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Universal Torsional Periodic Lattice Distortion in Twisted 2D Materials

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At low twist angles, moiré heterostructures periodically restructure and spontaneously break symmetry to form complex superstructures [1,2,3]. Here, we use a torsional periodic lattice distortion (PLD) model [4] to concisely describe the relaxed superstructure and simulate qualitatively accurate electron diffraction patterns across a variety of twisted 2D materials.

Periodic restructuring of twisted 2D materials is a direct consequence of competition between interlayer van der Waals registry energy and intralayer elastic energy cost. For example, moiré of twisted bilayer graphene (TBG) periodically unwinds energetically favorable AB/BA stacked regions to increase AB/BA area, and unfavorable AA stacked regions are further twisted to decrease their area. While the reciprocal structure of undistorted twisted 2D materials is a simple superposition of two constituent layers [5], periodic restructuring diffracts swift electrons into faint superlattice peaks around each Bragg peaks (Fig. 1a). The appearance of strong superlattice peaks around higher order Bragg peaks is a signature of PLDs. Notably, the distribution of superlattice peaks is stronger along the azimuthal direction; this is a signature for transverse PLDs [6,7]. These azimuthally distributed superlattice peaks appear universally among a variety of twisted 2D materials (Fig. 1b–d).

We use torsional PLD model to describe the in-plane distortion of twisted 2D materials. The torsional PLD ($\boldsymbol{\Delta}_{\mathrm{N}}$) is made-up of three non-orthogonal, transverse distortion waves of equal amplitudes (A_n) and harmonics thereof ($\boldsymbol{\Delta}_{\mathrm{n}}$):

$$\boldsymbol{\Delta}_{\mathrm{N}} = \sum_{\mathrm{n}=1}^{\mathrm{N}} \boldsymbol{\Delta}_{\mathrm{n}} \quad \cdots (\text{Eq. } 1) \qquad \boldsymbol{\Delta}_{\mathrm{n}} = \mathrm{A}_{\mathrm{n}} \sum_{i=1}^{3} \widehat{\mathbf{A}}_i \sin(n\boldsymbol{q}_i \cdot \boldsymbol{r}_0)\,; \qquad \widehat{\mathbf{A}}_i \perp \boldsymbol{q}_i \quad \cdots (\text{Eq. } 2)$$

$\mathbf{r}_0$, $\hat{\mathbf{A}}$, $\mathbf{q}$ denotes undistorted atom positions, PLD displacement direction, and PLD wavevector. Three $\mathbf{q}_i$'s are primitive reciprocal moiré lattice vector that are 120° apart.

Torsional PLDs in twisted 2D materials are a universal phenomenon at low twist angles and not limited to TBG [2,3,9,10]. Figure 1 shows SAED patterns that exhibit periodic relaxations of four distinct twisted 2D systems: a) low twist angle TBG, b) 4-layer (4L) of WS_2, c) twisted double bilayer CrI_3 and d) twisted WS_2/$MoSe_2$ heterostructure. For 4L-WS_2 (Fig. 1b), a surprisingly strong torsional PLD is observed, despite its large twist angle (θ≅4°) [2]. Figure 1c shows 4 layers of twisted CrI_3 but twist only between middle two layers. Xie et al. reported that this system shows magnetic behavior that cannot be explained by either 2-layer or 4-layer CrI_3 and periodic relaxation must be accounted for [3]. Figure 1d) shows that the periodic reconstruction in twisted materials is not limited to homostructures. WS_2/$MoSe_2$ heterostructure exhibit twist angle dependent excitonic behavior [11]. In Figure 5d, we revealed that the heterostructure with large twist angle ($\theta \cong 5°$) periodically relaxes, despite having difference lattice constants. Quantum mechanical multislice simulation [8] of electron diffraction patterns (Fig. 1 i–iii) with torsional PLDs applied show good agreement with the experimental diffraction patterns. Notably, for near magic-angle TBG, single-harmonic torsional PLD ($\mathbf{\Delta}_1$) matches quantitatively with the experimental SAED patterns [4].

By including higher harmonics ($\mathbf{\Delta}_n$), torsional PLD can express any arbitrary periodic relaxation pattern. For low twist angle TBG, single harmonic torsional PLD model does not fully reproduce experimental relaxation behavior and higher order superlattice peaks are more pronounced. Figure 2 depicts torsional PLD with single (Fig. 2d), triple (Fig. 2e) and 7- (Fig. 2f) harmonics. The length and direction of arrows describe local displacements, and the colored overlay shows amount of local rotation ($\propto (\mathbf{\nabla} \times \mathbf{\Delta})$). All three describes a periodic twist/anti-twist displacements while inclusion of higher harmonics introduces sharper features. Multislice simulated diffraction patterns (Fig. 2g–o) shows that higher harmonics enhance higher order superlattice peaks—qualitatively matches low twist angle TBG.

A torsional PLD model reduces the complexity of low-twist angle moiré crystals to a single order parameter across a variety of 2D materials ranging from graphene, metal dichalcogenides, metal trihalides homostructures to heterostructures of 2D materials.

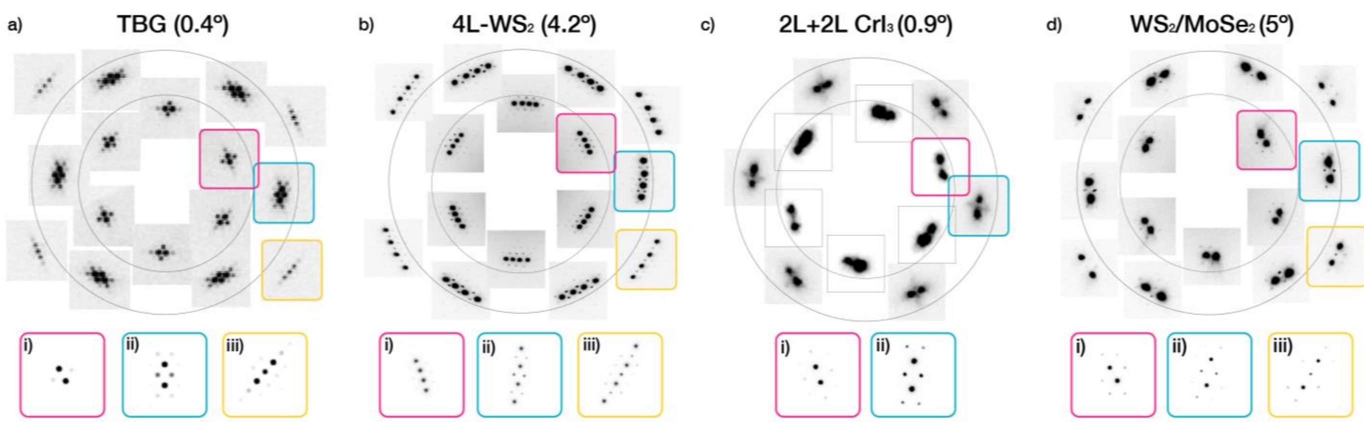

Figure 1. **Universal Torsional PLD relaxation of twisted 2D materials** Periodic relaxation is observed universally in multiple twisted 2D systems. SAED of a) low-θ TBG, b) twisted four layer (4L) WS_2 homostructure c) twisted bilayer (2L + 2L) CrI_3 d) twisted WS_2/$MoSe_2$ heterostructure shows bright Bragg peaks with small superlattice peaks. Insets i–iii are multislice simulated diffraction patterns with torsional PLD model. The torsional PLD model reproduces qualitatively accurate SAED patterns across multiple systems.

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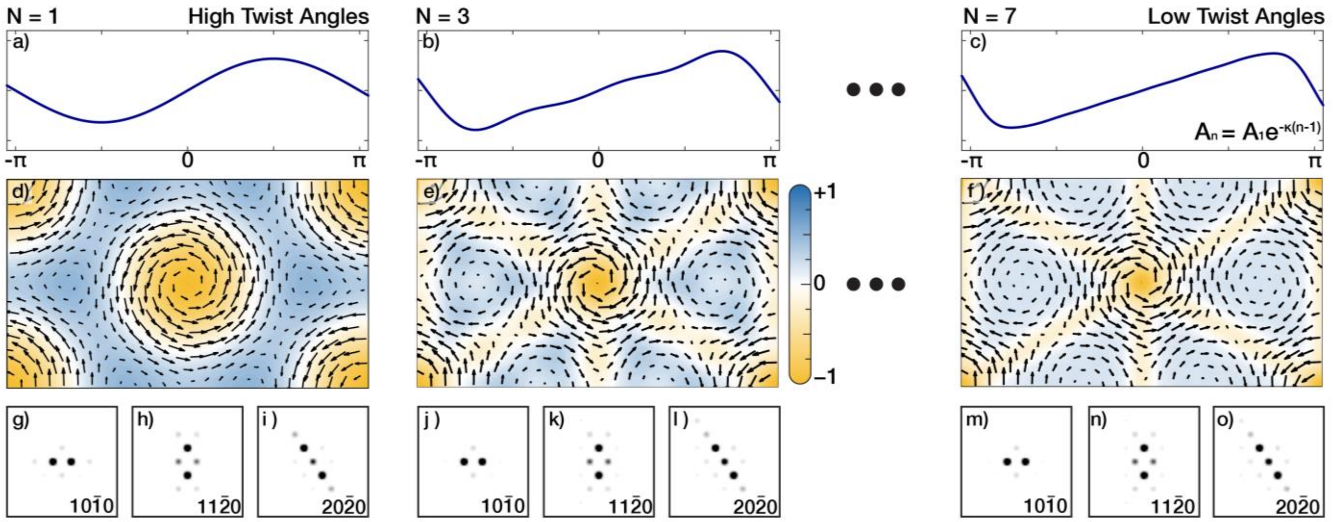

Figure 2. **PLDs as Fourier series in 2D moiré materials:** a–c) Evolution of periodic wave as higher harmonic waves (b) N=3, c) N=7) are included. Fourier coefficients (A_n) are tailored as exponential decay, which produces to a smooth `sawtooth'-like waveform. Including harmonic waves allows high frequency (i.e. sharp) features in resultant waves. d–f) Torsional PLD structure with higher harmonic PLD included. The color denotes the amount of local rotation $((\propto (\boldsymbol{\nabla} \times \boldsymbol{\Delta}_N))$ due to the PLD displacement field (arrows). g–i) Quantum mechanical electron diffraction simulations of TBG with single harmonic torsional PLD captures the distortions in high twist angles (i.e. near magic-angle and higher) well. j–o) Adding higher harmonics slightly modifies the diffraction patterns and shows qualitatively better matches with low twist angle systems.

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