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Supporting Information

Influences of Oxygen Source and Substrate temperature on the Unusual Growth Mechanism of Atomic Layer Deposited Magnesium Oxide Using Bis(cyclopentadienyl)Magnesium Precursor

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Table S1. Results of XRR curve fitting for SiO_2 bottom layer and Al_2O_3 top layer

Oxygen source	cycle	T_s (°C)	XRR (SiO_2 layer)			XRR (Al_2O_3 layer)		
			Thickness (nm)	Density (g/cm^3)	Roughness (nm)	Thickness (nm)	Density (g/cm^3)	Roughness (nm)
H_2O (t_4 = 0 s)	78	250	1.4	2.506	0.551	-	-	-
H_2O (t_4 = 12 s)	78	250	2.2	2.818	0.606	-	-	-
H_2O (t_4 = 60 s)	78	250	2.5	2.611	0.551	-	-	-
O_3	140	230	0.2	2.000	0.441	10.6	3.417	0.430
O_3	49	290	0.1	1.960	0.796	10.3	3.569	0.623
O_3	58	335	0.5	1.749	0.739	10.9	3.349	0.913
O_3	72	390	1.5	2.369	0.739	10.9	3.467	0.974

Bulk density: SiO_2 = 2.650 g/cm^3; Al_2O_3 = 3.950 g/cm^3.

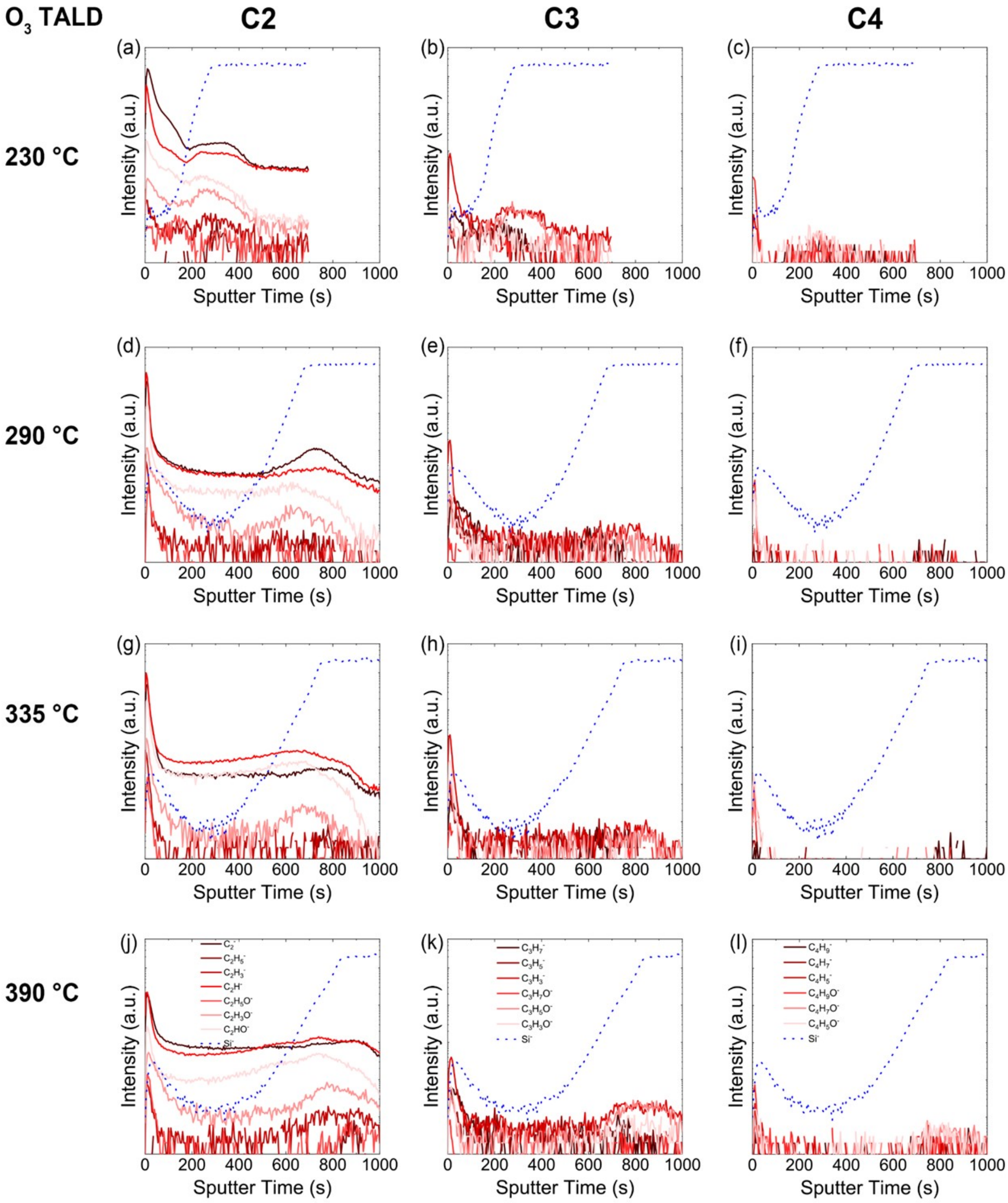

Fig. S1 Depth profiles measured by ToF-SIMS with substrate temperature dependence (a, d, g, j) x = 2. (b, e, h, k) x = 3 and (c, f, i, l) x = 4, where the x represents the carbon number in the ionized fragments. Blue dot lines represented Si^- fragment signals.

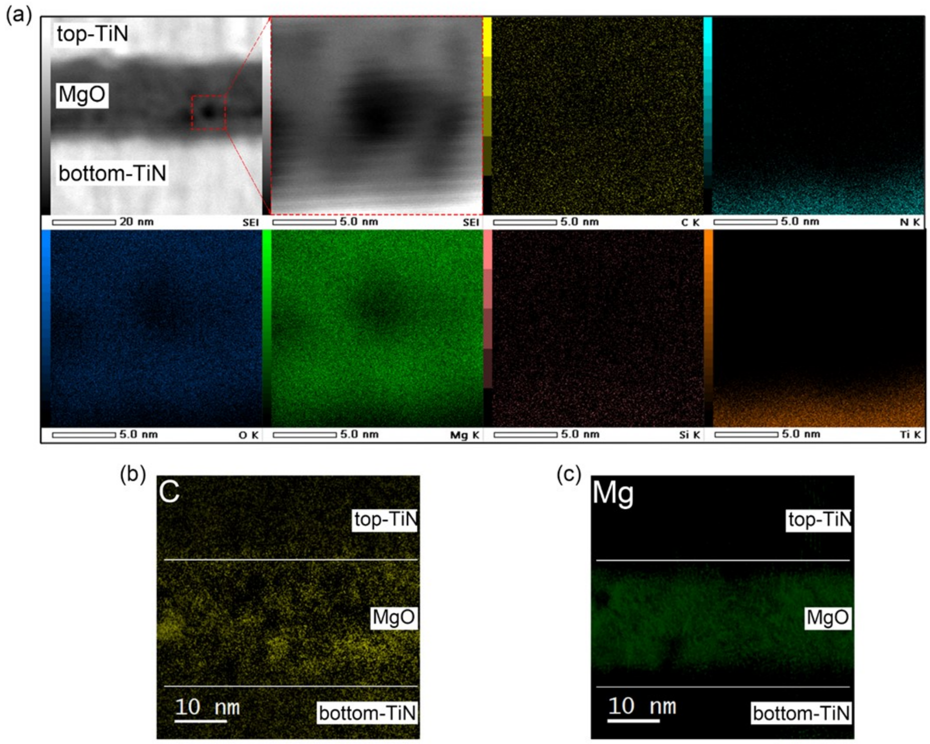

Fig. S2 (a) Secondary electron image (SEI) and elemental mapping by energy-dispersive X-ray spectroscopy (EDS) in scanning transmission electron microscopy (STEM). Elemental mapping of (b) carbon and (c) magnesium by Energy-Filtered TEM. (a) images were collected by FETEM (JEM-F200), while (b) and (c) images were collected by Tecnai (F20).

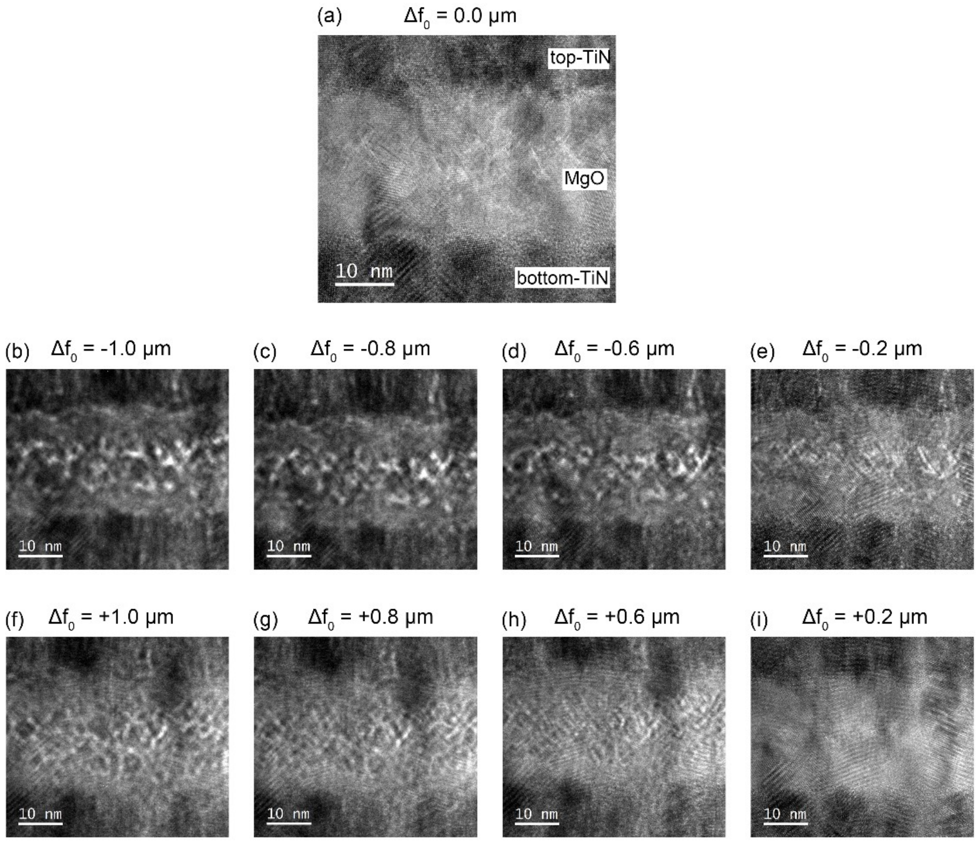

Fig. S3 (a) just-focus and (b) – (i) defocus TEM bright-field images with 360,000 magnification collected by EFTEM (Tecnai F20). The deposited MgO film thickness is around 25 nm, grown by O_3 ALD process at 290 °C.

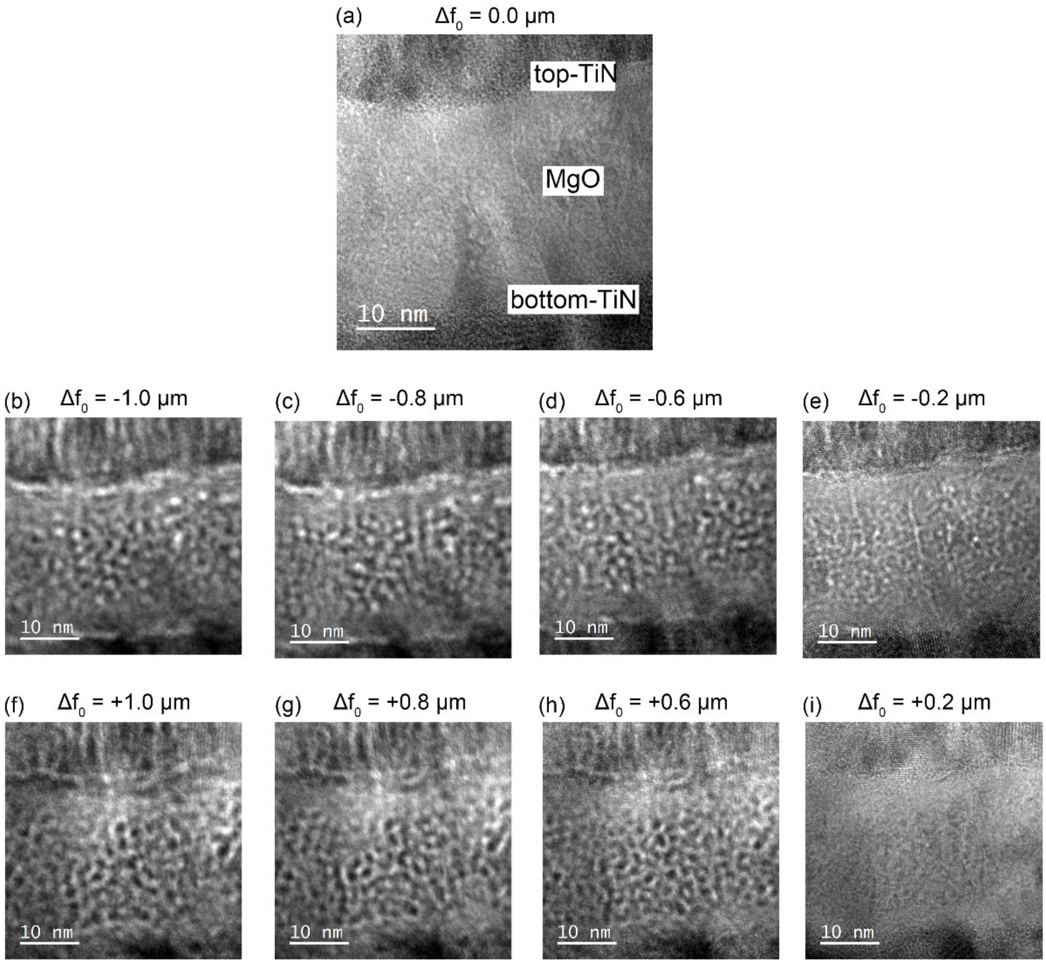

Fig. S4 (a) just-focus and (b) – (i) defocus TEM bright-field images with 360,000 magnification collected by EFTEM (Tecnai F20). The deposited MgO film thickness is around 25 nm, grown by O_3 ALD process at 390 °C.

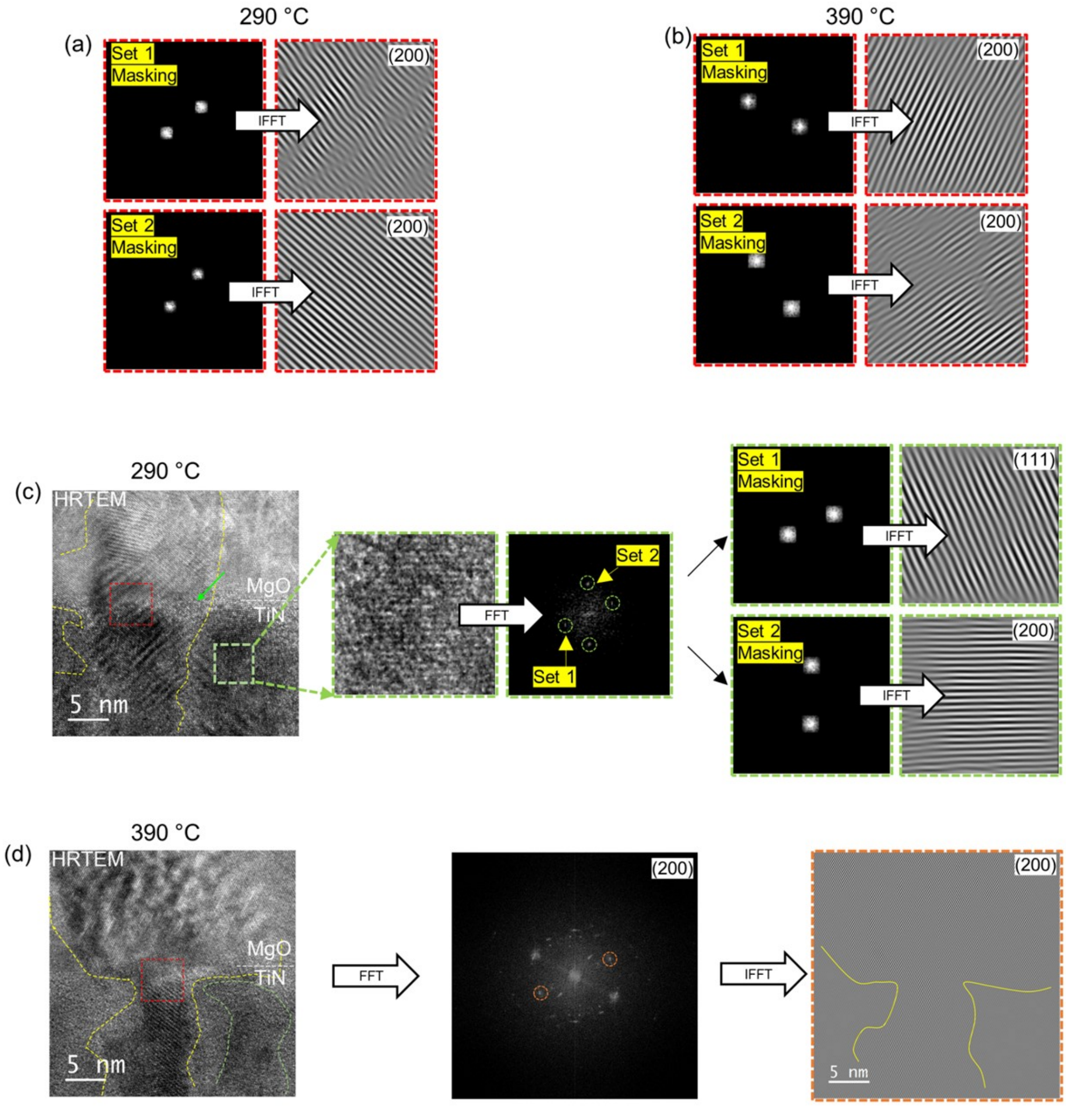

Fig. S5 (a – b) Fourier masking of FFT images in Figs. 11 (e) and (f) at MgO/TiN interfaces. MgO films were grown at T_s of (a) 290 °C and (b) 390 °C. (c) FFT and inverse FFT analysis for the TiN grain (in green square) without inducing epitaxial growth, at 290 °C. Non-preferable orientation (111) was identified. (d) FFT and inverse FFT analysis for the whole HRTEM image. Extensive (200) grain growth was confirmed. Due to the relatively low quality of HRTEM image of 390 °C sample, direct FFT and IFFT analysis for the TiN grain (with green line indicated grain boundaries) without inducing epitaxial growth was difficult. Therefore, Fourier masking was performed for the dominant orientation.

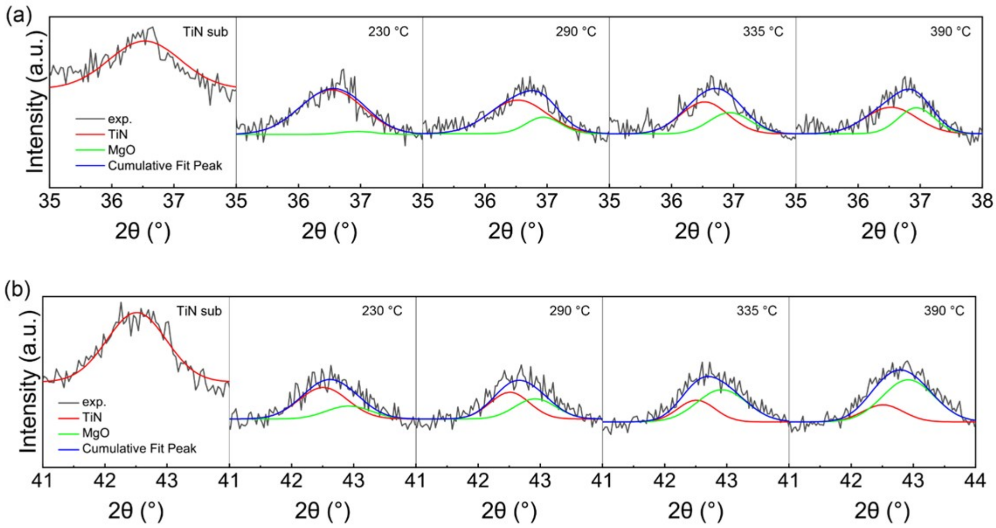

Fig. S6 GAXRD (ω = 0.5 ˚) of ~10 nm MgO thin films by O_3 ALD process as a function of T_s, deposited on TiN substrate. Deconvoluted peaks of (a) (111) and (b) (200), by Gaussian function fitting. The adopted sequence time is 6 – 30 – 6 – 30 s and the O_3 ALD cycle number is 140, 49, 58, and 72, respectively.

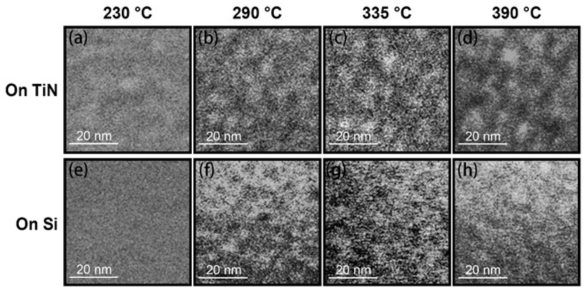

Fig. S7 FESEM plane-view of MgO deposited by O_3 ALD process with substrate temperature dependence (a) – (d) on TiN substrate. (e) – (h) on bare-Si substrate.