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Homophase bilayers: more than just the sum of their monolayers

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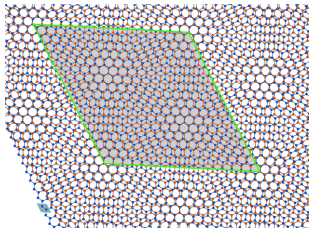
Twisted homophase bilayers, stacks of two rotated monolayers such as graphene, exhibit remarkable physical properties absent in their constituent monolayers. The structure of bilayer systems is dominated by a moiré effect and critically depends on the twist angle. Quiquandon & Gratias [*Acta Cryst.* (2025), A**81**, 94–106] develop a crystallographic framework for rigorous description of the structure of bilayers, including systems without a coincidence lattice. They offer a set of tools that can describe the structure of any arbitrary bilayer system and enable the connection with its physical properties.

In the beginning was graphene, a single atomic layer of carbon atoms arranged on a hexagonal honeycomb lattice, and it was the premier experimental realization of a true 2D material. It quickly turned out that graphene possesses remarkable physical properties including ballistic electronic transport, extreme strength and high optical opacity (for a recent review, see *e.g.* Urade *et al.*, 2023). The discovery of further single-layer materials, such as the 2D compound hexagonal BN (Kubota *et al.*, 2007) or the hexagonal transition-metal dichalcogenide MoS_2 (Radisavljevic *et al.*, 2011), advanced research of 2D materials into a mature scientific discipline.

In a next evolution step, homophase bilayers, stacks of two identical monolayers, were addressed (Novoselov *et al.*, 2004). These systems become particularly interesting when the two monolayers are twisted with respect to each other (Trambly de Laissardière *et al.*, 2010, 2012) and even more so if they are twisted at a small angle. Bistritzer & MacDonald (2011) predicted a 'magic angle' of about 1.1° at which the Dirac velocity vanishes, accompanied by a flat band structure and a sharp peak of the Dirac-point density of states. This was experimentally confirmed by Cao *et al.* (2018), who observed flat bands at zero Fermi energy and superconductivity below 1.7 K. This result is in contrast to monolayer graphene, which is not superconductive. Bilayer graphene thus displays properties that are not present in its constituent monolayers. There are more examples like this: while graphene monolayers are zero-bandgap semiconductors, bilayer graphene has a bandgap, which is even tunable (Min *et al.*, 2007). Bilayer graphene shows degenerate electronic states, which are not present in monolayers, leading to a fractional Hall effect that can be tuned by an electric field (Kou *et al.*, 2014). Observations like the latter have led to the term 'twistronics' for the electronics of twisted bilayer devices.



The appearance of these novel properties is directly linked to an increase in structural complexity. Graphene monolayers have a particularly simple structure, but adding a twisted

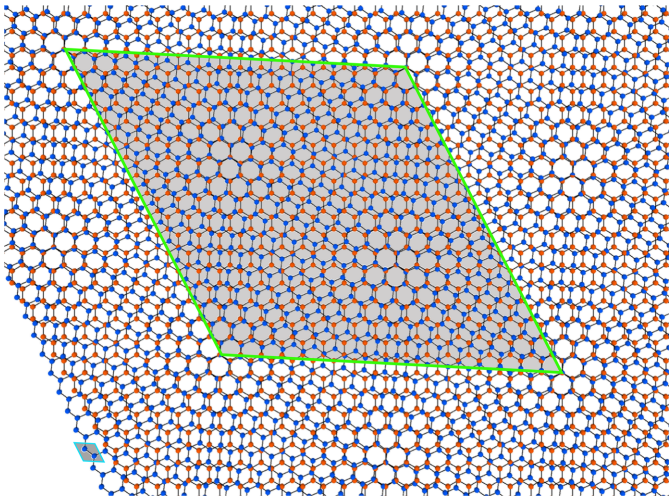


Figure 1
Bilayer system consisting of two graphene monolayers (red and blue) at a twist angle of 6.0°. The unit cell of a monolayer (small blue rhomb) and the resulting moiré crystal (green rhomb) are shown.

second layer fundamentally changes the situation. At small twist angles the atom arrangement in the bilayer system is dominated by a moiré effect creating a long-range modulation of the structure. Consequently, the system's lattice parameter by far exceeds that of the constituent monolayers, and the large unit cell contains numerous different local atomic environments (Fig. 1).

In a twisted bilayer, the structure is critically dependent on the twist angle (Feuerbacher, 2021), and the physical properties, in turn, are critically dependent on the structure. This demands a rigorous crystallographic description that includes the twist-angle dependence. It is essential to understand how the symmetries of the bilayer system interplay with the Coulomb interaction (Kou *et al.*, 2014), how they lead to phenomena like flat bands, the resulting electronic localization, or shifting Van Hove singularities.

Quiquandon and Gratias, therefore, embarked on an in-depth study with the goal of developing a crystallographic framework for twisted homophase bilayer systems. Their work resulted in a series of papers, the first dealing with bilayers that have a coincidence lattice (Gratias & Quiquandon, 2023). This is the case when the twist angle is a fraction of π, resulting in a periodic bilayer with a standard space group. The second paper describes the more general case of bilayers without a coincidence lattice, where the resulting crystal is not periodic but 'almost periodic' (Quiquandon & Gratias, 2025). Crystallographically, the system then is in a rather elusive state, yet demands rigorous description as it is still linked to specific physical properties. In order to approach this situation, the authors employ two innovative strategies: first, they introduce a descriptive formalism in terms of complex numbers, which makes operations in the plane such as rotations and translations very easy to handle through the well established framework of conformal transformations. Second, they use the concept of the 'zero locus', a generalization of the 0-lattice introduced for the treatment of grain boundaries (Bollmann, 1967). The zero locus enables the transition from bilayer systems with a coincidence lattice to the more general case of bilayer systems without a coincidence lattice. In the context of twisted homophase bilayers, the zero locus refers to the geometric set of points that remain invariant under the combined operations of rotation and translation between the two layers. The authors show that there always exists a set of invariant points, which do not move when one layer is rotated and translated relative to the other. Unlike a coincidence lattice, which forms only for rational twists, the zero locus exists for any arbitrary twist angle. This makes the zero locus a more general mathematical structure, which is capable of describing bilayer symmetries even in non-periodic cases.

What the eye, or the electron microscope, sees when looking at twisted bilayers is the moiré crystal, which corresponds to the interference pattern of the superposed functions describing the mass distribution or the electron density in the individual monolayers. The moiré crystal, again, is periodic when a coincidence lattice exists but it is almost periodic in the general case. The authors provide an explicit description of the general moiré crystal and use it to directly infer the crystallographic symmetries of the bilayer system.

With their work, Quiquandon and Gratias provide a novel crystallographic framework for twisted bilayer systems. Their approach offers a set of tools that can describe the structure of any arbitrary bilayer system in a closed manner. Not only does it enable one to make connections between almost periodic moiré patterns and known physical phenomena like quantum interference and electronic localization, but it can also be employed to localize configurations at which specific physical properties become most pronounced. Conversely, bilayer crystallography can be used to relate empirically pinpointed configurations like 'magic angles' to specific crystallographic conditions. In essence, this framework provides a solid crystallographic foundation for the burgeoning field of twistronics. It enables deeper insights into the structure–property relationships in twisted bilayer materials and will be invaluable for future progress in this rapidly developing field.

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