



HAL
open science

Strain Relaxation and Excitonic Absorption of Atomically-Reconstructed WSe₂ Moiré Superlattices

Steffi Woo, Fuhui Shao, Nianjheng Wu, Robert Schneider, Ashish Arora, Johann Preuß, Benjamin Carey, Steffen Michaelis de Vasconcellos, Andrew Mayne, Rudolf Bratschitsch, et al.

► **To cite this version:**

Steffi Woo, Fuhui Shao, Nianjheng Wu, Robert Schneider, Ashish Arora, et al.. Strain Relaxation and Excitonic Absorption of Atomically-Reconstructed WSe₂ Moiré Superlattices. Physical Sciences Symposium on Advanced Imaging and Spectroscopy for Nanoscale Materials, American Physical Society, Mar 2022, Chicago, United States. pp.2462-2463, 10.1017/S1431927622009424 . hal-04304699

HAL Id: hal-04304699

<https://hal.science/hal-04304699>

Submitted on 28 Nov 2023

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

Strain Relaxation and Excitonic Absorption of Atomically-Reconstructed WSe₂ Moiré Superlattices

Steffi Y. Woo¹, Fuhui Shao¹, Nianheng Wu^{1,2}, Robert Schneider³, Ashish Arora^{3,4}, Johann A. Preuß³, Benjamin J. Carey³, Steffen Michaelis de Vasconcellos³, Andrew J. Mayne², Rudolf Bratschitsch³, Luiz H.G. Tizei^{1,*}

¹Laboratoire de Physique des Solides, Université Paris-Saclay, CNRS, Orsay, France

²Institut des Sciences Moléculaires d'Orsay, Université Paris-Saclay, CNRS, Orsay, France

³Physikalisches Institut, Westfälische Wilhelms-Universität Münster, Münster, Germany

⁴Indian Institute of Science Education and Research, Dr. Homi Bhabha Road, 411008 Pune, India

* Corresponding author: luiz.galvao-tizei@universite-paris-saclay.fr

Atomically-thin layers of semiconducting transition metal dichalcogenides (TMDs), namely tungsten diselenide (WSe₂), have attracted considerable interest because of new properties that can be obtained when artificially fabricated into van der Waals superlattices. Interlayer coupling is responsible for various properties, including the indirect-to-direct band gap cross-over from bulk and few-layered to monolayer TMDs. It has also been demonstrated that twist angle can be used to tune the interlayer coupling strength in bilayer MoS₂ [1] and WS₂ [2] homostructures. In this work, the high spatial and spectral resolution of monochromated electron energy-loss spectroscopy (EELS) coupled with four-dimensional scanning transmission electron microscopy (4D-STEM) in an aberration-corrected STEM are used to investigate the excitonic response of twisted bilayer WSe₂ with a low moiré angle and the strain relaxation of those that are atomically reconstructed.

Monolayered WSe₂ flakes were mechanically exfoliated from synthetic bulk crystals, and transferred onto SiO₂/Si wafers prior to assembly. The twisted bilayer WSe₂ structures were fabricated using a viscoelastic ‘tear and stack’ stamping method for controlled stacking and twist angle [3], before depositing onto a Quantifoil holey carbon TEM grid. Monochromated EELS and nano-beam electron diffraction with 1 mrad convergence angle were performed on a modified Nion HERMES-S200 (or ChromaTEM) operated at 60 kV with the sample cooled using liquid nitrogen (T ~ 150 K).

Well-defined hexagonal moiré patterns with nanometer periodicity are apparent for the low twist angles with high-resolution imaging. At twist angles below ~3°, some of the high-symmetry stacking regions become more energetically favorable, leading to an expansion of selected high-symmetry points into domains by atomic reconstruction. A low-angle annular dark-field (LAADF) image can be used to visualize the domain boundaries [4]. The domain boundary geometry has been demonstrated to differ between those of R-type (AA) and H-type (AA') stacking order in twisted TMDs that designates layers without and with inversion symmetry, respectively. R-type takes on a triangular geometry [Fig. 1(b,c)], while H-type resembles a kagome-like pattern [Fig. 1(d)]. 4D-STEM of the reconstructed bilayers reveals the nanoscale deformation that correlates perfectly with the location of the domain boundaries [Fig. 2]. The excitonic absorption signatures of these nanometric twisted bilayers [Fig. 1(e)] demonstrate a good general correspondence to the thickness dependence of the exciton resonances, namely a pronounced decrease in C exciton energy with the number of layers [5]. Comparing different twist angles in the bilayers also shows sizable blueshifts in the C exciton energy up to 200 meV, with extremes between the zero-twist and towards the anti-aligned (28°) case, indicating underlying differences in interlayer coupling with respect to the moiré angle.

Acknowledgements: The authors acknowledge funding from the ANR, program of future investment TEMPOS-CHROMATEM (No. ANR-10-EQPX- 50), and support by the European Union in the Horizon 2020 Framework Program (H2020-EU) under Grant Agreement No. 823717 (ESTEEM3).

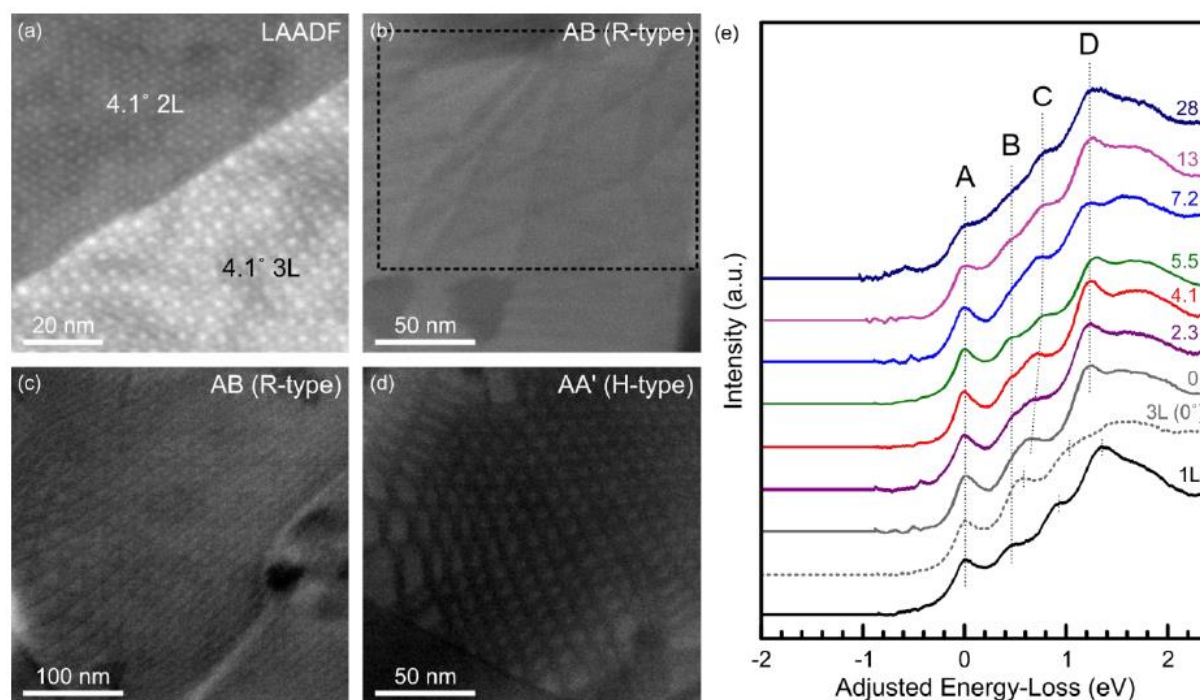


Figure 1. STEM-LAADF images of WSe₂ twisted bilayers (2Ls) with (a) 4.1° twist angle, and (b,c) AB (R-type) stacking, (d) AA' (H-type) stacking with atomic reconstruction. (e) Monochromated EELS spectra from twisted bilayers with various moiré angles compared to a representative monolayer (1L) and trilayer (3L) with zero twist. Spectra are offset in energy relative to the A exciton, and the dotted lines are to help guide the eye in the different exciton energy positions.

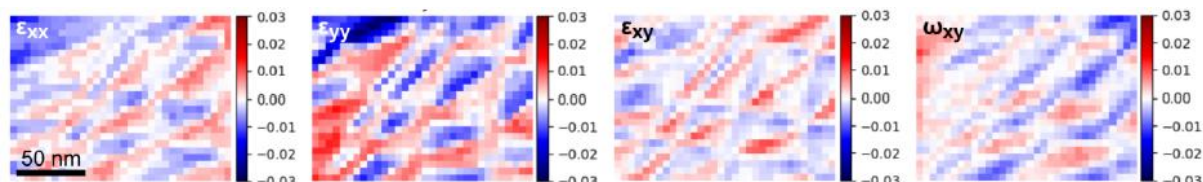


Figure 2. Strain maps obtained using 4D-STEM of the boxed region from Fig. 1(b), including axial strains, ϵ_{xx} , ϵ_{yy} , shear strain, ϵ_{xy} , and rotation ω_{xy} , showing strain relaxation corresponding to the distribution of alternating AB/BA domains, as a result of the atomic reconstruction.

References:

- [1] A. Castellanos-Gomez, H.S.J. van der Zant, & G.A. Steele, *Nano Research* **7**, 572–578 (2014).
- [2] W. Yan, L. Meng, Z. Meng, Y. Weng, L. Kang, L. and X. Li, *The Journal of Physical Chemistry C* **123**(50), 30684–30688 (2019).
- [3] K. Kim *et al.*, *Proceedings of the National Academy of Sciences* **114**(13), 3364–3369 (2017).
- [4] A. Weston, Y. Zou, Y., V. Enaldiev *et al.*, *Nature Nanotechnology* **15**, 592–597 (2020).
- [5] A. Arora, M. Koperski, K. Nogajewski *et al.*, *Nanoscale* **7**, 10421 (2015).