Supplementary Material

Monolithic Integration of Perovskites on Ge(001) by Atomic Layer Deposition: A Case Study with $SrHf_xTi_{1-x}O_3$

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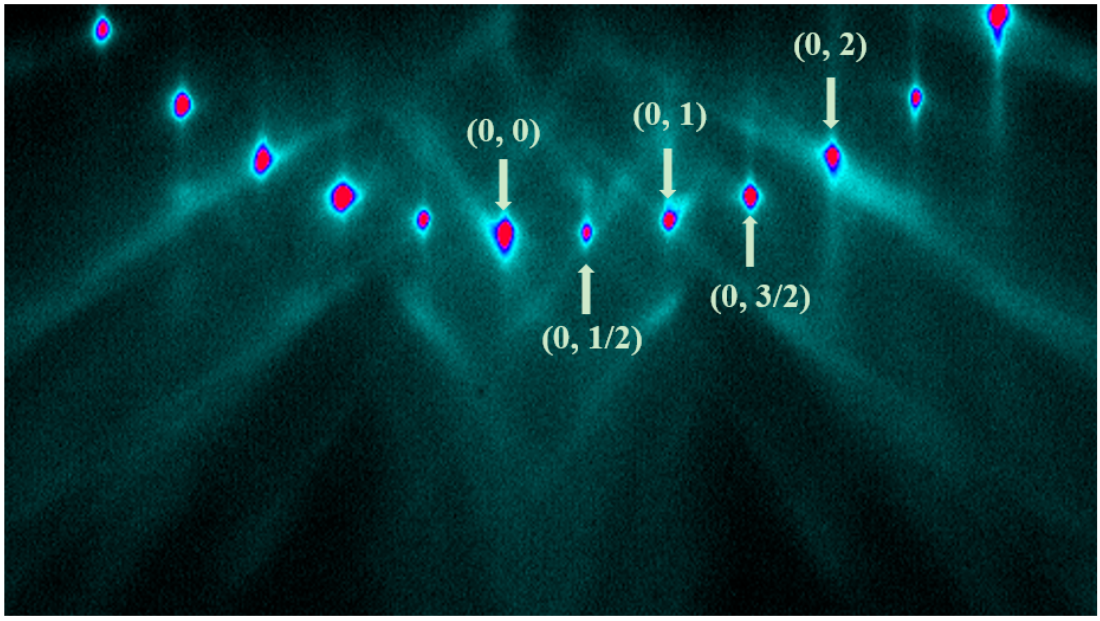

Figure S1. After 1 hr vacuum annealing at 650 °C, the Ge 2×1-reconstructed surface is observed by RHEED. The Kikuchi lines indicate the surface cleanliness and good long range order of the substrate, which are important for ALD growth on Ge (001). Image taken along the <110> direction.

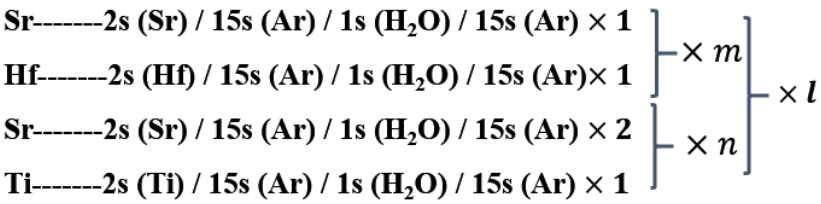

Figure S2. Schematic of the growth method for $SrHf_xTi_{1-x}O_3$ films by ALD. The subcycles *m* for SHO and *n* for STO are adjusted to realize different Hf content *x*. Different film thicknesses were realized by adjusting the number of supercycles *l*.

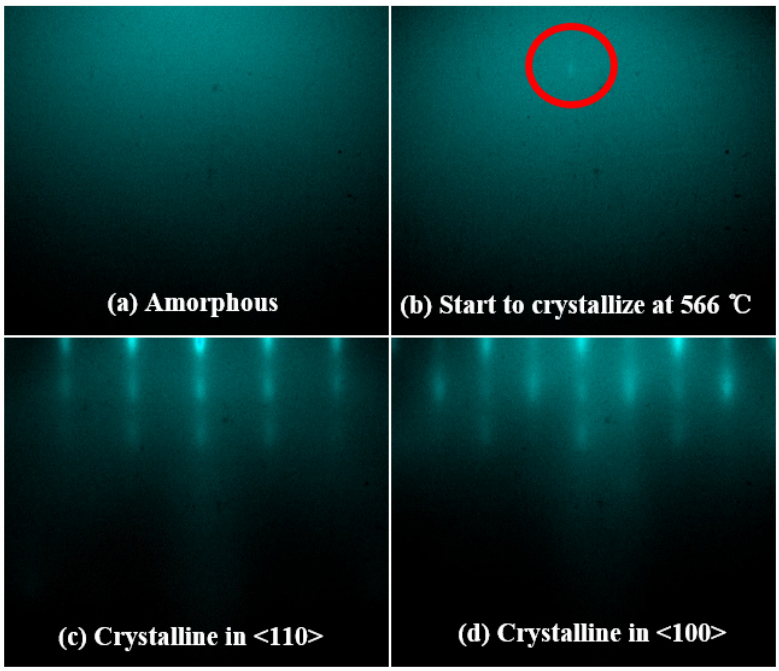

Figure S3. RHEED images for an 11.4-nm $SrHf_{0.34}Ti_{0.66}O_3$ film showing (a) the as-grown amorphous film, (b) the first pattern spot appearing at 568 °C, and (c) and (d) images were taken at 200 °C following annealing at 588 °C for 5 min. The beam was aligned along <110> and <100> directions for (c) and (d), respectively. This film was grown with a *m*:*n* = 1:1 subcycle ratio and *l* = 20 supercycles.

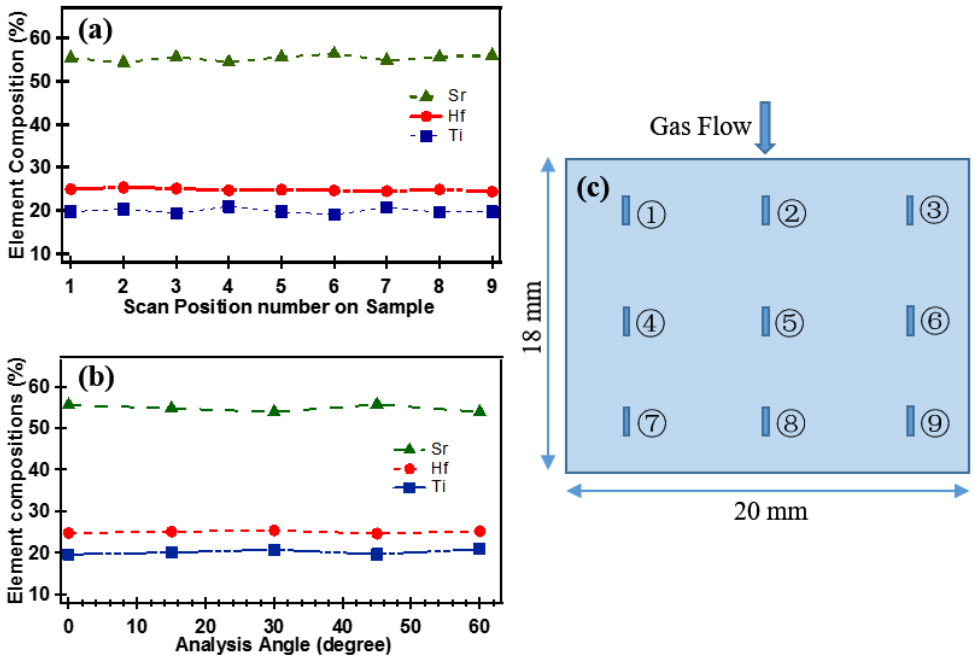

Figure S4. (a) XPS position analysis and (b) angle-resolved XPS at location Number 5 for the $SrHf_{0.56}Ti_{0.44}O_3$ film after annealing 654 °C. (c) Schematic showing the sampling points and orientation in the ALD chamber for the 18×20 mm^2 Ge substrate. The Numbers 1 to 9 denote different XPS sampling points; an area of 3×1 mm^2 is probed at each sample point. The Sr, Hf and Ti compositions are 55.3 ± 1.0%, 24.8 ± 0.5% and 20.0 ± 1.0%, respectively, for the nine positions. The analysis angles in (b) of 0, 15, 30, 45 and 60° correspond to sampling depths of 6.2, 6,0, 5.4, 4.4 and 3.1 nm, respectively. For all the AR-XPS scans, the Sr, Hf and Ti compositions have ranges of 54.8 ± 0.9%, 25.0 ± 0.6% and 20.3 ± 0.6%, respectively.

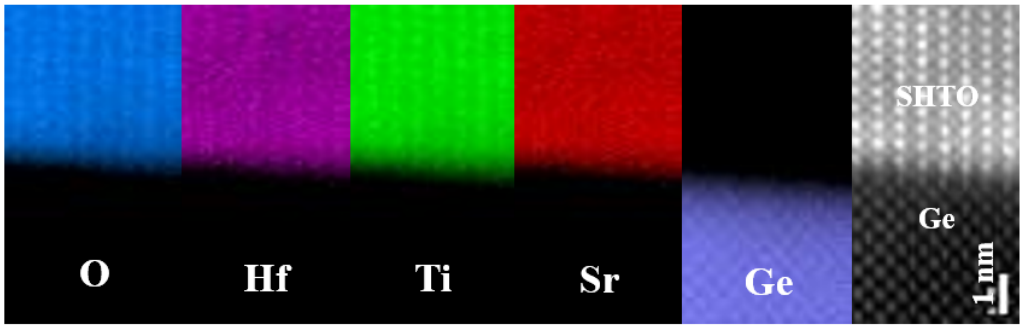

Figure S5. Electron-energy-loss spectroscopy (EELS) mapping to determine the elemental distribution in selected region for a 14.0-nm $SrHf_xTi_{1-x}O_3$ film with x = 0.55, post-deposition annealed at 645 °C for 5 min.