UPV/EHU is acknowledged.