Controlled Nucleation and Growth of 2D SnS2 via Solution Atomic Layer Deposition on Functionalized Surfaces

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Atomic layer deposition (ALD) is an established vapor-phase technique that utilizes sequential, self-limiting surface reactions to achieve sub-nanometer thickness control. By pulsing precursors individually, the process ensures the growth of high-quality, uniform 2D films [1]. Transitioning to solution-based ALD (sALD) offers significant advantages, including scalability, atmospheric pressure processing, and a broader precursor library, yet the fundamental precursor-surface interactions in solvents remain poorly understood [2].

As the initial cycles of sALD dictate final film quality and crystallinity, self-assembled monolayers (SAMs) are employed as templates to precisely control the density of nucleation sites. Previous *i n-situ* XRR investigations of SnS_2 growth on (3-mercaptopropyl)trimethoxysilane (MPTMS) SAMs revealed non-ideal island growth, linked to the distribution of thiol groups present at the SAM surface [3]. This necessitates optimization which might become possible by using phosphonic acid-based SAMs on AlO_x/Si as it offers a wider range of available SAM molecules and longer alkyl chains available for purchase to provide a more ordered template for controlled nucleation.

We investigate nucleation behaviour of 2D SnS_2 on thiol terminated SAMs on AlO_x/Si substrates after 10 sALD cycles. To control nucleation site density, we mixed 12-mercaptododecylphosphonic acid (SH-C12PA) with decylphosphonic acid (C10PA) spacers. By varying the SH-C12PA molar fractions at 100%, 5%, 1%, 0.2% and 0%, we systematically increased the average intermolecular distance of the functional –SH groups. The evolution of the SnS_2 films after 10 sALD cycles is characterized using X-ray reflectometry and atomic force microscopy. The primary objectives are to evaluate SAM homogeneity across different dilution systems and to tune the available SnS_2 nucleation sites on the functionalized surfaces. These insights into interfacial wetting behavior clarify how surface site density governs the transition between island and layer-by-layer growth, providing a pathway to defect-free 2D semiconductor fabrication.

Keywords:
X-ray reflectometry, SnS2, Solution atomic layer deposition, Self-assembled monolayers

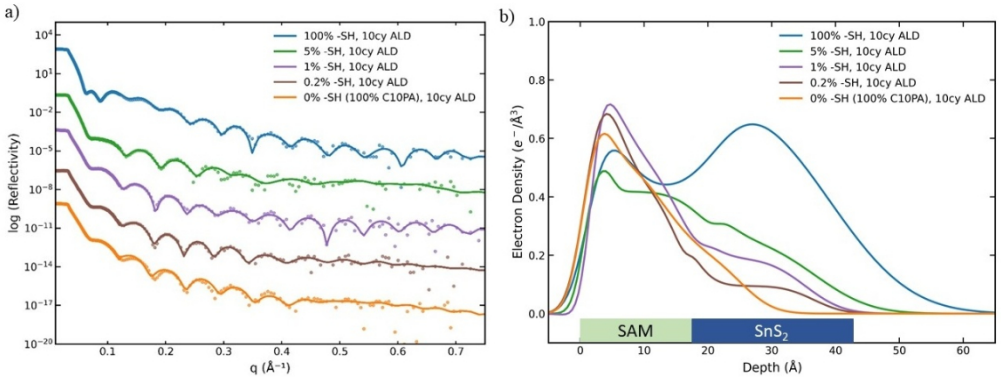

Figure 1: a) Ex-situ XRR data (dots) and fits (lines) of SnS_2 after 10 sALD cycles on AlOx/Si with varying thiol-densities SAM. b) Electron density profiles of SnS_2 and SAMs derived from a).

References:

[1] R. Johnson et al., Mater. Today, 17 (2014) 236.
[2] Y. Wu et al., Nano Lett., 15 (2015) 6379.
[3] K. Götz et al., Adv. Mater. Interfaces, 11 (2024) 2300990.

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