Supplemental Document

Beyond The Ligand Exchange Model
Time Resolved ALD Of HfO_2 On InAs Thermal Oxide

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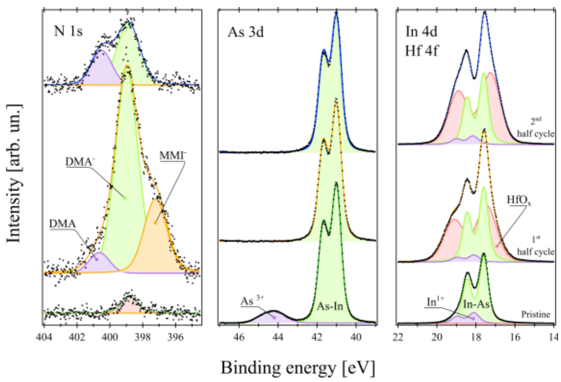
Ambient pressure X-ray photoemission spectroscopy (AP-XPS) papers:

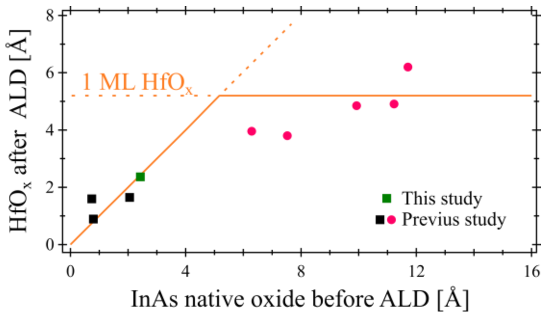
- S. Zhu, et al. - "*HIPPIE: a new platform for ambient-pressure X-ray photoelectron spectroscopy at the MAX IV Laboratory*"- J. Synchrotron Rad. (2021). 28, 624-636
- E. Kokkonen, et al. - *"Upgrade of the SPECIES beamline at the MAX IV Laboratory"* - J. Synchrotron Rad. (2021). 28, 588-601

Atomic Layer Deposition papers:

- G. D'Acunto, et al. - *"Atomic Layer Deposition of Hafnium Oxide on InAs: Insight from Time-Resolved in Situ Studies" - ACS Appl. Electron. Mater. (2020), 2, 12, 3915–3922*
- R. Timm, et al. - *"Self-cleaning and surface chemical reactions during hafnium dioxide atomic layer deposition on indium arsenide" - Nat Commun 9, 1412 (2018)*
- G. D'Acunto, et al. - *"Systematic study of atomic layer deposition of HfO2 on anatase TiO2: space state of Temperature and Pressure" – Manuscript in preparation*



N 1s, As 3d, and In 4d/Hf 4f core levels acquired before ALD (bottom), after the first (middle) and after the second (top) half cycle. The N 1s shows DMA- as well as MMI-; the Arsenic oxide is completely reduced after the metal pulse and it stays reduced even after severe oxidation condition (water dosing). Also the In oxide gets reduced but only partially, since it is needed to form the interface with the high-k dielectric. The Hf 4f shows the fingerprint of HfOx already from the first half cycle.



Correlation graph indicating the thickness of oxide in the InAs prior to the ALD deposition and the thickness of HfO_x after the first half cycle. The circle points indicate the samples with a native oxide thickness of more than a single monolayer(ML); the squared ones the samples with not enough oxygen to form a ML of HfO_x.