2D MATERIALS

Programming twist angle and strain profiles in 2D materials

Maëlle Kapfer1†, Bjarke S. Jessen1‡, Megan E. Eisele3, Matthew Fu1, Dorte R. Danielsen2,3, Thomas P. Darlington4, Samuel L. Moore4, Nathan R. Finney4, Ariane Marchese4, Valerie Hsieh1, Paulina Majchzak5, Zhihao Jiang6, Deepnarayan Biswas2†, Pavel Dudin6, José Avila6, Kenji Watanabe7, Takashi Taniguchi7, Søren Ulstrup5, Peter Bøggild2,3, P. J. Schuck4, Thomas P. Darlington4, Samuel L. Moore1, Nathan R. Finney4, Ariane Marchese4, Valerie Hsieh1,‡

Moiré superlattices in twisted two-dimensional materials have generated tremendous excitement as a platform for achieving quantum properties in demand. However, the moiré pattern is highly sensitive to the interlayer atomic registry, and current assembly techniques suffer from imprecise control of the average twist angle, spatial inhomogeneity in the local twist angle, and distortions caused by random strain. We manipulated the moiré patterns in hetero- and homolayers through in-plane bending of monolayer ribbons, using the tip of an atomic force microscope. This technique achieves continuous variation of twist angles with improved twist-angle homogeneity and reduced random strain, resulting in moiré patterns with tunable wavelength and ultralow disorder. Our results may enable detailed studies of ultralow-disorder moiré systems and the realization of precise strain-engineered devices.

The interference pattern between twisted or lattice-mismatched layers of two-dimensional (2D) materials leads to a periodic electrostatic scattering potential superimposed over the atomic lattice (1, 2). Additionally, periodic structural distortions from in-plane lattice relaxation and out-of-plane deformations can occur as a result of the interference pattern (3–5). This so-called moiré pattern can cause the appearance of flat bands in the electronic energy spectrum, which are found to host a variety of interaction-driven many-body states such as correlated insulators, superconductivity, and magnetic order as well as nontrivial topology (6, 7). The ability to laminate 2D materials with different atomic composition and lattice constants, and at arbitrary twist angles, allows for wide tunability of the moiré wavelength, amplitude, and symmetry (8), with further modification possible through in situ rotation (9–11), pressure (12, 13), and strain (13–16). Moiré patterning in 2D materials therefore provides a promising and flexible platform to realize correlated physics and topology in quantum materials by design (8).

Small variations in the moiré pattern, however, can lead to dramatically different behaviors, making it difficult to uncover the exact relationship between structural details of the superlattice and resulting electronic properties. For example, the flat band—hosting superconductor in twisted-bilayer graphene emerges for twist angles between the layers within only ±0.1° (17, 18), corresponding to a difference in moiré wavelength of just ±1 nm. This places stringent demands on the fabrication process (19). Moreover, experimental studies have found that spatially varying twist angle and distortion of the moiré unit cell caused by random strain fluctuations are ubiquitous when using current assembly techniques (20–23). Such nonuniformity compromises the ability to deterministically engineer specific band structures through well-defined moiré patterns or even correlate experimental measurement with predictive theoretical modeling (24–29). The inability to control this “moiré disorder” and the lack of any systematic understanding of its influence represents a critical roadblock toward continued progress in this field (29).

In this work, we demonstrate a technique that addresses these issues by using global strain fields to manipulate local moiré structures. We used an atomic force microscope to bend ribbon-shaped monolayers of 2D materials in-plane resulting in a continuously varying twist angle along the length of the ribbon. We confirmed through real-space imaging using piezoresponsive force microscopy (PFM) and lateral force microscopy (LFM), as well as optical mapping by means of Raman spectroscopy and nanophotoluminescence (nanoPL), that the resulting twist-angle and strain gradients vary smoothly and with a predictable spatial dependence. The ability to vary the relative twist angle by several degrees allows us to span wide-ranging moiré length scales in a single device. Most unexpectedly, we found substantially reduced moiré disorder in the bent ribbon geometry.

Bending 2D material ribbons

A schematic of the bending process is shown in Fig. 1A, illustrated for the case of a bent monolayer graphene (bMLG) ribbon on an hexagonal boron nitride (hBN) substrate. The heterostructure was first assembled by using the dry transfer technique in which we used the hBN to sequentially pick up a straight graphene ribbon, followed by a few-layer graphite manipulator shaped into a gear-like geometry (30). Using an atomic force microscope tip to slide the manipulator over the end of the ribbon allows us to apply an in-plane load (9). In response, the ribbon bends at the load end, with the far end effectively clamped owing to interfacial friction. Using the atomic force microscope tip, we thus directly control the displacement of the ribbon through a loop of topographic imaging and nanomanipulation.

We characterize the deformed ribbon using an (x, y) coordinate system, where x follows the neutral axis before bending, and y is the distance from the neutral axis perpendicular to x (Fig. 1B). We also introduce W and L as the width and bending length, respectively; d(x) as the deflection; and Δθ(x) as the local twist angle relative to the orientation of the unbent ribbon. The local twist angle relates to the slope of the deflection curve through

$$\Delta \theta(x) = \tan^{-1}(d(x)/y)$$  (1)

An atomic force microscopy (AFM) image of a bMLG on hBN is shown in Fig. 1C, with the bMLG highlighted in blue, showing the initially unbent (Fig. 1C, top) and final bent (Fig. 1C, bottom) geometries. The bending process is both reversible and robust because we can bend and unbend the ribbon up to several degrees without evidence of plastic deformation (fig. S5). Upon releasing the load, static friction is sufficient to secure the deflection profile of the bent beam, and conventional dry transfer techniques can be used to add additional layers to the heterostructure without negatively affecting the geometry. We have applied the bending geometry to varying combinations of graphene, hBN, and transition-metal dichalcogenides and observed similar results in all cases.

Owing to the closely matched lattice constants of graphene and hBN, a large-wavelength moiré superlattice λ emerges in the small twist angle regime, varying with the twist angle, \(\theta\), according to

$$\lambda = \frac{(1 + \delta)a}{2(1 + \delta)|1 - \cos(\theta)| + \delta^2}$$  (2)

where a is the graphene lattice constant, \(\theta\) is the twist angle, and \(\delta\) is the lattice mismatch...

Kapfer et al., Science 381, 677–681 (2023) 11 August 2023

1 of 5
(0.017 for graphene on hBN) (2). Thus, measuring the moiré wavelength provides a way to directly determine the local twist angle (31, 32). Shown in Fig. 1D are real-space and Fourier-space images acquired with PFM from two different locations on the bMLG shown in Fig. 1C. From the positions of the peaks in the fast Fourier transform (FFT), we extracted the average moiré wavelength and used Eq. 2 to determine the average twist angle within the scan window. Repeating this measurement along multiple points on the bMLG, we spatially mapped the twist angle along the ribbon. The measured change in twist angle, Δθ, is shown in Fig. 1E as a function of x for three different ribbon widths of 0.8, 1.3, and 5 μm.

In terms of modeling the deflection curves of the bent ribbon, d(x), we observed key differences compared with a classical cantilever: (i) upon bending, only part of the ribbon, closest to the loading point, is deflected; (ii) the length, L, over which bending occurs is larger for wider ribbons; and (iii) upon releasing the load at the end of the ribbon, the ribbon retains its bent shape. We found that all deflection curves for bent graphene ribbons, regardless of width or orientation to the underlying substrate, follow

$$d(x) = \frac{d_0}{L} (L - x)^3$$ (3)

where $d_0$ is the deflection at the load point, and L is the length over which bending occurs (fig. S4). We found an approximately linear relation between W and L, which we surmise reflects the role of friction in the system (fig. S4). On the basis of this understanding, we explored the limits of the bending approach.

In Fig. 1F, we show the measured maximum twist angle variation as a function of the ribbon width for different graphene ribbons on hBN. We observed that beyond a critical deflection, some bMLGs buckle (33), suddenly transitioning into two mostly straight sections separated by a highly strained fold (fig. S5). By balancing the van der Waals energy between the ribbon and the substrate with the elastic energy from strain, we calculated a critical strain of ~2.5%, after which buckling becomes energetically favorable, shown as the dashed line in Fig. 1F (30). This sets an upper limit on Δθ of ~5° for bMLG, which is consistent with the approximate trend seen in Fig. 1F.

**Mapping strain**

The strain caused by bending varies both in the longitudinal ($x$) and transverse ($y$) directions and is given by

$$\varepsilon(x, y) = -\frac{y}{\rho(x)}$$

where $\rho(x) = \{1 + [d(x)]^2 / 3C_0^2/d(x)\}$ is the local radius of curvature. The calculated spatial strain maps for two ribbons are shown in Fig. 2A, one 1 μm wide and one 5 μm wide, each bent by a maximum twist-angle variation of 1°. The strain evolves linearly in both the transverse and longitudinal directions, with the maximum strain found at the load end of the ribbon. In the transverse direction, the ribbon has compressive strain on the inside radius and tensile strain on the outside, with an unstrained neutral axis separating the two. The maximum transverse strain gradient, $-\varepsilon_{\text{max}}/W/2$, reaches as high as 0.47% per micrometer for the 1-μm-wide ribbon but only 0.09% per μm for the 5-μm-wide ribbon. Similarly, the maximum longitudinal strain gradient, $-\varepsilon_{\text{max}}L$, reaches 0.03% per micrometer and 0.006% per micrometer for the narrow and wide ribbon, respectively. This indicates that the bending geometry allows us to effectively program the twist-angle and strain gradients individually through choice of ribbon width and maximum displacement. We confirmed this independent tuning through the use of (nano) angle-resolved photoemission spectroscopy, in which we spatially mapped out the band structure of a 4-μm-wide bMLG bent 4°. As expected from a low-strain ribbon, we observed a significant Brillouin zone (BZ) rotation but otherwise minimal impact on the band structure (fig. S9).

Analysis of the moiré pattern allows us to map the local strain field, in addition to twist angle, because the moiré pattern magnifies any distortion in the constituent lattices (34). This is visualized in Fig. 2B, where we show a schematic of a bent bMLG on hBN. The impact of the strain is apparent from the heavily distorted moiré lattice (35–37), which is especially visible for the large moiré periods in the low-twist-angle regime. The effect is maximal at the top and bottom edges of the bMLG and gradually decreases toward the unstrained neutral axis, near the middle. The resulting BZs of the twisted layers in the unstrained and tensile regions are shown in Fig. 2, C and D, respectively.

Examples of distorted moiré patterns acquired by means of PFM from a 1.3-μm-wide bMLG on hBN are shown in Fig. 2, E to G. The dashed line in Fig. 2E indicates the initial position of the bMLG (highlighted in blue) before bending. High-resolution moiré patterns and corresponding FFTs are shown in Fig. 2, F.
and G, top and bottom, respectively, acquired from two locations (indicated in Fig. 2, F and G, with white symbols) of the bBLG shown in Fig. 2E. The scan windows correspond to two different y positions (different strain points) but fixed x positions (same twist angle). Following (38) and (39), the average of the reciprocal lattice vectors measured from the FFT gives the local twist angle, whereas the ratio of the reciprocal vectors gives a measure of the local strain. The pattern in Fig. 2F, acquired close to the neutral axis, gives \( \theta \sim 1.37^\circ \) and \( \epsilon \sim 0.2\% \), whereas that in Fig. 2G, taken in the compressive part of the bMLG, gives \( \theta \sim 1.34^\circ \) and \( \epsilon \sim -0.65\% \).

A plot of the strain versus normalized y position \( y/w \) is shown in Fig. 2H, where \( y \) is the distance from the neutral axis, and \( w = W/2 \), the half-width of the bMLG (Fig. 2H, blue circles). The strain gradient is approximately linear, with a slope that matches the value of \(-1\%\) per micrometer calculated from the displacement curve (Fig. 2H, dashed blue line) with no free parameters. For comparison, strain versus \( y/w \) from a wider 5-\( \mu m \) ribbon is also shown (Fig. 2H, yellow circles). Again we found good agreement between the strain

**Fig. 2. Tunable strain gradients.** (A) Calculated strain gradient in a (top) 1-\( \mu m \)-wide and (bottom) 4-\( \mu m \)-wide bMLG, each bent to a maximum twist-angle variation of 1°, with the dashed lines indicating their initial positions. The deflection has been exaggerated by a factor of 5 for illustrative purposes. (B) Sketch of a distorted moiré superlattice, highlighting the effect of strain in the low-angle limit. For moderate twist angles (left side; \( \theta = 27^\circ \)), the strain has only a minute effect on the fidelity of the moiré superlattice, whereas for low twist angles (right side; \( \theta = 2^\circ \)), the lattice is heavily distorted for both compressive and tensile strain. (C and D) BZs of two twisted graphene layers at no strain and tensile strain, respectively, highlighting the sensitivity of the superlattice BZ toward strain. (E) AFM image of a thin bMLG on hBN. The dashed line indicates the position before bending. (F and G) (Top) PFM scans taken across the bMLG in (E), with positions of the PFM scans marked on the AFM image of the sample. (Bottom) The corresponding FFTs, highlighting the increasingly distorted moiré pattern away from the neutral axis of the sample. (H) Summary of the strain gradient across a thin (blue) and wide (yellow) bMLG as a function of the normalized y position. The dashed lines indicate the strain gradient predicted with a mechanical model for the corresponding geometry and deflection.

**Fig. 3. Characterization of strain without moiré magnification.** (A and B) (Top) An AFM image of a 1.6-\( \mu m \)-wide bMLG. (Bottom) The corresponding strain map extracted from Raman spectroscopy. Red indicates tensile strain, and blue indicates compressive strain, with a white neutral axis identified in the middle of the bMLG. (C) Raman spectra at fixed twist angle but varying strain for the bMLG shown in (A). (D) Extracted strain of the sample in (A), with the dashed line indicating the beam bending model for a 1.6-\( \mu m \)-wide beam deflected by \( d_0 = 300 \) nm, yielding a strain gradient of 0.64% per micrometer. (E) An AFM image of a 2.4-\( \mu m \)-wide ribbon of monolayer WSe2 on hBN. (F) A strain map of the sample, from the dashed area in (E). The strain is extracted from nano-PL mapping, where the peak position is related to changes in band alignment caused by strain. (G) Representative spectra of areas of compressive (blue) and tensile (red) strain in the WSe2. (H) Extracted strain versus distance from the neutral axis of the sample in (E), with the dashed line indicating a linear fit, yielding a strain gradient of 0.33% per micrometer.
field measured from the distortions in the moiré pattern and the strain calculated from the displacement curve (Fig. 2H, dashed yellow line). Moreover, as expected, the wider ribbon gives a substantially reduced strain gradient, confirming the ability to use the bMLG width to tune the strain gradient independently of the twist angle.

So far, we have focused on bMLGs with a visible moiré pattern. However, for large twist angles (>5°), the resulting moiré wavelength is below our resolution limit. For those systems, the strain can instead be probed from optical measurements. Shown in Fig. 3A is a bMLG on hBN where no moiré was detected in the PFM scans, which indicates a twist angle larger than 10° (31). The corresponding strain map extracted from Raman spectroscopy is seen in Fig. 3B. The strain is calculated from the position of the 2D peak, where we expect a shift of ~27 cm−1 per percent of strain (40). Spectra taken along various positions to the neutral axis are shown in Fig. 3C, with the red and blue curves corresponding to tensile and compressive strain, respectively. Using multiple peak positions at a constant x position, we extracted the strain evolution along y (Fig. 3D), with the dashed line showing the strain gradient expected for a 1.6-μm-wide ribbon deflected by d0 = 300 nm.

Shown in Fig. 3E is a bent monolayer tungsten diselenide (WSe2) ribbon on hBN, demonstrating the capability to apply this technique to 2D materials other than graphene. Because of the substantial lattice mismatch between these two materials, no moiré superlattice was observed. We instead used nanoPL to determine the strain in the WSe2. When subject to strain, the direct band gap of WSe2 is expected to shift by ~50 meV per percent (41). Figure 3F is a spatial map of the strain distribution in the WSe2 ribbon calculated from the shift of the peak wavelength (Fig. 3G). Similar to bMLG, we observed a strain gradient evolving from tensile (Fig. 3H, blue) to compressive (Fig. 3H, red) strain and with a linear slope that matches theory (Fig. 2H).

Ultralow disorder in bBLG

Last, we exploited the bent geometry to fabricate twisted-bilayer graphene with continuously varying twist angles through the magic angle. An optical image of the bBLG is shown in Fig. 4A (details of the device fabrication are provided in fig S2). We performed LFM scans along the ribbon and report the results for five different scan areas, all near the region where the magic angle is reached. The scan locations, shown schematically at the bottom of Fig. 4A, span in total 1 μm in the x direction and 2.5 μm in the y direction. We show one of these scans with the corresponding FFT in the Fig. 4B inset; the remaining can be found in figs S10 and S11. The real-space image shows a notably uniform moiré pattern, with little evidence of varying twist angle or notable distortion. We observed similar high-quality moiré patterns in each of the scan regions. We extracted the moiré wavelength from the FFTs and used Eq. 2 to calculate the twist angle. Repeating this for different positions along the ribbon, we found that the twist angle between the layers varies from θ = 2.5° in the unbent region to θ = 1° at the end of the bBLG (fig S10), with an approximately linear gradient of 0.095° per micrometer.

Although we can extract an average twist angle over a single LFM scan from its FFT, the high quality of our scans opened the possibility to analyze the real-space moiré pattern and extract the local twist angle and strain by locating the center of each moiré site (Fig. 4B, bottom right). The spatial distributions of twist angles and strain, extracted from Fig. 4B, are shown in Fig. 4, C and D, respectively. Whereas the overall twist-angle variation remains small (±0.05°), we observed a gradient of the twist angle along the x axis (bending axis) and minimal evolution along the y axis. For the strain, we observed highly uniform values of around ~0.05% over the whole area, which is in line with the visually uniform pattern seen in Fig. 4B.

To characterize the apparent twist-angle gradient, we averaged twist-angle values along the y axis, and we report this average as a function of the position along the x axis, or bending axis. We repeated this averaging for all five scans, and we report the twist-angle evolution in Fig. 4E. We observed a uniform gradient of the twist angle of 0.095° per micrometer, which is identical to the global twist-angle gradient found with FFT peak positions. The identical local and global twist-angle gradients confirmed that the twist-angle evolution along the bBLG is smooth and continuous.
over the whole bMLG. Having established the bend-induced twist-angle gradient as the primary source of angle variation in our samples, we set out to estimate the intrinsic twist-angle disorder as a deviation from the global twist-angle gradient. The resulting distribution of the gradient-corrected twist angles is presented in Fig. 4F. We found an intrinsic disorder value of 0.0074°, which is around three times lower than results obtained with conventional twisted-bilayer samples prepared by using a tear and stack method (20, 38).

Discussion and outlook

The ability to precisely tune the twist angle and strain within a 2D heterostucture, in the absence of uncontrolled distortions, paves the way for moiré band structure engineering in the disorder-free limit, including the exciting possibility that moiré patterning can be used as a generalized quantum simulation platform to study strongly correlated physics and topology in quantum materials (8). The dramatically reduced moiré disorder observed in our bent geometry is not yet understood. We conjecture that this may relate to the lattice relaxation dynamics in the presence of an externally applied strain field, but further theoretical and experimental work will be required to fully understand both the origin of this behavior and how this interplay may be exploited to realize new control opportunities. Last, the reversible in-plane bending geometry that we demonstrate, realized through local mechanical actuation, provides an alternative approach toward generalized strain engineering (42, 49) beyond moiré patterning.

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SUPPLEMENTARY MATERIALS

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Materials and Methods
Supplementary Text
Figs. S1 to S15
References (46–61)
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