In Situ Investigation of Doping of 2D Semiconductors During Atomic Layer Deposition of Dielectrics

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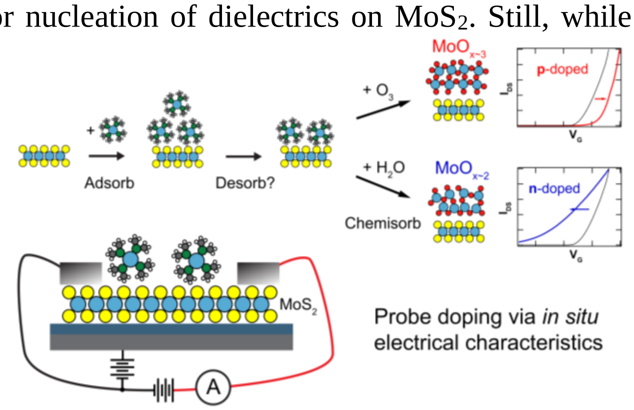
The sensitivity of ultrathin and 2-dimensional (2D) semiconductors to the surrounding environment provides a key opportunity for control of material and device behavior. Especially as substitutional dopants may be difficult to control and lead to increased scattering, adlayers are a promising approach to tuning the Fermi level in 2D semiconductors [1]. Despite the growing body of results using oxide dielectrics to this end, there is a lack of mechanistic investigation and understanding of scope and limitations.

In this talk, we build on results using atomic layer deposition of a tunable oxide to dope MoS_2 [2], and investigate mechanisms of growth and doping via *in situ* electrical measurements. Using a modified atomic layer deposition (ALD) reactor, we can measure field-effect transistors during dielectric growth at temperatures up to 300°C. As well as being more efficient than *ex situ* measurements for some studies (e.g. carrier concentration vs. thickness [3]), it enables otherwise-impossible observation of dynamics and changes with each half-cycle of deposition. We are therefore also positioned to learn about growth and reactivity. The first and to date only other such *in situ* electrical measurements identified that physisorption of ozone promotes dielectric growth on graphene [4]. We further observe reversible adsorption of metal-organics for nucleation of dielectrics on MoS_2. Still, while physisorptive nucleation of ALD is not unique to graphene, neither is it universal to van der Waals materials. Even moderately air-stable transition metal dichalcogenides such as $MoTe_2$ can differ notably in reactivity, growth mechanism, and thus semiconductor-dielectric interface. As such, *in situ* measurements are a powerful tool to understand growth on 2D materials.



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Supplementary Pages

We have demonstrated doping of MoS_2 using MoO_x with a range of stoichiometries, where both the magnitude and direction of carrier concentration change can be varied (Figure S1). We have also implemented convenient electrical measurement for a commercial atomic layer deposition reactor (Ultratech Savannah S200) and observe device characteristic changes *in situ* (Figure S2).

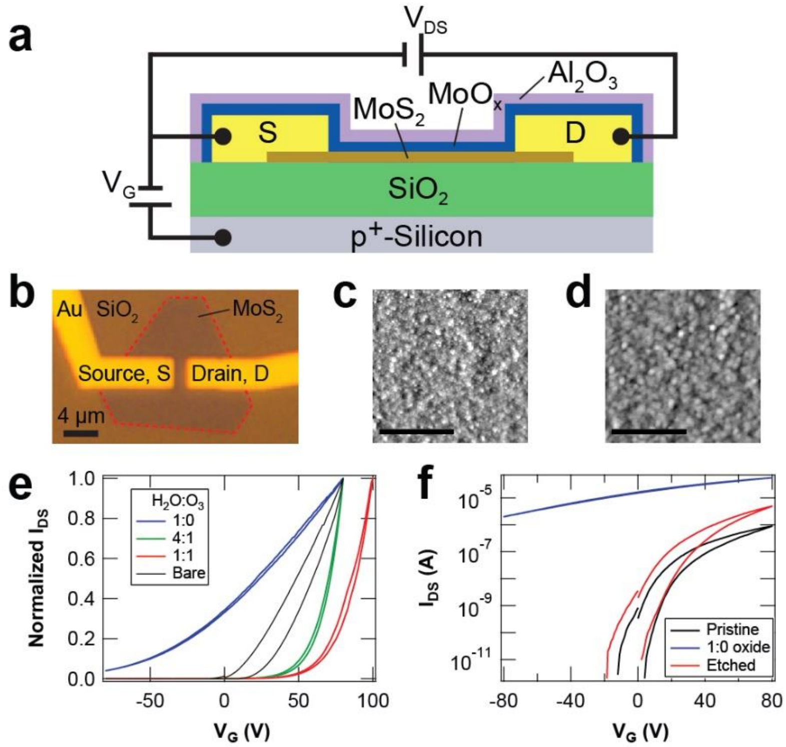

Figure S1: Continuous MoO_x layers as a dopant for MoS_2 (a) scheme showing blanket deposition on fabricated FETs to dope the channel (b) optical micrograph of representative device (c) AFM image of MoS_2 before and (d) after oxide deposition, showing that oxide is continuous and low roughness (e) normalized and (f) log-scale I_{DS} illustrating shift in threshold voltage (adapted from [2])

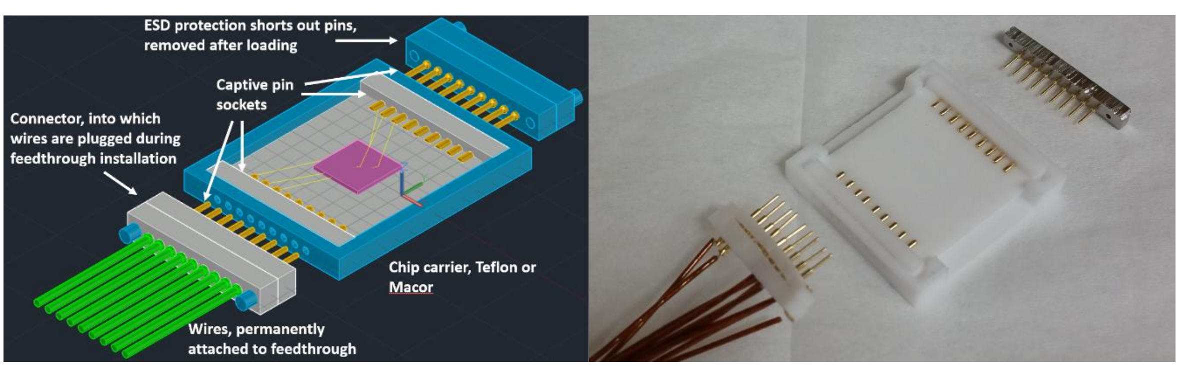

Figure S2: High temperature chip carrier (a) schematic and (b) photograph