Superlattice Structures in Twisted Bilayers of Folded Graphene

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The electronic properties of bilayer graphene strongly depend on relative orientation of the two atomic lattices. Whereas Bernal-stacked graphene is most commonly studied, a rotational mismatch between layers opens up a whole new field of rich physics, especially at small interlayer twist. We investigate magnetotransport measurements on twisted graphene bilayers, prepared by folding of single layers. These reveal a strong dependence on the twist angle, which can be estimated by means of sample geometry. At small rotation, superlattices with a wavelength in the order of 10 nm arise and are observed by friction atomic force microscopy. Magnetotransport measurements in this small-angle regime show the formation of satellite Landau fans, which are attributed to additional Dirac singularities in the band structure.

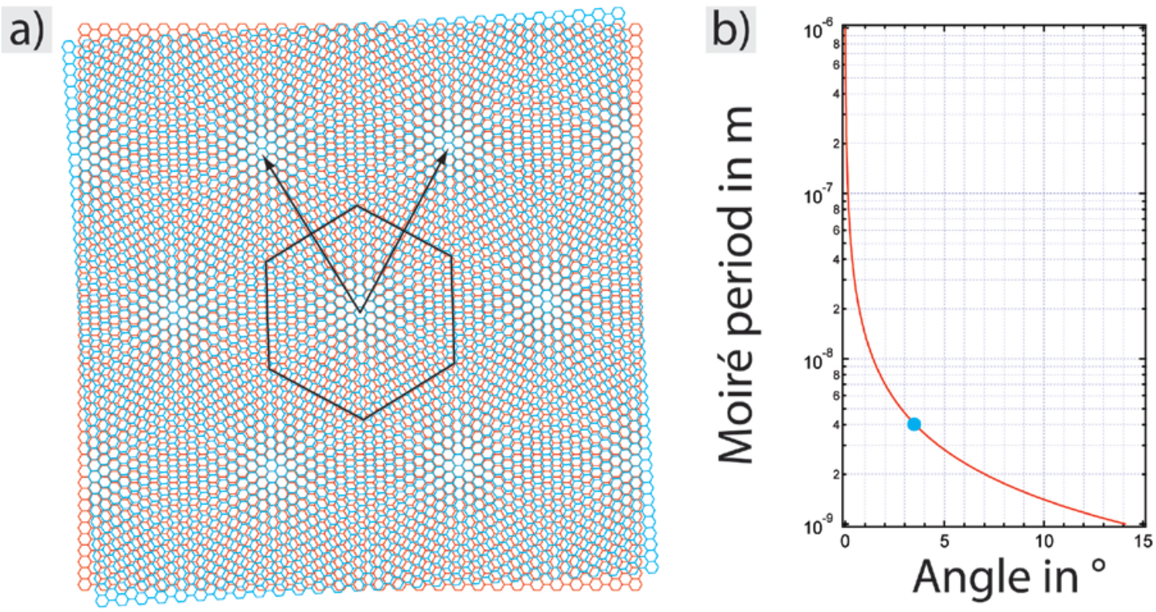

Fig. 1: (a) Two rotated honeycomb lattices arranging in a moiré superstructure. **(b)** Dependence of the superlattice periodicity on the twist angle.

The electronic structure of bilayer graphene can significantly differ from that of a single layer, depending on the interlayer stacking order. Apart from the common Bernal-stacked variety, twisted graphene bilayers (TBGs) constitute a whole new field of their own: Layers of large rotational mismatch effectively decouple, exhibiting reduced Fermi velocities for decreasing interlayer twist in many cases[1,2]. At the smallest angles, totally different electronic structures are expected[3,4]. While TBGs of various angles have been grown and optical studies as well as scanning tunnelling spectroscopy were performed on samples of different interlayer twist, we here present a study on magnetotransport in high-quality folded monolayers of small interlayer twist[0], revealing novel transport features in the form of satellite Landau fans, caused by twist-induced long-wavelength superlattices (Fig. 1).

Our folded layers are obtained by mechanical manipulation via atomic force microscope (AFM) or incidental flip-over during the exfoliation of natural graphite. As graphene is most commonly terminated by armchair- or zigzag-edges, according crystallographic reference directions in relation to the folded edge (relative angle φ) can be used to estimate the interlayer rotation by $\theta=2\cdot\varphi$. In case of the shown example (Fig. 1a), the twist angle can thus be narrowed down to 1.5°±0.5° (projected into the range of $0°<\theta\leq30°$ due to graphene's sixfold symmetry). The two rotated lattices may now arrange in periodic superstructures reproducing the original honeycomb pattern on a twist-dependent length scale of $\lambda(\theta)=a/[2\sin(\theta/2)]$, a being the length of graphene's lattice vector. Figure 1c shows a lateral force microscopy scan on the TBG area. The dashed white star marks three distinct symmetry directions in the friction force plot, which are clearly confirmed by the prominent hexagonal pattern in the Fourier transform (Fig. 1c) pointing to a trigonal lattice of period λ=9 nm. The corresponding moiré twist angle of 1.6° fits the geometrically estimated value of 1.5°±0.5° very well.

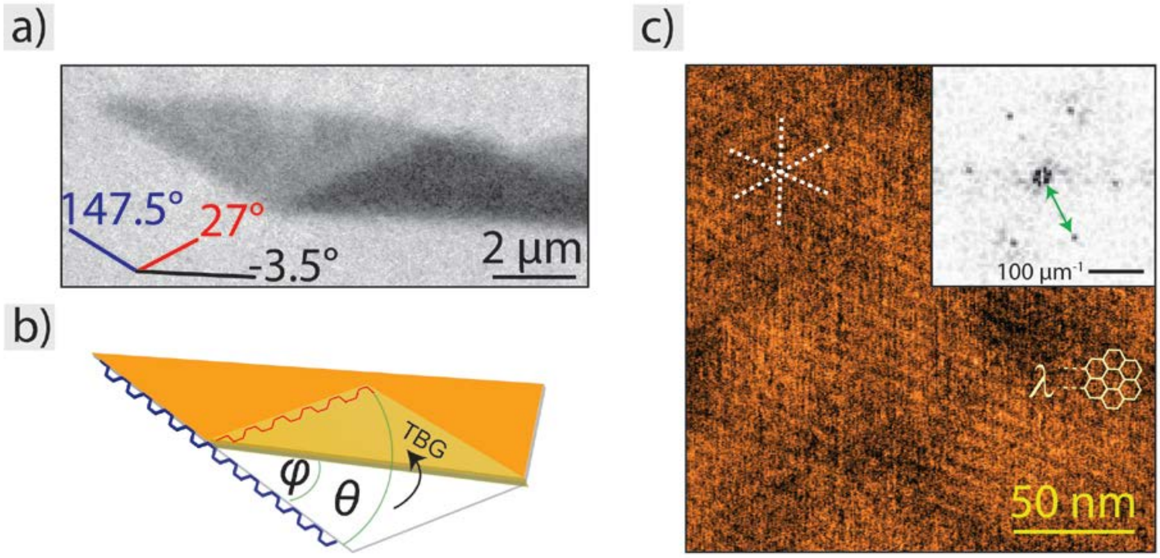

Fig. 2: (a) Optical picture of a folded graphene sample. The orientation of edges can be used to estimate interlayer twist, as illustrated in the schematic in **b**. **(c)** AFM scanning image of the TBG in friction force mode and Fourier transform in the inset.

To investigate the electronic properties of graphene bilayers with different stacking, magnetotransport measurements in perpendicular fields up to B=13 T were performed. For several samples of small rotational mismatch ($\theta < 3°$), systematic satellite Landau fans could be observed in longitudinal resistance, as depicted in the example in figure 3a (main fan originating at charge neutrality, secondary fan origination at $n = 2.75 \times 10^{12}$ cm^{-2}). This indicates the generation of secondary Dirac singularities in the band structure comparable to observations on heterostructures of closely aligned graphene on hexagonal boron nitride[5]. This phenomenon can be attributed to a large wavelength moiré pattern between the two rotated hexagonal lattices. Bragg scattering by the correspondingly small superlattice Brillouin zone (mini Brillouin zone, mBz) will lead to replica satellite Landau fans at higher energies (Fig. 3b), while periodically alternating interlayer coupling should result in a more complex electronic spectrum[3,4], as discussed in [0].

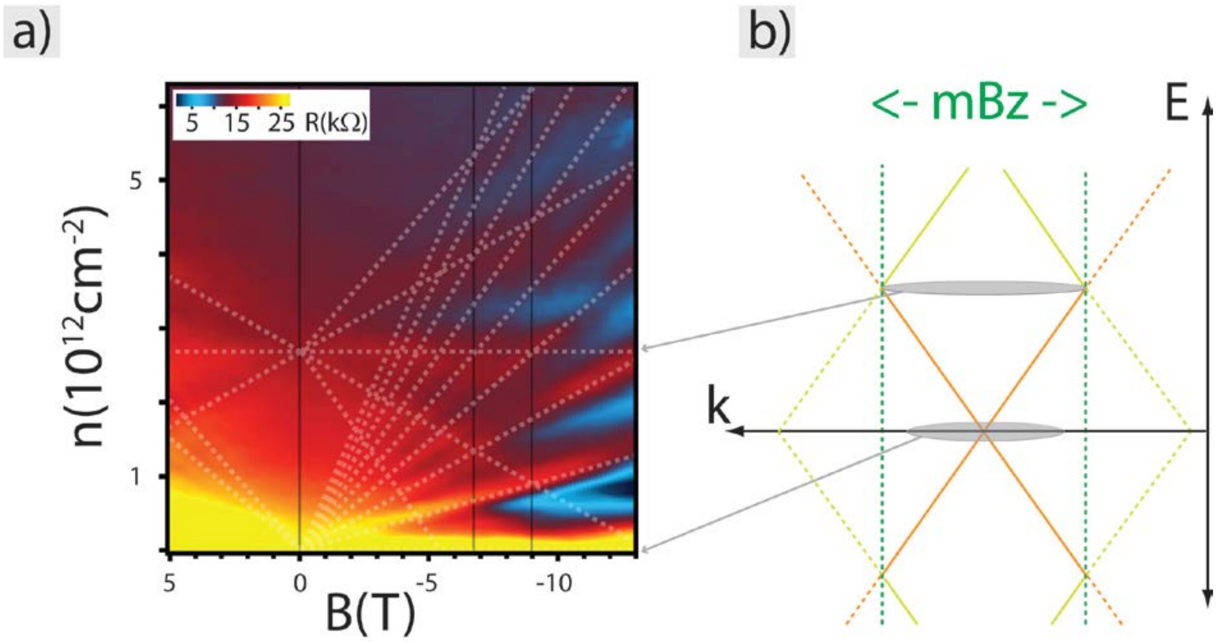

Fig. 3: (a) Longitudinal resistance vs. magnetic field and charge carrier density for a twisted bilayer of small rotation. Dashed white lines trace main and satellite Landau fan. **(b)** Schematic of graphene´s low energy dispersion (orange). Reflection on the mini Brilloun zone (green) leads to replica Dirac points at higher energies.

References

[0] Schmidt, H., Rode, J. C., Smirnov, D. & Haug, R. J. Superlattice Structures in Twisted Bilayers of Folded Graphene. *Nat. Commun.* **5**, 5742 (2014).

[1] Lopes dos Santos, J. M. B., Peres, N. M. R. & Castro Neto, A. H. Graphene Bilayer with a Twist: Electronic Structure. *Phys. Rev. Lett.* **99**, 256802 (2007).

[2] Schmidt, H., Lüdtke, T., Barthold, P. & Haug, R. J. Mobilities and Scattering Times in Decoupled Graphene Monolayers. *Phys. Rev. B* **81**, 121403(R) (2010).

[3] Mele, E. J. Band symmetries and singularities in twisted multilayer graphene. *Phys. Rev. B* **84**, 235439 (2011).

[4] Bistritzer, R. & MacDonald, A. H. Moiré bands in twisted double-layer graphene. *Proc. Natl. Acad. Sci. USA* **108**, 12233-12237 (2011).

[5] Hunt, B. *et al.* Massive Dirac fermions and Hofstadter butterfly in a van der Waals heterostructure. *Science* **340**, 1427-1430 (2013).