Local Surface Crystal Structure of 2D materials Characterized by Dynamical μLEED-*IV*/LEEM Analysis

Z. Dai[1*], W. Jin[2], J. Sadowski[3], R. M. Osgood Jr.[2] and K. Pohl[1]

[1]Department of Physics and Materials Science Program, University of New Hampshire, Durham, NH, 03824, USA
[2]Columbia University, New York, NY
[3]Center for Functional Nanomaterial, Brookhaven National Lab

Email: zbr5@wildcats.unh.edu

Two-dimensional (2D) materials have attracted much attention as an emerging category of materials over the past decade due to their novel mechanical, optical and electronic properties with many potential applications in photovoltaics, photo-catalysts, and modern electronics. However, the detailed atomic structural information has been rarely experimentally investigated due to the following difficulties: (*i*) the limited sample size of 2D materials prepared through mechanical exfoliation of a few μm, and (*ii*) the easy oxidation and surface instability of various 2D materials under high energy probing techniques. Selected area low-energy electron diffraction analysis (μLEED-*IV*) performed in a low-energy electron microscopy (LEEM) system, is a modern surface sensitive and non-intrusive surface characterization technique, which has the advantage of μm sampling size selectivity. I present, for the first time, detailed experimental characterizations of atomic crystal structures of a series of technologically promising 2D materials: MoS_2 [1], black phosphorus (BP) [2] and the topological crystalline insulator (TCI) SnSe [3]. We find a slight asymmetry of the relaxation of the interlayer spacing of a suspended single S-Mo-S sandwich layer, which is most likely caused by a small amount of warping or strain. In the case of both bulk BP and exfoliated few layer phosphorene (FLP), with a thickness of about 10 nm, we find that the surface undergoes a significant distortion in the form of buckling of 0.2 Å, an order of magnitude larger than two previously reported theoretical values. Using first-principles calculations, we propose a vacancy defect driven mechanism as cause of this surface distortion. The topological properties of the rock-salt TCI SnSe is strongly related to its surface structure. The Sn-terminated surface and Se-terminated surface have been previously shown to have distinctively different electronic properties. We show that our SnSe (111) thin films have a Sn-terminated surface without surface reconstruction. Furthermore, we reveal an oscillatory, contraction-expansion-contraction pattern for the structural relaxation in the top few layers of SnSe. We show that μLEED-*IV* is a powerful tool for study of atomic crystal structure of 2D materials. We believe the detailed surface structural information is of fundamental importance and provides crucial input for better understanding the intriguing electronic properties of various 2D materials and a more solid guidance for engineering 2D materials based devices.

References:

[1] Z. Dai, W. Jin, M. Grady, J. Sadowski, J. Dadap and R. Osgood et al., Surf. Sci. **660**, 16 (2017).
[2] Z. Dai, W. Jin, J. Yu, M. Grady, and J. Sadowski et al., Phys. Rev. Materials. **1**, 074003 (2017).
[3] W. Jin, S. Vishwanath, J. Liu, L. Kong, R. Lou and Z. Dai et al., Phys. Rev. X **7**, 041020 (2017)