

The Importance of Relaxation Effects on the Properties of Twisted van der Waals Materials

Florian M. Arnold, Agnieszka Kuc, Jens Kunstmann, Thomas Heine

TU Dresden, Theoretische Chemie, Bergstraße 66c, D-01069 Dresden, Germany

HZDR, Institut für Ressourcenökologie, Permoserstraße 15, D- 04318 Leipzig, Germany

Florian.Arnold@tu-dresden.de

The stacking of single layer 2D crystals, such as transition-metal dichalcogenides, creates a new class of materials: van der Waals [hetero]structures. [1] In these materials, a variety of structures can be realized by controlling the stacking order and the twist angle between adjacent layers. In our work, the influence of the twist angle on the structural and electronic properties of MoS₂ bilayers was studied using reactive force field (ReaxFF) and density functional theory (DFT) calculations.

Interlayer twisting leads to the formation of moiré patterns of rapidly increasing size with decreasing twist angles towards 0° and 60°, resulting in huge simulation cells that are only accessible computationally using force fields. [2] This concept of constructing twisted bilayers, by a rigid rotation between the individual layers, is used in many studies. However, it neglects the importance of relaxation effects – these result in a significant deformation of the individual layers, especially for small twist angles, and strong changes of the interlayer distance in different areas of the structure. Our work gives a detailed overview of the influence of relaxation on the structural properties in such systems and how different structural regimes – soliton, transition, and moiré regime – form depending on the interlayer twist angle. We further demonstrate how these changes can strongly influence the analysis of electronic properties, as shown in the Figure 1, thus emphasizing the need for thorough relaxation in computational studies of van der Waals [hetero]structures.

References

- [1] A. K. Geim, I. V. Grigorieva, *Nature*, 499 (2013) 419.
- [2] G. C. Constantinescu, N. D. Hine, *Physical Review B*, 91 (2015) 195416

Figures

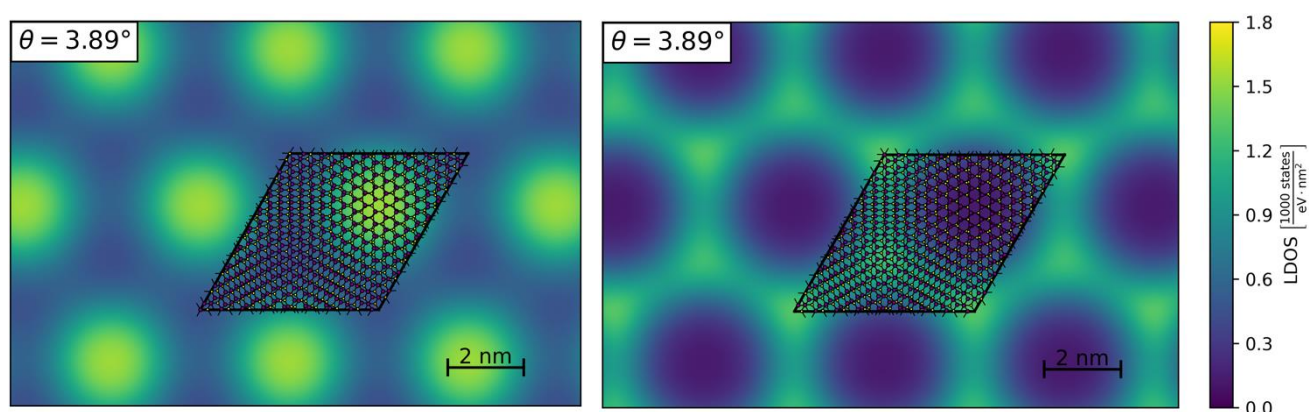


Figure 1: Atom-projected density of states map in the valence band region ($E = -0.268$ eV) of a bilayer MoS₂ with a twist angle of 3.89°. The atomic structure is included for the primitive moiré cell. In the case of (left) rigidly twisted layers the valence band is localized in strongly confined regions. Extensive changes can be observed in the case of (right) a fully relaxed structure where a delocalized hexagonal network, associated with the appearance of strain solitons, is formed.