2D materials

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Twisted growth by design

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Twisted bilayer graphene is epitaxially grown between two adjacent Cu(111) surfaces, with the twist angle controlled by the rotation of the Cu foils as designed.

An artificial crystal with lattice structures that can be tuned arbitrarily provides an exciting platform for flexibly programming material properties. Twisted bilayer graphene (TBG) is a system where one layer is twisted relative to the other at a twist angle (θ) that is broadly tunable. Depending on θ, a moiré superlattice with a specific crystal symmetry and lattice parameter appears at the van der Waals interface. The TBG structure becomes non-centrosymmetric with finite θ and chiral for $\theta \neq 30°$, producing interesting properties, such as chiral plasmons[1] and the chiro-optical response[2], that are absent in non-twisted graphene layers. In addition, the moiré periodic potentials modulate the electronic band structures to host superconductivity and to form a Mott insulator at a specific θ close to 1.1° (ref. [3]). Researchers have been motivated by the continuous discovery of the surprising properties of these artificial crystals and have been exploring the potential electronic and optoelectronic applications based on twisted graphene films. Large-scale TBG with a twist angle that can be tuned on demand is a prerequisite for the applications, which researchers are eagerly pursuing[4]. TBG samples with atomically clean van der Waals interfaces are typically fabricated by assembling mechanically exfoliated flakes, but this process has limitations in terms of reproducibility and the sizes of fabricated samples that are usually several tens of micrometres. While direct-growth methods would be the simplest way to achieve large-scale layers, the precise control of θ is considered to be challenging with the direct-growth methods currently used. Now, writing in *Nature Materials*, Can Liu and co-workers report[5] a simple chemical vapour deposition (CVD) technique for growing centimetre-scale TBG structures with arbitrarily controlled θ.

Growth with a specific θ requires special guidance. The twisted structure is energetically less favourable than the non-twisted crystal because of the reduced interlayer interactions caused by the lattice mismatches as a result of the rotation. Epitaxy from a growth template is a common way to obtain a desired crystalline structure. For example, TBG with $\theta = 30°$ has been grown on SiC (ref. [6]). However, arbitrary control of θ was difficult because of the thermodynamic constraints on the possible atomic configurations of the epitaxial surfaces. To solve this problem, Liu and colleagues have proposed a method in which epitaxial growth occurs between two closely spaced Cu(111) surfaces that are aligned in parallel but with an in-plane rotation of angle α (Fig. 1). Graphene films with crystalline orientations aligned with the underlying Cu surfaces are grown on both sides, and then form an interface with a twist angle θ that replicates α. Therefore, TBG with arbitrary θ, including a small value of approximately 1°, can be achieved with an accuracy of <1°, uniformly over the growth region of approximately 2 cm^2, as confirmed by high-resolution transmission electron microscopy.

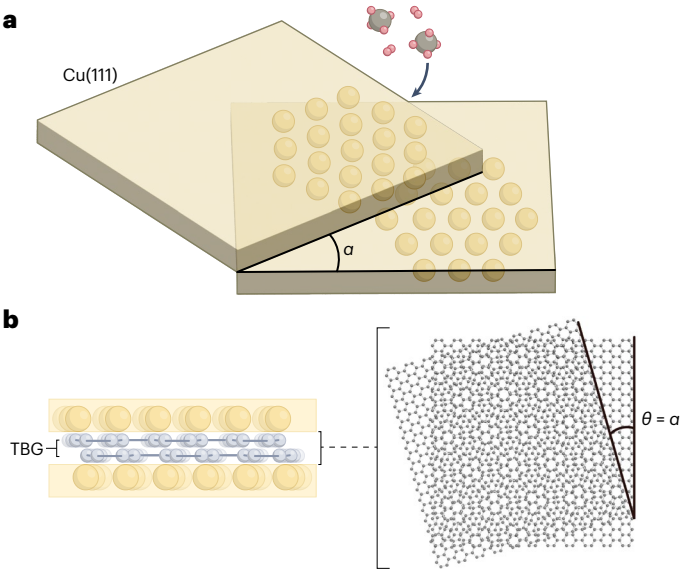

Fig. 1 | Epitaxial growth of TBG. a, Schematic illustration of the growth method. Two pieces of single-crystal Cu(111) foil are stacked with a controlled rotation angle of α. Growth precursors diffuse into the Cu interface and monolayer graphene films are grown on both sides of the interface. **b**, Schematic illustration of TBG with a twist angle (θ) sandwiched between the growth substrates. The crystalline orientations of the graphene layers are defined by the adjacent Cu surfaces. As a result, θ replicates α.

It is worth noting that TBG films encapsulated within two Cu foils after growth can be isolated and transferred. One side of the Cu layer can be selectively removed by electrochemical etching with a uniformly applied potential on the etching side. The TBG with the exposed surface is then transferred to a target surface using a conventional poly(methyl methacrylate)-assisted method. The transfer method enables the integration of the TBG layers with diverse materials and device systems to explore the opportunities for potential applications.

The direct growth of TBG with a controlled θ has several key strengths. Specifically, this method is ideal for obtaining high-quality interfaces between graphene layers. In a layer-by-layer assembly method based on CVD-grown films, the interface is easily contaminated by the different chemicals and polymers it is exposed to. Contaminants cause variations in θ and even prevent the hybridization of the electronic states of individual layers, which is critical for hosting a variety of physical phenomena[1–3]. By contrast, the direct growth of twisted bilayers in a more tightly controlled gas environment results in interfaces that are atomically pure. Importantly, the growth occurs on Cu substrates, which are the most widely researched materials for graphene growth. Therefore, previously reported techniques may be used to achieve ultrafast growth[7] and to control the number of layers[8]. Notably, the same substrate has also been used to grow monolayer

hexagonal boron nitride, indicating that the technique could be used to fabricate other twisted material families[9].

The technique developed by Liu and colleagues provides a powerful tool for making designed twisted 2D materials, but there is still room for exploration. First, exotic phenomena[1,3] based on the moiré superlattice require precise control of not only θ but also the doping level. However, the wet transfer process used by Liu and colleagues can introduce heterogeneous contaminants on the surface of TBG. Second, the number of layers is currently limited to two, but there is growing interest in creating twisted multilayer structures to further engineer the properties of artificial crystals[10] and to produce thicker films for other applications[11]. Additionally, although the Cu substrate has its own benefits, other growth substrates[4,8] may provide opportunities to meet various application demands and are worth further investigation.

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Competing interests

The authors declare no competing interests.