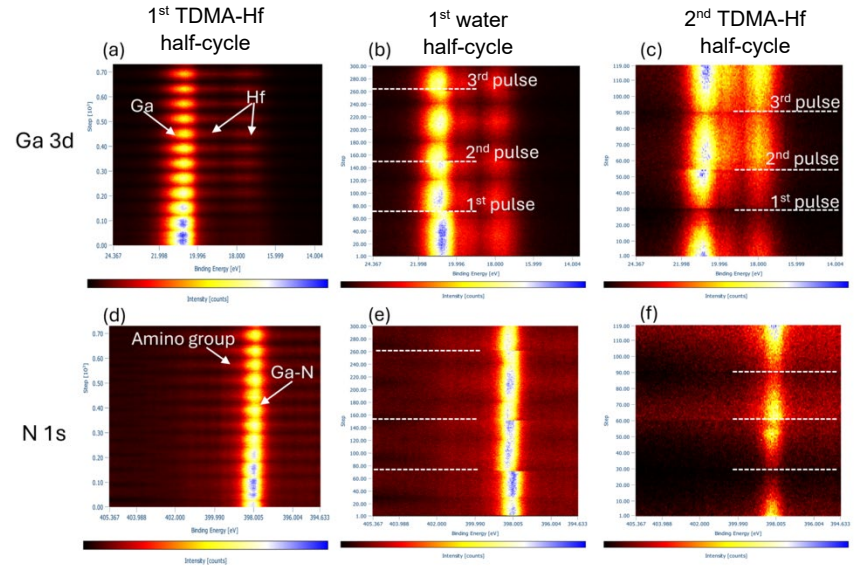


**Figure 1**: Time-resolved XPS data of $HfO_2$ ALD on GaN, with color-coded intensity from (a-c) Ga 3d and Hf 4f, which are overlapping in energy, and (d-f) N 1s core levels, obtained in snap-shot mode during (a,d) the 1st TDMA-Hf deposition, (b,e) the 1st water deposition, and (c,f) the 2nd TDMA-Hf deposition. Individual precursor pulses, using Ar as carrier gas, can induce shifts in binding energies. Periodic fluctuations of the intensity occur as the sample is continuously scanned through the X-ray beam, in order to avoid X-ray induced reactions, moving it slightly in and out of focus.

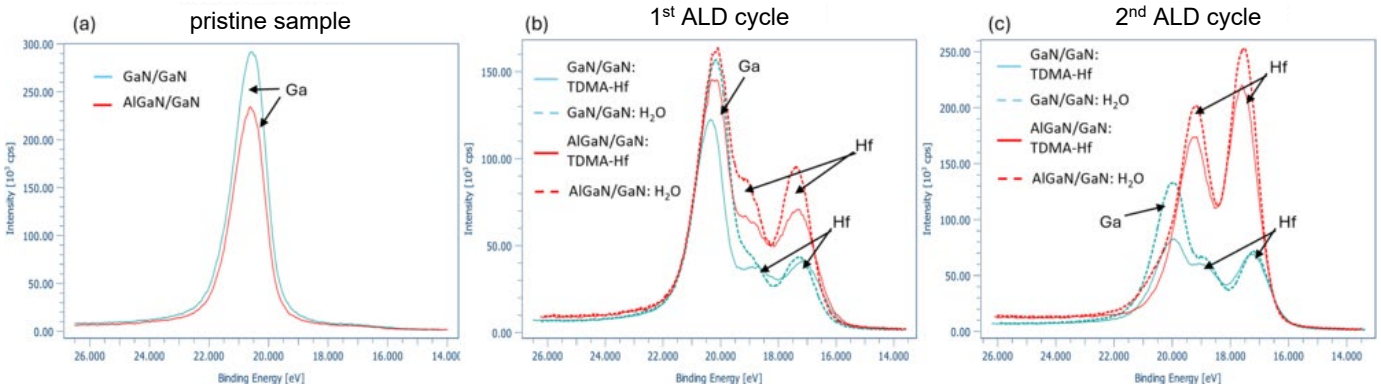


**Figure 2**: Ga 3d/Hf 4f XPS core-level spectra from a GaN and an $Al_{0.06}Ga_{0.94}N$ sample before (a) and after 1st (b) and 2nd (c) ALD cycle. Ga 3d and Hf 4f core-level peaks are partially overlapping. More Hf is found on the AlGaN sample. Note that in (c) the Ga peak has almost vanished for the AlGaN sample due to strong attenuation by the Hf-containing surface layer.

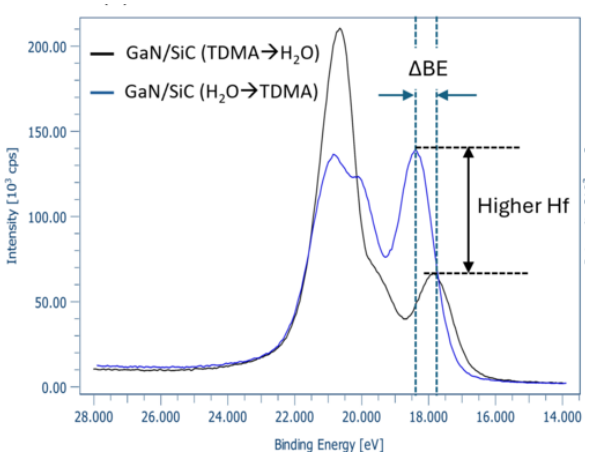


**Figure 3**: Ga 3d/Hf 4f XPS core-level spectra from a thick GaN film grown on SiC, obtained after one full $HfO_2$ ALD cycle, with either TDMA-Hf precursor first and water second (black curve) or vice versa (blue curve). In the latter case, we observe not only a much higher Hf intensity, but also a significant shift in the binding energy of the Hf peaks, indicating oxygen-deficient Hf-oxide.

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