Atomic layer deposition of Al_2O_3: new insights from sum-frequency generation

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Atomic layer deposition (ALD) is the method of choice to deposit thin films with sub-nanometer sensitivity and with an excellent step-coverage or conformality on 3D-structured materials with high-aspect ratio structures. In this presentation the surface chemistry during ALD of Al_2O_3 – by far the most used and studied ALD material – is revisited on the basis of broadband sum-frequency generation (BB-SFG) experiments. BB-SFG is a nonlinear optical technique that is excellently suited for *in-situ* studies of the surface chemistry governing ALD because of its inherent interface selectivity, submonolayer sensitivity, and short acquisition times. In contrast to BB-SFG, conventional absorption spectroscopy, based on differential measurements, monitors only changes on the surface. On the other hand, due to its surface selectivity, BB-SFG reveals information about both persistent and changing surface groups. Therefore, with this technique, open questions have been addressed to obtain a more complete picture of the ALD surface chemistry. Aspects that will be addressed in this presentation are:

- What are the reaction kinetics during the $Al(CH_3)_3$ and H_2O exposure steps?
- What are the sticking probabilities of $Al(CH_3)_3$ and H_2O during the ALD process
- What's the origin of the decrease in growth per cycle (GPC) of Al_2O_3 at low temperatures?
- What about so-called "cooperative" effects during ALD surface reactions?
- What about the initial growth during ALD on substrate materials?

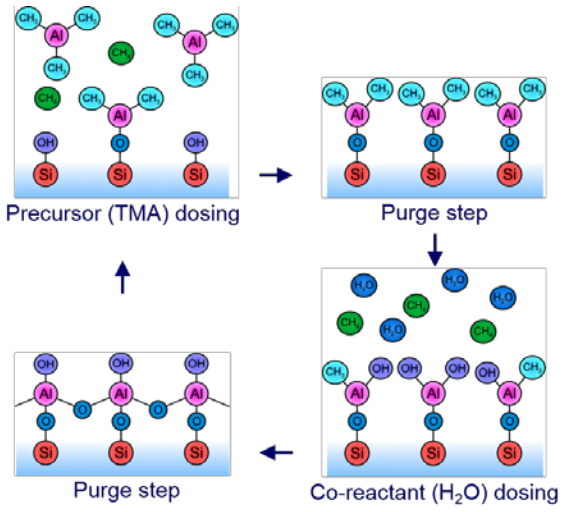

Fig. 1: Cycle for ALD of Al_2O_3 from $Al(CH_3)_3$ (trimethylaluminum, TMA) and H_2O. The precursor and co-reactant dosing steps are alternated by purge steps. Every cycle results in an increase of thickness (1 Å at 250 °C) such that the targeted film thickness can be reached by repeating the right number of cycles.

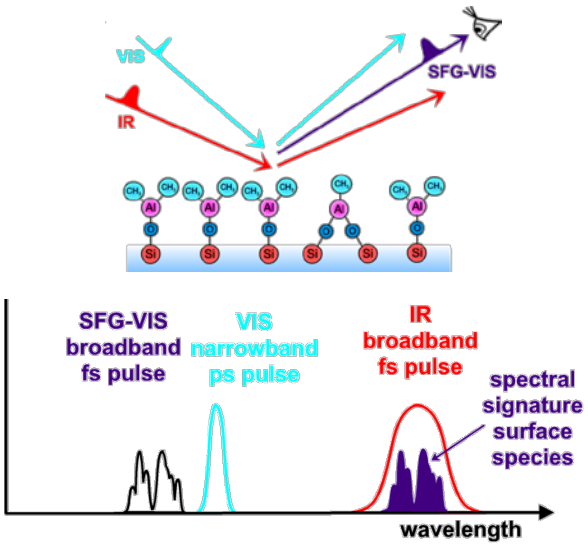

Fig. 2: Broadband sum-frequency generation (BB-SFG) on surfaces: (top) schematic illustration of the technique applied to an amorphous Al_2O_3 surface; (bottom) schematic showing that a wide spectral coverage in the IR can be obtained within one laser shot and with femtosecond time-resolution.