Growth Kinetics and Atomistic Mechanisms of Native Oxidation of ZrS_xSe_{2-x} and MoS_2

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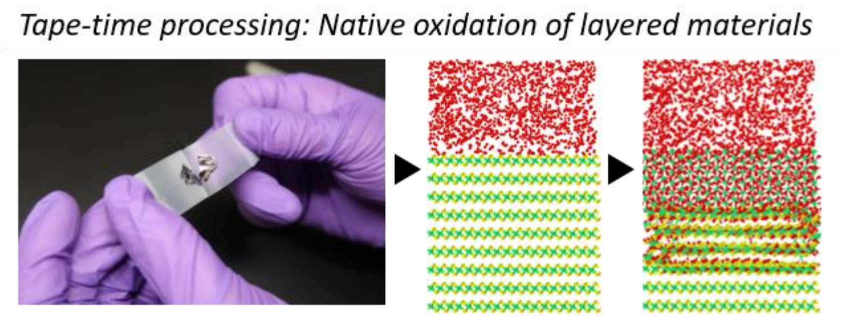
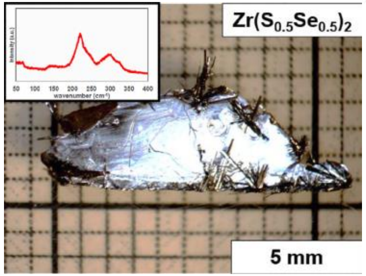
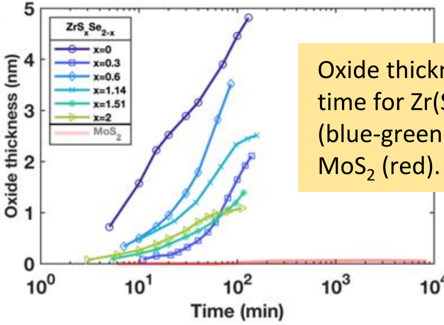
Project Summary: Semiconductor oxidation is essential to understand and control for device processing, as revealed by the history of silicon microelectronics. Much is known empirically about the oxidation of 2D materials, but quantitative and atomistic understanding is lacking and is critical to the development of these materials for practical applications. A close combination of experiment (cleaving and spectroscopic ellipsometry) and simulation (reactive molecular dynamics simulations) finds that $Zr(S,Se)_2$ single crystals oxidize quickly in air, at up to 0.5 Å/min, while MoS_2 single crystals are extremely air-stable, with no oxide formation for >1 year. A Zr-O bond switching mechanism was identified that enables fast oxygen transport and breaks down the layered crystal structure. Next steps will compare synthetic thin films to bulk crystals and extend to other TMDs. Published in *Nano Letters 20 (12), 2020, 8592-8599.*

2DCC Role: The research resulted from a collaboration between two external users of the 2DCC facility (both assistant professors) who met at the 2019 User Committee meeting. At the meeting, Prof. Patrick Vora from George Mason Univ. presented results on the optical properties of $Zr(S,Se)_2$ crystals synthesized in the 2DCC facility, which sparked interest from Prof. Rafael Jaramillo from MIT to study the oxidation kinetics. Graduate students from Prof. Vora's group also visited the 2DCC facility to learn how the samples were synthesized by chemical vapor transport and receive training in layer exfoliation.



Experiments reveal time-dependent oxide growth after tape exfoliation.
Simulations reveal atomic mechanisms that promote and suppress oxidation.





Oxide thickness vs. time for $Zr(S,Se)_2$ alloys (blue-green series) and MoS_2 (red).