Direct growth of orthorhombic Hf$_{0.5}$Zr$_{0.5}$O$_2$ thin films for hysteresis-free MoS$_2$ negative capacitance field-effect transistors

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Herein, the direct growth of polar orthorhombic phase in Hf$_{0.5}$Zr$_{0.5}$O$_2$ (HZO) thin films is reported using Pulsed Laser Deposition (PLD). The growth of HZO onto a preheated ($700\,^\circ\text{C}$) silicon substrate mimics the rapid thermal annealing, which allows the formation of smaller crystallites ($\sim9.7\,\text{nm}$) with large surface energy leading to the stabilization of metastable orthorhombic phase. Unlike atomic layer deposition (ALD) of HZO, PLD is more advantageous for depositing highly crystalline thin films through optimized parameters, such as laser fluence and background gas pressure. Further, the PLD-HZO is integrated with HfO$_2$ dielectric and the resulting gate stacks have been used in the bottom gate FET architecture -Si/PLD-HZO/HfO$_2$/MoS$_2$/Ti/Au’. The NCFETs have yielded a sub-thermionic subthreshold swing ($\text{SS}_{\text{for}} = 33.03 \pm 8.7\,\text{mV/dec.}$ and $\text{SS}_{\text{rev}} = 36.4 \pm 7.7\,\text{mV/dec.}$) and a negligible hysteresis ($\sim28\,\text{mV}$), which is capable in realizing low power integrated digital/analog circuits.

**INTRODUCTION**

Incorporation of a thin ferroelectric layer in the gate stack of Field Effect Transistors (FETs) is an innovative approach to achieve steep switching and hysteresis-free devices$^{1,2}$. Among the widely available options of ferroelectric materials, ranging from metal oxides to polymers, metal oxides have gained considerable technological importance owing to their compatibility with current complementary-metal-oxide-semiconductor (CMOS) technology as well as large-scale integration$^3$. A well-known, high dielectric permittivity (κ) hafnium oxide (HfO$_2$) can also act as a ferroelectric with proper modification in the crystal structure. In other words, amorphous HfO$_2$ is the most popular high κ dielectric; but, its crystalline counterpart with metastable-orthorhombic phase (o-phase, Pca$_2_1$) depicts large remnant polarization due to inherent ferroelectricity originated from the displacement of oxygen anions (i.e., non-centrosymmetry). The stabilization of o-phase is challenging due to the affinity of HfO$_2$ toward the thermodynamically stable, monoclinic phase (m-phase, P2$_1$/c)$^4$. However, numerous works have been established to crystallize the as-deposited thin films of HfO$_2$ into prominent o-phase, such as - substitutional doping (Zr, Al, La, and so on)$^4$-$6$, achieving finer grains with large surface energy$^7$, encapsulating with electrodes$^8$, depositing via distinct techniques such as Atomic Layer Deposition (ALD), Chemical Solution Deposition (CSD) and designing specific processing conditions such as annealing temperature, time, and heating/cooling rates$^9$. Notably, high heating rates are often required to restrict grain growth, which leads to the formation of finer grains with a large grain boundary area. Besides, low-temperature ($<250\,^\circ\text{C}$) growth using ALD is the most popular and widely reported deposition method, which demands an additional crystallization process. The low-temperature deposition through precursors retains the carbonaceous impurities at the junctions of as-deposited granular domains$^{10}$, which significantly hampers the growth when subjected to post-processing, such as rapid thermal annealing. Given this analogy, the post-processing-crystallization step is unavoidable as far as ALD and/or CSD techniques are concerned. Thus, an additional processing step is always necessary to achieve outstanding ferroelectric properties. In this regard, the present work demonstrates the possibility of direct growth of o-phase in Zr doped HfO$_2$ (Hf$_{0.5}$Zr$_{0.5}$O$_2$; HZO) on Si substrate, with finer crystallites using PLD technique. Of course, a lot of research on the growth of HZO on various substrates with the PLD process has been conducted, confirming the case with excellent ferroelectric phase$^{11-13}$. In this study, prominent o-phase was obtained by growing directly on the Si substrate for ferroelectric response. Substrate selection was also intended to show that it can be used as a useful device in an industry that is currently focused on silicon processing, and a two-dimensional (2D) molybdenum disulfide (MoS$_2$) NCFETs was investigated based on the grown HZO thin films. Despite tunable bandgap and colossal theoretical high carrier mobility (at room temperature) MoS$_2$ based FETs suffer from various instabilities, such as large hysteresis, threshold voltage, and high subthreshold swing (SS). The origin of instabilities is attributed to the prodigious surface area of MoS$_2$, which allows the atmospheric gas molecules to chemically adsorb on to the surface, resulting in hindered transport of charge carriers. These instabilities are generally observed regardless of the device architecture and channel layer thickness. Furthermore, high SS magnitudes demand increased power inputs, thereby promoting the heating effect. Thus, the problem of power dissipation becomes more severe in the case of large-scale integration of billions of FETs. The fundamental thermionic limit restricts the magnitude of SS to 60 mV/dec at room temperature$^{14,15}$. Among

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the various conceptual alternatives, in overcoming and/or nearing the thermionic limit, negative capacitance field-effect transistors (NCFETs) uses the amplification of channel surface potential more than the applied gate voltage. The negative capacitance (NC) effect transpires when the factor \( \frac{dP}{dE} \) is negative, where \( E \) is the electric field, and \( P \) is the ferroelectric polarization charge. This effect prevails under the switching of ferroelectric polarization\(^{14-16}\). In this contribution, a multi-stacked gate insulator based on the ferroelectric-HZO grown using PLD and ALD grown HfO\(_2\) afford MoS\(_2\) NCFETs to be operated at a low input power and negligible hysteresis\(^{17}\). The NCFETs operated below the thermionic limit dictated by Boltzmann’s Tyranny. The minimum SS obtained in both forward and reverse conditions are 33.03 ± 8.7 mV/dec and 36.42 ± 7.7 mV/dec, respectively.

RESULTS
Direct growth and characterization of Hf\(_{0.5}\)Zr\(_{0.5}\)O\(_2\) on Si
Most previous studies (~85%) have focused on ALD-grown HZO, which is advantageous in achieving large-area and uniform thin films. However, it suffers not only from the difficulty of controlling process time but also utilization of toxic and costly volatile chemical precursors makes the technique capital intensive. Unlike ALD, PLD can control the growth rate and chemical composition of thin films through laser fluence, ambient gas pressure. Besides, by controlling the substrate temperature and cooling rate, thin films can be grown with high crystallinity\(^{18}\). PLD uses ceramic targets as a source of material to be deposited, allowing easy transfer of the target composition and making the process chemical-free and non-toxic. A schematic of the PLD technique for the deposition of HZO thin films is presented in Fig. 1. Pulses of laser irradiation onto the HZO target allow the disintegration of the material from it and gets captured on to the pre-heated (700 °C) silicon substrate. In the case of ALD deposited thin films, the retained carbonaceous bonds are utilized to restrict grain growth during the post crystallization treatment\(^5\). This two-step process (growth followed by crystallization) has widely been reported and generalized to achieve metastable phases in HfO\(_2\) based ferroelectrics (strategy I, Fig. 1). On the contrary, PLD grown HZO does not have growth retarders due to pure ceramic targets. Nevertheless, the sudden striking of HZO, which has disintegrated from the target surface, is subject to abrupt heating at the preheated silicon substrate. The HZO is allowed to crystallize on a silicon substrate, whose temperature is maintained to be 700 °C (strategy II, Fig. 1). Thus, in PLD grown HZO, an additional crystallization processing step stands inapplicable. In other words, the PLD technique amalgamates both thin film deposition as well as crystallization steps to grow crystalline HZO thin films. To understand the effect of working \( P_O \) the HZO thin films were grown at three different working \( P_O = 0.1, 1, \) and 100 mTorr with fixed cooling \( P_O = 20 \) mTorr. Figure 2a depicts the X-ray diffraction patterns of HZO thin films grown.

The Bragg diffraction signatures at 28.3° and 34° correspond to m-phase and m-phase/tetragonal (t)-phase, respectively, while the diffraction peak centered at ~30.2° is attributed to polar o-phase. However, the slight shift in the Bragg angle from the reference, ~30.5°, suggests that the HZO films are strained in the compressive in-plane state\(^{11-13}\). Although the present thin films possess a mixed phase of m- and o- which is consistent with previous reports. Table 1 presents the status of present PLD grown
HZO on Si substrate in comparison with previously reported studies. Most of the attempts are used to grow HZO via ALD, requiring an additional crystallization step. Furthermore, the crystallite size estimated from the Scherrer equation (Supplementary Table 1) for all working $P_{O_2}$ conditions and it is found to decay with working $P_{O_2}$. Furthermore, the minimum of which is estimated to be $\sim 9.7$ nm. Consequently, the orthorhombic phase fraction estimated from the relative intensities of the diffraction responses (Eq. (1))\(^{19}\) is found to increase in thin films with an increase in the working $P_{O_2}$.

$$\text{Phase fraction} = \left[ \frac{l_0}{l_0 + l_m} \right] \times 100$$

(1)

where, $l_0$ and $l_m$ are the relative intensities of diffraction responses of o- and m-phases, respectively. Although in the 0.1 mTorr and 100 mTorr cases, the intensities are not so prominent to estimate the relative fractions of the phases, the usual trend of diminishing m-phase concerning the 1 mTorr case is worth noting. Furthermore, these variations convey that the smaller grains contribute to large surface energy, being one of the requirements to stabilize the o-phase. The grain sizes possess a strong dependency on the thickness of the film. Thicker films generally promote grain growth due to the large volume of material in all directions. However, the thin films impose geometrical constraints to grain growth. The PLD grown HZO thin films (with constant thickness) have depicted minimum grain/crystallite sizes as a direct consequence of working $P_{O_2}$, which is of its first kind, ever reported in Fig. 2b. Apart from the smaller crystallites, it is essential to have pinhole-free thin films. The HZO thin film (working $P_{O_2} = 1$ mTorr) is presented as an example in Fig. 2c, where both the density and thickness of the film are estimated from X-ray reflectivity measurements, found to be 7.2 g/cc, 12 nm, respectively.

Furthermore, from a cross-sectional TEM micrograph, it is clear that the thin film possesses a thickness of $\sim$12 nm (Fig. 2d). Thus, an ultrathin and dense film with finer grains is the ingredient for

Fig. 2  Structural analysis of PLD-grown HZO on Si. a XRD diffractograms of HZO thin films deposited at different working $P_{O_2}$. (b) variation of crystallite size and the fraction of o-phase estimated from Scherrer equation and relative peak intensities. c XRR results for an as-deposited (working $P_{O_2}$ = 1 mTorr) HZO thin film on Si. (d) TEM cross-sectional micrograph and EDS elemental mapping of HZO thin film; (e) Cross-sectional HR-TEM micrograph, filtered inverse FFT, and FFT pattern of the selected area.

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The shape of the P-V curve conveys improved ferroelectric electrodes, within a voltage range of ±5 V at a frequency of 10 kHz. In-line with the diffraction studies, depicting significant Fig. 2a. The cross-sectional TEM micrographs reveal the polycrystalline nature of the HZO film (deposited with a working \( P_0 \), of 1 mTorr). In Fig. 2e, the cross-sectional TEM micrographs and inverse FFT patterns reveal the planar d-spacing of two distinct phases (m-phase ~2.6 Å and o-phase ~3.0 Å)\(^{11}\), which are in-line with the diffraction studies, depicting significant peaks of both the phases (Fig. 2a).

The surface chemical compositions of the PLD-HZO grown with three distinct working \( P_0 \), = 0.1, 1, and 100 mTorr are analyzed using XPS, and the spectra are calibrated using carbon 1 s (C 1 s) peak positioned at 284.6 eV. The details of peak fitting and calibration using C 1 s are described in Supplementary Fig. 1 and Supplementary Tables 2–4. Furthermore, the presence of Hf and Zr at an expected ratio of 1:1 is confirmed, along with the oxygen from low- and high-resolution XPS spectra (Fig. 3a–b). Furthermore, the high-resolution O 1 s is deconvoluted into three synthetic signatures centered at 530.3 ± 0.1 eV, 531.7 ± 0.1 eV, and 532.6 ± 0.1 eV related to lattice oxygen \((O^2-)\), oxygen vacancy \((V_O)\) and surface adsorbed oxygen \((O-ads)\) species, respectively (Fig. 3c)\(^{20}\). From the ratio of relative area fractions of the principal components; it is confirmed that the ratio of oxygen vacancy to the lattice oxygen does not significantly depend on the selected range of working \( P_0 \) (Supplementary Table 5). It is essential to understand the defect chemistry of ferroelectric-HZO, especially oxygen vacancies whose diffusion gives rise to leakage current in ferroelectric capacitors\(^{21}\).

### Ferroelectric properties of the Hf\(_{0.3}\)Zr\(_{0.7}\)O\(_2\)

Furthermore, the HZO thin film deposited with a working \( P_0 \), of 1 mTorr is probed to understand the ferroelectricity. Along with the previously analyzed 12 nm thick HZO, two more thin film samples with the thickness of 4 and 7 nm were deposited and whose film characteristics such as density and roughness were quantified through the x-ray reflectivity measurements (Supplementary Fig. 2). Figure 4a shows the polarization vs. voltage (P vs. V) response of HZO thin films, sandwiched between Si and TiN electrodes, within a voltage range of ±5 V at a frequency of 10 kHz. The shape of the P-V curve conveys improved ferroelectric behavior compared to the previous report\(^{22}\). The reason behind the unsaturated P–V curve is attributable to the charge tunneling and lossy dielectric effect in the thin film\(^{23–25}\). In Fig. 4b, the coercive voltage \((V_c)\) and remnant polarization \((P_r)\) values as a function of varying thickness (4, 7 and 12 nm) magnitudes are presented. The \( V_c \) and \( P_r \) tend to show an upward trend with the HZO thickness. Notably, the asymmetric ferroelectric response in forward and reverse bias conditions are presented in Fig. 4b, which follows an inverse trend with the HZO thickness. The increasing thickness promotes a more symmetric P-V response.

### Electronic properties of the MoS\(_2\) NCFETs

Figure 5a shows the architecture of HZO-based MoS\(_2\) NCFETs. In order to successfully utilize ferroelectricity of HZO in the devices, two essential factors are needed to be considered. First, capacitance matching, which is fundamentally responsible in achieving stable NCFET operation. Since the metastable NC effect of ferroelectrics does not guarantee the stability of NCFET operation, a stable NC state can be achieved when a dielectric is connected in series with the ferroelectric layer and the stacked combination of those is used as a passive layer in place of a dielectric, in the regular device architecture. Matching the capacitance magnitudes results in the hysteresis-free operation of the devices, which is fundamentally distinct from the ferroelectric-FETs\(^{33,34}\). The relation between capacitance and SS is expressed as: \( SS = \left[ 1 + \frac{C_{semi}}{C_{ox}} \right] \times 60 \text{ mV/dec} \), where C is the capacitance with subscripts, semi, Fe, and ox corresponds to semiconductor, ferroelectric, and oxide dielectric, respectively\(^{35}\). Given this, the capacitance of oxide dielectric must be greater than the ferroelectric to realize sub-60 mV/dec. switching. The phenomena is caused by the amplification of the surface potential \((\psi_s)\) at the semiconductor-dielectric interface. As shown

### Table 1. Comparison of previous reports of HZO thin films on Si substrate with the present study.

| Deposition method | Post-deposition treatment | Characterization method | Phases | Reference |
|-------------------|---------------------------|-------------------------|--------|-----------|
| ALD RTA           | GIXRD                     |                         | ✓ ✓ ✓  | \(^{1}\)  |
| ALD RTA           | --                        | GIXRD                   | ✓ ✓ ✓  | \(^{29}\) |
| ALD RTA           | GIXRD                     |                         | ✓ ✓ ✓  | \(^{36}\) |
| ALD RTA           | GIXRD                     |                         | ✓ ✓ ✓  | \(^{37}\) |
| ALD RTA           | --                        | GIXRD                   | ✓ ✓ ✓  | \(^{39}\) |
| PLD (Ar gas)      | --                        | XRD                     | ✓ ✓ ✓  | \(^{22}\) |
| PLD (O\(_2\) gas) | --                        | XRD                     | ✓ ✓ ✓  | Present work |

\( RTA \) rapid thermal annealing, \( XRD \) X-ray diffraction, \( GIXRD \) grazing incidence X-ray diffraction, m monoclinic, o orthorhombic, t tetragonal.
Fig. 3  Surface chemical composition of PLD-HZO thin films by XPS. a Survey spectra of HZO thin films grown with different working \( P_{O_2} (= 0.1, 1, \text{ and } 100 \text{ mTorr}) \). b high-resolution spectra of Hf 4f and Zr 3d for three cases of working \( P_{O_2} \). c high-resolution O 1s spectra for three cases of working \( P_{O_2} \).

Fig. 4  Ferroelectricity of PLD-grown HZO on Si. Dependency of (a) P–V (at 10 kHz) curves (b) magnitudes of \(|V_C|, |P_r| \) and curve-asymmetry on the thickness of HZO (4 nm, 7 nm, and 12 nm) (c) PFM hysteresis loop averaged from 25 measurements (d) Histograms of PFM phase, before and after poling, over the same area (e) PFM of HZO thin films after poling with applied tip voltages of ±5 V.
**Fig. 5** Fabrication and electrical properties of MoS₂ NCFET. a Schematic of fabricated MoS₂ NCFET based on HZO with HfO₂ (9 nm). b Equivalent capacitance schematic of the MoS₂ 2D-NCFET (Vg,NC, ψψ represent the negative capacitance gate voltage and surface potential, C_DOS, C_emi, Cavr, and Cr, represent the capacitance of MOSFET, MoS₂, HfO₂, and HZO, respectively.) c Transfer characteristic curve (I_d–V_g) and SS versus I_d characteristics of the MoS₂ NCFET at V_d = 0.1 V.

In Fig. 5b, it is briefly expressed in the equivalent capacitance schematic of the MoS₂ NCET. Second, the thickness of MoS₂, qualitatively thin film of MoS₂ is a suitable candidate semiconductor material for highly efficient gate modulation in low-power electronic devices. The optimized few layers of MoS₂ (~7 nm, Supplementary Fig. 4) is selected as the channel and transferred onto the HZO/HfO₂ surface via mechanical exfoliation. Use of thin MoS₂ is capable of minimizing the adverse charge trap effect leading to hysteric behavior. The channel length of the fabricated devices is 15 μm and the width is 28 μm. Figure 5c shows I_d–V_g characteristics of MoS₂ NCFETs with a step size (ΔV_g) of 0.01.

The maximum gate voltage range is maintained to be −1 to 1 V at a fixed V_d (≥0.1 V). From the transfer curve of the NCFET, a steep-switching behavior can be visualized, which is considerably above the thermionic limit of 60 mV/dec, in both forward and reverse bias conditions. The SS and SSrev of NCFETs are found to be 29.22 mV/dec. and 35.31 mV/dec., respectively, with negligible hysteresis of 28 mV (measured at a fixed I_d of 1 nA). Statistically, the results measured on multiple devices is averaged to be 33.03 ± 8.7 mV/dec. and 36.4 ± 7.7 mV/dec., for SS and SSrev, respectively. The significant reduction in SS magnitude is attributable to the NC effect offered by HZO thin films. The amplification of voltage allowed the devices to work well within the physical limit set by Boltzmann's tyranny. Furthermore, the performance of the present NCFETs is compared with the previously reported data with various other ferroelectrics, such as CuxPbSrTiO₃ (PVDF-TrFE), Al₂HfO₂, and HZO (Supplementary Table 6). The fabrication and electrical properties of MoS₂ NCFETs are comprehensively assessed by both P–V and PFM measurements.

**DISCUSSION**

In this study, a ferroelectric HZO thin film is successfully grown at 700 °C on a silicon substrate using the PLD without any additional crystallization treatments. An optimum working P_d of 1 mTorr allows HZO to crystallize in the prominent ferroelectric-α-phase, which is confirmed by both XRD and HR-TEM analysis. The thin film with a maximum α-phase of 64% and a minimum crystallite size of ~9.7 nm has been achieved. Besides, the ~12 nm film possesses uniform distribution of elements throughout the thickness. Furthermore, the evaluation of HZO ferroelectricity is comprehensively assessed by both P–V and PFM measurements. The PLD-grown HZO in the gate stack with HfO₂ is the key to overcome generic issues such as large hysteresis, threshold voltage instabilities, and large SS of MoS₂-based FETs. The introduction of the PLD grown ferroelectric HZO proved that the steep-switching device enables low-power operation at SS = 33.03 ± 8.7 mV/dec. and SSrev = 36.4 ± 7.7 mV/dec. with free-hysteresis. Thus, the present work suggests a way to achieve low operating voltage of NCFETs with smaller SS and negligible hysteresis. Also, the additional crystallization treatment may not always necessary to achieve ferroelectric α-phase in HZO. The present work is probably the report which demonstrates the feasibility of PLD grown ferroelectric HZO, without additional crystallization treatment, to be used to develop NCFETs, which can further be used in energy-efficient logic circuits.

**METHODS**

**HZO growth on Si**

PLD (KrF excimer laser, λ = 248 nm) deposition was used to grow HZO on a heavily doped <100>-type silicon (Si) wafer (resistivity ≤ 0.005 Ω·cm). Initially, the Si substrate was cleaned by dipping it in buffered oxide etchant (6:1), which removes the native oxide layer. The cleaned Si substrate was then loaded into the PLD chamber in a high vacuum of 1.5 × 10⁻⁶ Torr. The substrate is allowed to reach 700 °C; meanwhile, laser fluence of 3 J/cm² along with the repetition rate of the laser pulse of 5 Hz was maintained. The deposition was carried out with a varied working oxygen partial pressure (P_d) of 0.1, 1, and 100 mTorr, while the oxygen pressure was kept 20 Torr during cooling.

**Thin film characterization**

The thin film structure was determined by X-ray diffraction (XRD: D8 DISCOVER) at the MEMS-Sensor Platform Center of SungKyunKwan University (SKKU). The high-resolution transmission electron microscopy (TEM: JEM-ARM 200 F, JEOL), coupled with energy-dispersive spectroscopy (EDS: Quantax-400, Bruker), was utilized at an accelerating voltage of 300 kV to estimate the thickness and elemental distribution. The chemical
composition was estimated by X-ray photoelectron spectroscopy (ESCA 2000 (MultiLab 2000)). The obtained XPS spectra were calibrated using the C 1s peak positioned at 284.7 eV. Furthermore, the characteristics such as film density, roughness and thickness were estimated using X-ray reflectivity (XRR: Rigaku, smart lab). Planar TiN/HZO/Si capacitors were measured using a ferroelectric tester (Precision LC II, Radiant technologies). The PFM measurements were conducted using an atomic force microscope (AFM: Park Systems, NX-10) equipped with a lock-in amplifier (Stanford Research Systems, SR844), a function generator, and data acquisition systems (National Instruments, NI-PXIe 5122/5412). PFM image measurements were performed with 0.7 Vp-p and 17 kHz. The PFM hysteresis loop was measured by applying a DC voltage ranging from −7 to 7 VDC, with a band excitation waveform of 1 VDC at 320–400 kHz. All AFM experiments were conducted using the conductive Pt/Cr-coated tip (BudgetSensors Multi75E-G).

Device fabrication and characterization

The PLD grown HZO layer on Si substrates were further subjected to the deposition (~9 nm) of high permittivity dielectric-HfO2 at 250 °C using ALD (Lucida D100, NCD Co., Ltd, Daejeon, Korea). Multi-layered MoS2 films (SPI Supplies, West Chester, PA, USA) were mechanically exfoliated using scotch tape and transferred on Si/HZO/HfO2 stack. The AFM measurements were conducted for measuring MoS2 thickness using an atomic force microscope (AFM: Park Systems, XE-7). Furthermore, E-beam-evaporated titanium/gold (20 nm/80 nm) was used as the top electrode. The source/drain was patterned by photolithography and the wet-etching process. The electrical properties of the fabricated devices were characterized using a semiconductor parameter analyzer (SCS-4200A, Keithley).

DATA AVAILABILITY

The data that support the findings of this study are available from the authors on reasonable request, see author contributions for specific data sets.

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ADDITIONAL INFORMATION

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