



## First spatial observation of an entire second-order superlattice in twisted bilayer graphene

A team led by ICFO was able to map, for the first time, the entire spatial distribution of a second-order superlattice within a twisted bilayer graphene device, offering a deeper fundamental understanding and revealing previously hidden features of this kind of structures.

The study shows that these structures are very sensitive to strain and to the twist angle between layers, and thus they could serve as a 'magnifying glass' to detect deviations of these parameters when engineering bilayer graphene devices.

October 02, 2024

In March of 2018, an international team led by Pablo Jarillo-Herrero from the MIT reported the discovery of [superconductivity](#) when two layers of [graphene](#) (a two-dimensional, that is, one-atom thick, layer of graphite) were stacked on top of each other with a twist angle of  $1.1^\circ$

The small twist between the layers induces an interference pattern (known as moire pattern with a periodicity that depends on the twist angle. For the very specific 'magic angle' of 1.1°, the electronic properties are changed in such dramatic fashion that it leads to exotic physics such as the discovered superconductivity

. After that, material scientists from all over the world started to stack and twist [two-dimensional materials](#), hoping for interesting effects -such as correlated interactions and superconductivity- to arise. As expected, slight rotations consistently gave rise to exotic physical phenomena, dramatically altering the electronic properties of the mono-layers on their own.

Nowadays, twisted bilayer devices are already quite a 'standard' practice, and the community has started to increase the number of twisted layers. This can lead to an interference effect between the moire lattices corresponding to different pairs of twisted bilayers. Consequently, such 'moire superlattice of two moire lattices' can have periodicities of hundreds of nanometers, exceeding those of the underlying moire lattices. Due to this dramatically increased periodicity, the so-called second-order superlattice (SOSL) can be spatially observed with optical techniques. Even though some techniques (for instance, STM) have provided insights on the mechanisms taking place inside this kind of structures, they can only address a small subsection of the device, offering limited information. Instead, a direct visualization of SOSLs' entire spatial distribution across a device would provide a deeper fundamental understanding and potentially reveal undiscovered features.

The observation of the spatial distribution of a whole SOSL was still lacking until, recently, an article in Nature Materials came out. [ICFO researchers](#) **Dr. Niels C. H. Hesp, Sergi Batlle-Porro, Dr. Roshan Krishna Kumar, Dr. Hitesh Agarwal, Dr. David Barcons Ruiz, Dr. Hanan Herzig Sheinfux, Dr. Petr Stepanov**, led by ICREA Prof. Frank H. L. Koppens, in collaboration with [NIMS](#) and [University of Notre Dame](#) **have reported on a new type of experiment that, for the first time, maps the entire SOSL inside a twisted bilayer graphene device.**

The team stacked two layers of graphene on top of a hexagonal Boron Nitrate (hBN) layer. They then performed cryogenic nanoscale photovoltage measurements (in short, cryoSNOM) and electronic transport measurements. The former combines high spatial resolution with extreme sensitivity to local electronic properties, while the latter was used to verify the twisting angles between layers and to confirm the presence of magic-angle physics.

Supported by simulations, the team finally unveiled the **unprecedented sensitivity of SOSLs to the strain experienced by the material and the twist angle between the layers**, but the journey until arriving to these results was full of surprises.

### **From measurement issues to signs of a SOSL**

The whole story started when the team applied cryogenic nano-imaging and electronic transport measurements to the sample. The initial goal was to obtain hints of correlated physics and superconductivity, but throughout the process they observed some unusual

patterns that no one had seen before.

Their immediate reaction was to assume that there was something wrong with the measurement system. The cryoSNOM was facing many issues, so we suspected that the tip (the contact point of the measurement setup with the sample) was broken, explains Dr. Niels Hesp, first author of the article. But it seemed that, at least at that particular moment, the measurement system was perfectly fine.

They therefore decided to further explore these unexpected patterns, which presented a large period (of about 400 nanometers), in an attempt to discover their nature and origin.

In the end, researchers realized these long-range periodicities were a clear indication of SOSLs.

That was very extraordinary as well, because it is very unlikely to have a device prepared in this state, recalls Hesp, since it is very hard to get the twist angles exactly right. The team was not intending for this, but they embraced the opportunity, changed course of action and focused on the next step: to get proper measurements done on the sample to fully characterize it. They knew it was a challenge, as with the least voltage the device could get burnt. They needed to be extremely careful. At some point, the cryoSNO unexpectedly started warming up, which partially broke the sample. I was able to fix it in time, avoiding irreparable damage. Luckily, the unique device had only a little hole in the middle, leaving the second-order superlattice surprisingly unaffected, shares Dr

Petr Stepanov. Another surprise came right after that, when researchers noticed in the simulations that **second-order superlattice manifests itself when the alignment between the graphene layers coincides exactly with the 'magic angle' of twisted bilayer graphene**. Hesp, though, remarks the lack of a fundamental relation between both phenomena: **We don't see any causality between the twist angle where magic-angle physics occurs and where a SOSL occurs, and therefore it is to us a coincidence**. Yet, this means that when the twist angle of the hBN layer is tuned right, **the twisted bilayer graphene device not only shows magic-angle physics but also hosts a SOSL, which makes the physics of magic-angle twisted bilayer graphene even richer**. Hesp also suspects that, even though the reported configuration does not modify the magic-angle physics at first sight, it will affect how some exotic phenomena (such as the anomalous Hall effect) manifest within these systems.

### **A 'magnifying glass' to precisely tune strain and twist angles in bilayer graphene**

The holistic spatial observations that the team carried out showed that minuscule variations in the strain and twisting-angle led to drastic changes in the second-order superlattice structure. In this sense, **the reported method serves as a 'magnifying glass' to spot small deviations with respect to the desired strain and twist angles**. Thus, **it could be used to realize quality controls in magic-angle twisted bilayer graphene devices, avoiding inaccuracies during its fabrication process**.

Hesp, who started this project right at the end of his PhD, is proud of how everything turned out: *If you ask me what the beauty of this work is, I would say it is not only the fascinating*

g richness in superlattices that one can create, but also that it reveals a sizeable effect that is present in part of the twisted-bilayer graphene devices that people study without realizing it?

**Reference:**

Hesp, N.C.H., Batlle-Porro, S., Krishna Kumar, R. et al. Cryogenic nano-imaging of second-order moire superlattices. Nat. Mater. (2024).  
<https://doi.org/10.1038/s41563-024-01993-y>

**Acknowledgements:**

F.H.L.K. acknowledges financial support from the Government of Catalonia through the SGR grant, and from the Spanish Ministry of Economy and Competitiveness through the Severo Ochoa Programme for Centres of Excellence in R&D (no. SEV-2015-0522) and Explora Ciencia (no. FIS2017-91599-EXP). F.H.L.K. also acknowledges support from the Fundacio Cellex Barcelona, Generalitat de Catalunya, through the CERCA program and the Mineco grant Plan Nacional (no. FIS2016-81044-P) and the Agency for Management of University and Research Grants (AGAUR) (no. 2017-SGR-1656). Furthermore, the research leading to these results has received funding from the European Union's Horizon 2020 programme under grant agreement nos. 785219 (Graphene Flagship Core2), 881603 (Graphene Flagship Core3) and 820378 (Quantum Flagship). This work was supported by the ERC under grant agreement no. 726001 (TOPONANOP). P.S. acknowledges support from the European Union's Horizon 2020 research and innovation programme under the Marie Skłodowska-Curie grant no. 754510. N.C.H.H. acknowledges funding from the European Union's Horizon 2020 research and innovation programme under the Marie Skłodowska-Curie grant agreement no. 665884. K.W. and T.T. acknowledge support from JSPS KAKENHI (grant nos. 19H05790, 20H00354 and 21H05233). This project has received funding from the 'Presidencia de la Agencia Estatal de Investigacion' within the PRE2020-094404 predoctoral fellowship.