Plasma-enhanced atomic layer deposition of MoS_2: from 2-D monolayers to 3-D aligned nanofins

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Plasma-enhanced atomic layer deposition (PE-ALD) might prove to be a key enabler for tackling the current challenge of large-area growth of 2-D materials with wafer level uniformity and digital thickness controllability. In this contribution, we have implemented PE-ALD to synthesize large-area WS_2 and MoS_2 thin films with tuneable morphologies i.e. in-plane and vertically standing nano-scale architectures on CMOS compatible SiO_2/Si substrates. The large scale 2D in-plane morphology has potential applications in nanoelectronics, while the 3D nanofin structures could be ideal for catalysis applications such as water splitting.

The PE-ALD process was characterized over a wide temperature range between 150°C - 450°C by using a combination of a metal organic Mo or W precursor and a $H_2S + H_2 +$ Ar plasma as the co-reactant. The use of plasma species as reactants allowed for more freedom in processing conditions and for a wider range of material properties compared with the conventional thermally driven ALD. The number of layers in the resulting films could be controlled accurately down to a mono-layer just by tuning the number of ALD cycles. HAADF TEM analysis of the showed that during the initial ALD cycles, MoS_2 islands expeditiously extended in the lateral direction and merged to form a film which continued to grow in a layer-by-layer fashion until a certain thickness. Thereafter, an aligned out-of-plane growth mode started to dominate as shown by cross-sectional TEM analysis. The origin of this transition from in-plane to out-of-plane growth mode might be attributed to the enhanced precursor adsorption on high surface energy locations such as grain boundaries, kinks or ledges. Due to crowding effects at these favourable adsorption sites subsequent vertical growth of MoS_2 is observed.

We show that the in-plane grown 2D structures can be successfully applied in electronic devices, while the 3D aligned fins a very suitable catalysts for the Hydrogen Evolution Reaction. These results show that plasma enhanced ALD might be instrumental in realizing not only the large area growth of high-quality 2-D materials but can also be applied as a tool to control the morphology of thin films.

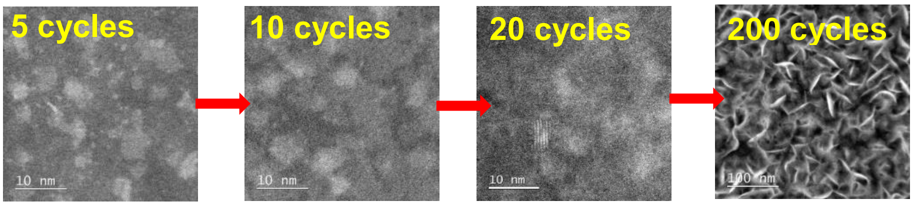

Figure 1. HAADF images showing the nucleation of MoS_2 on SiO_2/Si substrates as a function of number of ALD cycles and the variation of morphological structures.