## **Supporting Information**



Fig. S1. The schematic of the HT-LCVD apparatus.



Fig. S2. A diagram showing the evolution of temperature in HfO<sub>2</sub> films grown using

## HT-LCVD.



Fig. S3. A schematic model for the growth of <002>-oriented grains in Region II showing (a) atomic nodes and (b) exposed surface.



Fig. S4. The schematics of FM, VW, and SK growth models.



Fig. S5. The dielectric measurements of HfO<sub>2</sub> films in different regions, as the (a) real part permittivity, (b) imaginary part permittivity, (c) dielectric loss (tanδ), and (d) variation chart of permittivity and dielectric loss.