Peer Review File

Torsional Periodic Lattice Distortions and Diffraction of Twisted 2D Materials

Reviewer Comments, first round -

Reviewer #1 (Remarks to the Author):

Sung et al present a mathematical description of periodic lattice relaxation in twisted 2D heterostructures. The modulation of stacked 2D lattices when twisted to small angles is an important topic in physics and material science with the effect of lattice relaxation having previously shown to modify local band structure. It is also a subject where the modulation has been mostly described in a qualitative rather than a quantitative manner in existing literature. This work also presents the verification of the reconstruction behaviour through the observation of superlattice spots in the diffraction data making it applicable even for materials which are highly electron beam sensitive. The manuscript therefore offers the potential for significant enhancement over existing work.
However, the key description of torsional periodic lattice distortions is verified by comparison of "multislice" diffraction simulations with experimental data. Little information is given regarding how the diffraction spot simulations are achieved and we have been unable to reproduce these results from the methodological information provided in the manuscript. Particularly, the size of the real space supercell necessary for small diffraction angles and the small step size required in reciprocal space to effectively resolve superlattice reflections location and intensity, results in a complex multislice simulation which is hard to achieve using the widely adopted E. Kirkland multislice code [ref 31] and the processing power of a conventional PC.
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In this paper by Sung and co-workers, a torsional periodic lattice distortion (PLD) model is proposed to describe lattice reconstruction in twisted 2D materials. Theoretical results, including simulated electron diffraction patterns and displacement fields as well as strain and stacking energy calculations are accompanied by experimental selected area diffraction data. The orientations and relative intensities of superlattice peaks observed in diffraction provide evidence of transverse PLDs in moiré superlattices that evolve as a function of twist angle.

Discussion of lattice reconstruction in twisted 2D materials is currently of significant interest due to its impact on the exotic physical behavior observed in these systems. This paper provides an atomic scale description of the reconstruction process, whereas current literature has thus far focused on nanoscale pictures of lattice relaxation. The authors also show how the PLD model explains the changing complexity of reconstruction when transitioning from a low-twist to an extreme low-twist angle regime, which has been previously observed experimentally but not fully understood. Overall, the results from this work build nicely on the existing literature on lattice reconstruction (i.e. DF-TEM, 4D-STEM and HAADF-STEM studies, theoretical predictions) and fill in important gaps in the understanding of how reconstruction occurs in twisted 2D systems. I recommend this paper for publication in Nature Communications once the following comments are addressed:

a. It would be very insightful if more detail were provided regarding the application of the PLD model to the multilayer systems shown in Figures 5b and c. Should one assume that the model described for the bilayer in the Methods section can be directly applied to the 4L system by continuing to alternate the sign of the PLD amplitude? For the 2L + 2L system, what happens in the outermost untwisted layers? Do they pin the distortions of the innermost layers, or do they also undergo PLD?

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To clarify this, the main text on page 5 now reads "… For multi-layered system, relaxation may not be equivalent between layers. For 4L-WS_2, for example, the PLD amplitudes are strongest for the inner-most layers. Here equal and opposite PLDs of the inner two layers matches simulated diffraction patterns (Fig. 5b)."

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If they have been rotated, then this would answer my questions but should be mentioned. If they have not been rotated, then 1) why are the superlattice peaks oriented azimuthally instead of radially with respect to the primary Bragg peaks? Is this because the twist angle is so large? And 2) why do the WS2 and MoSe2 Bragg peaks have approximately the same radial distance?

Reviewer #3 (Remarks to the Author):

Dear Authors,

Thank you for your responses to the comments and queries raised about the original manuscript submission. I am pleased to see a more complete description of the simulation parameters used and a deeper analysis of some of the other twisted 2D materials presented.

I am happy that the original review comments have been addressed satisfactorily and I am happy to approve the revised manuscript for publication

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We appreciate the response and glad to hear our revisions are well received.

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We now provide simulated diffraction data that includes PLD in the outermost layers to confirm whether additional superlattice peaks arise from the relaxation of those layers. Diffraction of torsional PLDs in 4L-WS$_2$ with different amplitudes in each layer are now shown in Supplemental Figure 11. To be comprehensive, we make no assumption about the energy landscape and simulate several PLD direction directions. As the reviewer intuited, the outer most superlattice peaks are more consistent with the data when a small PLD is present in the outer layers and Figure 5b has been updated for better accuracy. The conclusions and statements pertaining to Fig.5 remain unchanged—a qualitative demonstration of PLD relaxation behavior across different twisted 2D materials.

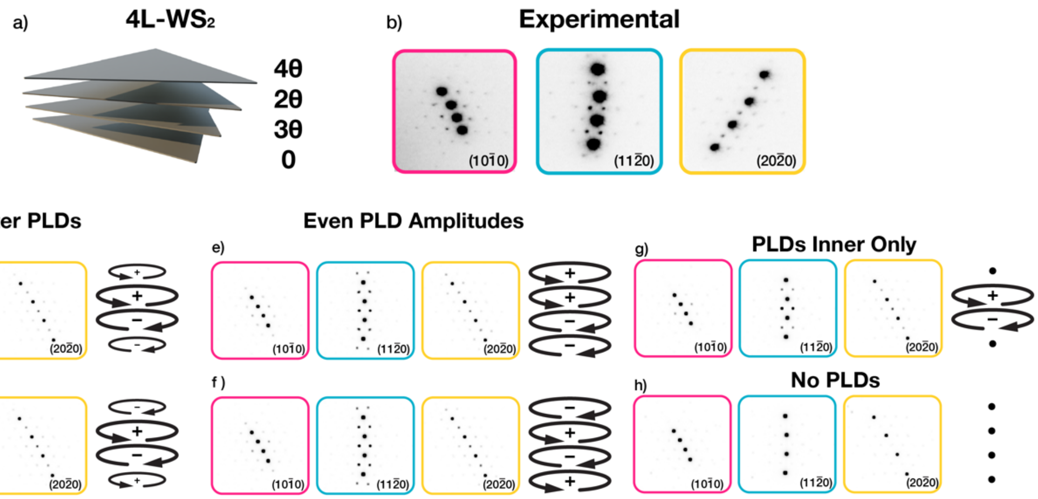

Fig. S11 | **Torsional PLDs in 4L-WS$_2$** a) Schematic diagram of 4L-WS$_2$ showing twist configuration of each layer. b) Experimental SAED patterns for 4L-WS$_2$. c–h) Simulated electron diffraction patterns for 4L-WS$_2$ with different torsional PLD amplitudes across layers. Accompanying pictographs denotes sign (+/–) and amplitude of torsional PLDs. c, d) PLD amplitudes are weaker (50%) in outer two layers than in inner layers. e, f) PLD amplitudes are equal in all four layers. g) PLDs exists in inner two layers with no PLD in outer layers. h) Simulations without PLDs. c, e) The sign of PLD in upper two and bottom two layers are equal. d, f) PLD sign alternates. In all cases, superlattice peak intensities are qualitatively similar, while some superlattice peaks are different. Experimental SAED patterns (Fig. 5b) matches closely with c) and d).

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We thank the reviewer for calling attention to a severe typo causing undue confusion. The heterostructure presented in this manuscript is $MoSe_2/WSe_2$ not WS_2. $MoSe_2$ and WSe_2 have comparable lattice constants ($\lesssim$ 1%). This explains why the longitudinal component of the PLD is not apparent in figure 5d. This correction has been made and we now also include the full diffraction pattern of the heterostructure as Supplemental Figure 12. We are sincerely grateful for the attention here.

Reviewer Comments, third round -

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