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## Subthreshold swing improvement in MoS<sub>2</sub> transistors by the negative-capacitance effect in a ferroelectric Al-doped-HfO<sub>2</sub>/HfO<sub>2</sub> gate dielectric stack

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Obtaining a subthreshold swing (SS) below the thermionic limit of 60 mV dec<sup>-1</sup> by exploiting the negative-capacitance (NC) effect in ferroelectric (FE) materials is a novel effective technique to allow the reduction of the supply voltage and power consumption in field effect transistors (FETs). At the same time, two-dimensional layered semiconductors, such as molybdenum disulfide (MoS<sub>2</sub>), have been shown to be promising candidates to replace silicon MOSFETs in sub-5 nm-channel technology nodes. In this paper, we demonstrate NC MoS<sub>2</sub> FETs by incorporating a ferroelectric Al-doped HfO<sub>2</sub> (Al:HfO<sub>2</sub>), a technologically compatible material, in the FET gate stack. Al:HfO<sub>2</sub> thin films were deposited on Si wafers by atomic layer deposition. Voltage amplification up to 1.25 times was observed in a FE bilayer stack of Al:HfO<sub>2</sub>/HfO<sub>2</sub> with a Ni metallic intermediate layer. The minimum SS (SS<sub>min</sub>) of the NC-MoS<sub>2</sub> FET built on the FE bilayer improved to 57 mV dec<sup>-1</sup> at room temperature, compared with SS<sub>min</sub> = 67 mV dec<sup>-1</sup> for the MoS<sub>2</sub> FET with only HfO<sub>2</sub> as a gate dielectric.

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## Introduction

For decades, computer chips have been solely made of silicon. However, doubling the number of chips on the same integrated circuit area, and thus following Moore's law, is becoming increasingly challenging as silicon technology reaches its physical limit. Two main paths are currently being pursued to continue the scaling of CMOS technology in the next few decades: 1. finding a new channel material that would allow electronics to move beyond silicon in miniaturization of the transistor channel length, and 2. determination of a new device mechanism to overcome the thermionic limit in metal oxide semiconductor field effect transistors (MOSFETs), thereby enabling reduction of the power consumption by further reducing the supply voltage.

Recently, layered two-dimensional (2D) semiconducting crystals of transition metal dichalcogenides (TMDs), such as molybdenum disulfide (MoS<sub>2</sub>) and tungsten diselenide (WSe<sub>2</sub>), have been proposed to enable aggressive miniaturization of

FETs.<sup>1–4</sup> The atomically-thin body thickness of TMDs improves the gate modulation efficiency. This can be seen in their characteristic scaling length,<sup>5</sup>  $\lambda = \sqrt{\frac{\epsilon_{\text{semi}}}{\epsilon_{\text{ox}}} t_{\text{ox}} \cdot t_{\text{semi}}}$ , where

$\epsilon_{\text{semi}}/t_{\text{semi}}$  and  $\epsilon_{\text{ox}}/t_{\text{ox}}$  are the dielectric constant/thickness of the channel and oxide, respectively.  $\lambda$  determines important short channel effects such as drain-induced barrier lowering and the subthreshold swing (SS). In particular, MoS<sub>2</sub> has a low dielectric constant of  $\epsilon = 4–7$  (ref. 6 and 7) and an atomically thin body ( $t_{\text{semi}} \approx 0.7 \text{ nm} \times \text{number of layers}$ ), which facilitate the decrease of  $\lambda$ , while its relatively high bandgap energy (1.85 eV for a monolayer) and high effective mass allow for a high on/off current ratio ( $I_{\text{on}}/I_{\text{off}}$ ) via reduction of direct source–drain tunneling.<sup>8</sup> These features make MoS<sub>2</sub>, and wide-bandgap 2D semiconductors in general, interesting candidates for low-power subthreshold electronics.

We have previously reported methods to reduce the channel length in MoS<sub>2</sub> FETs to 15 nm using graphene contacts<sup>3</sup> and 7.5 nm using directed self-assembly patterning.<sup>9</sup> Despite the advances made in TMD FET miniaturization, the power scaling in such devices suffers from the same issues than in Si MOSFET technology, where the supply voltage is limited by SS, which is the gate voltage change ( $\Delta V_{\text{g}}$ ) required to increase the source–drain current ( $I_{\text{ds}}$ ) by one decade:

$$\text{SS} = \frac{dV_{\text{g}}}{d\psi_{\text{s}}} \times \frac{d\psi_{\text{s}}}{d(\log I_{\text{ds}})}, \quad \frac{dV_{\text{g}}}{d\psi_{\text{s}}} = \left(1 + \frac{C_{\text{s}}}{C_{\text{ins}}}\right)$$

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where  $C_s$  and  $C_{\text{ins}}$  are the semiconductor and dielectric insulator capacitance,  $k$  is the Boltzmann constant,  $T$  is the temperature, and  $q$  is the elementary charge.

Limited by thermionic emission, Boltzmann statistics limits the second term in the above formula to  $\ln(10)kT/q$ , which is about 60 mV  $\text{dec}^{-1}$  at room temperature. The first term, which is known as the body factor  $m$ , is always greater than one. Therefore,  $SS > 60 \text{ mV dec}^{-1}$  at room temperature in a standard MOSFET, limiting the minimum supply voltage possible for a given drain current capability. To lower the SS and, in that way, the supply power, we need to overcome the limit of at least one of the terms in the above formula.

To overcome the limit of  $\frac{d\psi_s}{d(\log I_{\text{ds}})}$  a transport mechanism different from thermionic emission is required. The most widely studied method is tunnel-FETs, where carriers are injected into the channel by band-to-band tunneling from the source to the drain so over-barrier emission, as in MOSFETs, is circumvented. This allows steeper change of the current and thus overcomes the limit of 60 mV  $\text{dec}^{-1}$ . However, the experimental results reported so far severely suffer from the interface trap assisted tunneling mechanism, which generates a large background current obscuring the steepness of turn on of the band-to-band tunneling.<sup>10</sup>

An alternative method to reduce the SS below 60 mV  $\text{dec}^{-1}$  in a MOSFET is to decrease the body factor to less than one.<sup>11</sup> This requires the capacitance to be mathematically negative (*i.e.*,  $C_{\text{ins}} < 0$ ). The body factor is the inverse of the gate efficiency ( $\beta = d\psi_s/dV_g$ ), which is the rate that the semiconductor surface potential  $\psi_s$  changes by changing  $V_g$ . Therefore, if  $C_{\text{ins}} < 0$  then  $\beta > 1$ , a condition that is always impossible with conventional dielectric insulators.

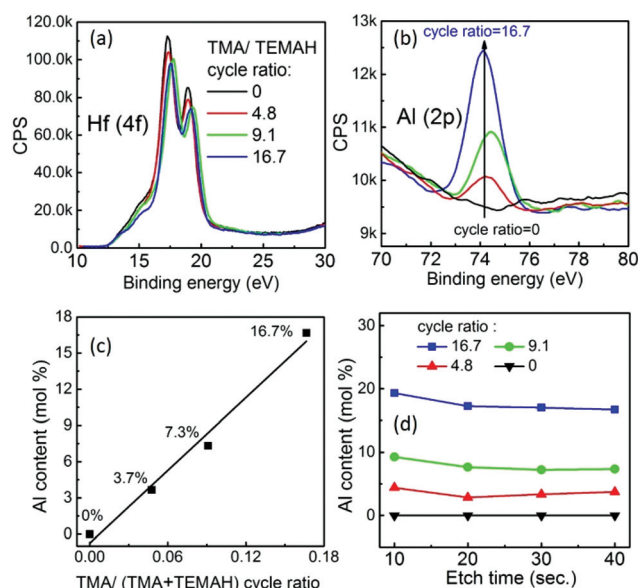
A negative differential capacitance effect has been experimentally observed in ferroelectric materials during the phase transition when the system is in nonequilibrium.<sup>12</sup> In addition, sub-60 mV  $\text{dec}^{-1}$  switching characteristics have been observed in several negative-capacitance (NC) FETs using lead zirconate titanate,<sup>13</sup> bismuth ferrite<sup>14</sup> and polymer ferroelectric dielectrics such as P(VDF)-TRFE.<sup>15,16</sup> However, neither of these materials is technologically compatible with standard Si CMOS technology. In this regard,  $\text{HfO}_2$  and  $\text{ZrO}_2$ <sup>17</sup> based ferroelectric materials are more favorable because they are fully compatible with the CMOS manufacturing process and their thin films can be obtained by standard deposition techniques, such as atomic layer deposition (ALD), and they can thus be easily integrated into transistor structures. The  $\text{HfO}_2$ -based gate dielectric has been extensively developed for CMOS applications with well-defined ALD processing technology, and it is used in current technology nodes. In addition, robust ferroelectricity has been reported in thin-films of  $\text{HfO}_2$  systems with various dopants, such as rare-earth,<sup>18</sup> Si,<sup>19</sup> and Al<sup>20</sup> dopants. Here, we used Al-doped  $\text{HfO}_2$  (Al: $\text{HfO}_2$ ) ferroelectric thin films to develop NC-MoS<sub>2</sub> FETs. We believe that incorporation of the NC mechanism in transistors with a 2D channel (*e.g.*, MoS<sub>2</sub> FETs) could be a solution to extend the boundaries of transistor dimension scaling as well as power scaling, and extend Moore's

law to its ultimate limit to enable the continued increase of the functionality of semiconductor electronics.

## Results and discussion

The atomic accuracy in the thickness of ALD films allows precise control of the molar ratio of the dopant to the host metal in bimetal oxide systems such as Al :  $\text{HfO}_2$ . In our work, ALD Al :  $\text{HfO}_2$  deposition was carried out at a substrate temperature of 250 °C using trimethylaluminum (TMA) as the Al precursor, tetrakis(ethylmethylamino)hafnium (TEMAH) as the Hf precursor, and  $\text{H}_2\text{O}$  as the oxidant. A Si wafer with a native  $\text{SiO}_2$  layer was used as the substrate. The Al composition of the films was varied from 0% to 16.7% by controlling the TMA/TMAHA pulses. The ALD process consisted of TMA cycles in the middle of sequences of TEMAH cycles. After deposition, the samples were rapid thermal annealed (RTA) in forming gas at 850 °C for 5 min to crystallize the films and intermix Al with the  $\text{HfO}_2$  body to form a homogenous Al :  $\text{HfO}_2$  matrix.

XPS analysis was performed on the Al :  $\text{HfO}_2$  films after varying the ratio of TMA cycles to TEMAH cycles (Fig. 1(a) and (b)). Fig. 1(c) shows a plot of the Al content of ~10 nm thin films extracted from the XPS analysis *versus* the TMA/TEMAH cycle ratio. The linear fit with near unity slope shows that the Al content can be precisely controlled in the range 0%–16.7%. Moreover, in-depth analysis showed negligible variation in the Al content after different etching times by Ar plasma, which indicates that the Al dopant atoms are uniformly dispersed in the  $\text{HfO}_2$  lattice.



**Fig. 1** (a) and (b) XPS analysis of Al-doped  $\text{HfO}_2$  films with different Al contents deposited on Si wafers with TMA/TEMAH cycle ratios ranging from 0 to 16.7%. (c) Al content of the Al :  $\text{HfO}_2$  films extracted from the XPS spectra shown in (a) and (b) as a function of the TMA/TEMAH cycle ratio. The inset shows a schematic of the deposited stacked  $\text{HfO}_2$  and  $\text{Al}_2\text{O}_3$  and homogeneously Al-doped  $\text{HfO}_2$  (Al :  $\text{HfO}_2$ ) formed after RTA. (d) Al contents for different etch times.

Fig. 2(a) compares the grazing incident X-ray diffraction (GIXRD) patterns of the undoped and 7.3% Al:HfO<sub>2</sub> samples. The XRD analysis suggests a phase transition from the monoclinic phase to the orthorhombic phase upon doping. This noncentrosymmetric transition phase is a prerequisite for ferroelectric characteristics. See ref. 17 for more detail on different phases of Al:HfO<sub>2</sub>. Next, we analyzed the ferroelectric capacitor with a 10 nm 7.3% Al:HfO<sub>2</sub> film. For this

measurement, a highly doped Si wafer was used as the back electrode and Ni pads were patterned on the thin film as the top electrode. An extra RTA step was performed at 450 °C after deposition of the Ni electrodes to eliminate charged interfacial trapped states.

Fig. 2(b) shows plots of the polarization *versus* electric field (*P*–*E*) hysteresis loop of 10 nm 0% and 7.3% Al:HfO<sub>2</sub> capacitors. Compared with the undoped sample, the 7.3% sample shows a *P*–*E* hysteresis loop, which confirms the ferroelectricity of the ALD grown thin film with a coercive field of 1.55 MV cm<sup>-1</sup> and remnant polarization of 9.5 μC cm<sup>-2</sup>. Fig. 2(c)–(e) show the transient voltage (*V<sub>F</sub>*), current (*I<sub>R</sub>*), and charge (*Q*) of the capacitor on application of a square voltage pulse (*V<sub>S</sub>*). A schematic of the measurement setup is shown in the insert of Fig. 2(d). *I<sub>R</sub>* is determined by  $I_R = (V_S - V_F)/R$ , where *R* is a 10 kΩ resistor and *Q* is calculated using  $Q = \int I_R(t) dt$ . The regions of interest are marked with dashed lines in Fig. 2(c) and (e), where *V<sub>F</sub>* and *Q* show different trends and  $dV_F/dt$  and  $dQ/dt$  have opposite signs. This indicates the presence of a negative capacitance feature in these regions ( $C = dQ/dV < 0$ ).

As previously discussed, the aim of incorporating a ferroelectric material in the gate stack of a MOSFET is to increase the gate efficiency above one by exploiting the NC effect of the ferroelectric material. Regardless of its absolute value, any NC can make the gate efficiency greater than one. However, it should be noted that the total gate capacitance *C<sub>g</sub>* has to remain positive to avoid instability in the system, which requires that  $|C_f| > C_s$  (where *C<sub>f</sub>* is the ferroelectric NC), otherwise it will result in strong hysteresis during device operation.<sup>21</sup> This condition can fail because *C<sub>s</sub>* is a nonlinear function of *V<sub>g</sub>* and *V<sub>d</sub>*, which can push the transistor to an unstable condition during its transition from depletion (low *C<sub>s</sub>*) to the inversion condition (high *C<sub>s</sub>*). To guarantee positive *C<sub>g</sub>* in the whole operation regime, a positive-capacitance dielectric oxide (*C<sub>ox</sub>*) can be added in series with the ferroelectric dielectric layer and the semiconductor. In this bilayer configuration,  $|C_f|$  has to be greater than the oxide dielectric capacitance ( $|C_f| > C_{ox}$ ) to amplify the gate efficiency ( $\beta > 1$ ) and greater than *C<sub>mos</sub>* =  $(C_s^{-1} + C_{ox}^{-1})^{-1}$  to meet the stability condition (*C<sub>g</sub>* > 0), which is always the case as long as the first condition ( $|C_f| > C_{ox}$ ) is met. However, despite this theoretical conditions for stabilization of NC, it should be noted that in a bilayer gate stack, the presence of any finite gate leakage does not allow NC full stabilization.<sup>22</sup> Nevertheless if the gate voltage is ramped up faster than the time required to discharge the capacitance (RC delay), the transistor can enter the NC regime.

Here, we used undoped HfO<sub>2</sub> as the positive-capacitance oxide component in the bilayer gate stack. To protect the surface potential in the device against the charge nonuniformity induced by microdomains in the ferroelectric film, the ferroelectric and the normal dielectric layers need to be separated by a metal layer to average out such non-uniformities.<sup>23</sup> To evaluate the potential amplification in the bilayer structure, we fabricated a test capacitor structure composed of Ni pads formed on the 10 nm 7.3% Al:HfO<sub>2</sub>/Si substrate, a 10 nm HfO<sub>2</sub> layer deposited on the Ni pads by ALD, and Au top elec-

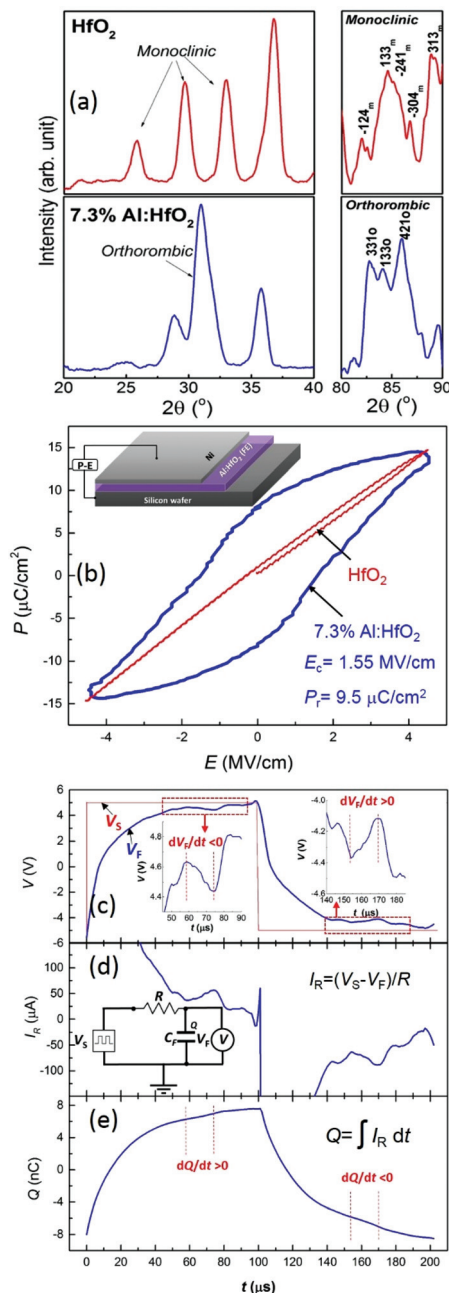
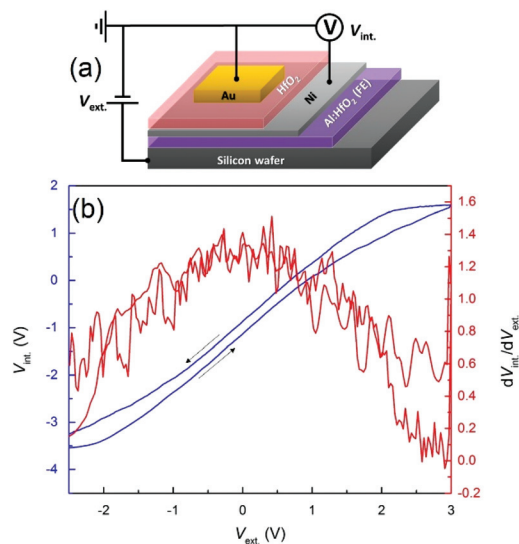


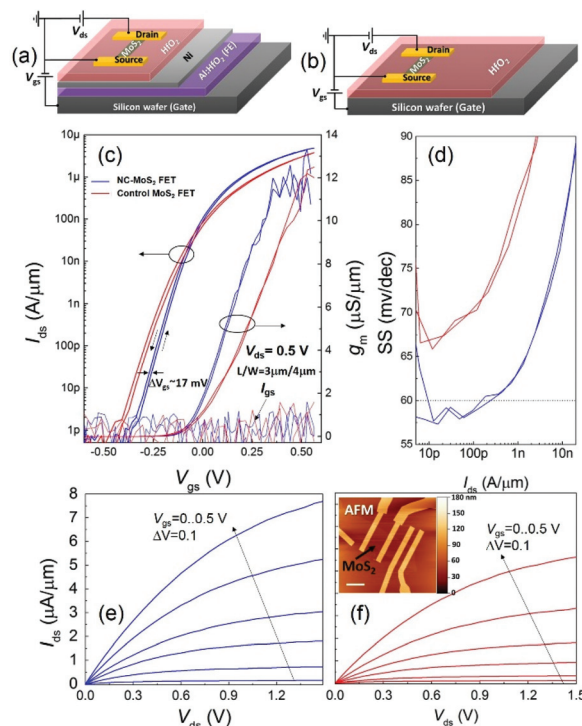
Fig. 2 (a) GIXRD patterns of undoped and 7.3% Al-doped HfO<sub>2</sub>. (b) Polarization *versus* electric field hysteresis of undoped and 7.3% Al-doped HfO<sub>2</sub>. (c–e) Transient voltage, current, and charge of 10 nm 7.3% Al-doped HfO<sub>2</sub> with a 200 μs period. The insert of (d) shows a schematic of the measurement setup.



**Fig. 3** (a) Schematic of the measurement setup used to characterize the potential amplification in the nominal 10 nm-Al:HfO<sub>2</sub>/10 nm-HfO<sub>2</sub> bilayer system with a Ni intermediate electrode.  $V_{\text{ext}}$  was applied to the Si wafer and the induced potential on the intermediate Ni electrode  $V_{\text{int}}$  was read in voltage mode (voltmeter). (b)  $V_{\text{int}}$  and  $dV_{\text{int}}/dV_{\text{ext}}$  versus  $V_{\text{ext}}$ .

trodes deposited on the HfO<sub>2</sub> layer (see Fig. 3(a)). In the device, contact holes were etched through HfO<sub>2</sub> to access the intermediate Ni layer.

Fig. 3(b) shows a plot of the potential of the intermediate Ni electrode  $V_{\text{int}}$  versus the potential applied to the doped-Si substrate,  $V_{\text{ext}}$ , when the top electrode is grounded. This shows how much  $V_{\text{int}}$  can be amplified by the NC effect. Using the capacitor divider method, the amplification factor is  $dV_{\text{int}}/dV_{\text{ext}} = [1/(1 + C_{\text{ox}}/C_f)]$ . The factor monotonically increases, exceeds one at  $V_{\text{ext}} \sim -1.5$  V, and reaches an average maximum gain of  $\sim 1.25$ . This confirms that the bilayer NC stack works properly and can potentially enhance the switching rate of a transistor when used as the gate stack. Next, we fabricated MoS<sub>2</sub> FETs by transferring an exfoliated tri-layer MoS<sub>2</sub> film onto the bilayer gate stack and formed source/drain contacts by electron-beam lithography. Fig. 4(a) and (b) show schematics of the NC-MoS<sub>2</sub> FET with a bilayer gate stack and the reference MoS<sub>2</sub> FET with only HfO<sub>2</sub> as the gate dielectric. Fig. 4(c) compares the transfer characteristics of the reference and NC devices. The measurements were performed in vacuum to eliminate the hysteresis induced by physisorbed moisture on the MoS<sub>2</sub> channel. Both devices show small hysteresis ( $\sim 17$  mV). To guarantee that the device was in transient mode during the measurement and thus enters the NC regime, the point-to-point delay time during the gate voltage sweep was set to 100  $\mu\text{s}$ . This was the minimum value allowed by the measurement setup and was sufficiently smaller than the gate RC delay time ( $>5$  ms). The device has an on current of  $5 \mu\text{A} \mu\text{m}^{-1}$  and maximum transconductance of  $12 \mu\text{S} \mu\text{m}^{-1}$ , and the off current is as low as  $1 \text{pA} \mu\text{m}^{-1}$ . The subthreshold characteristics of the device are of particular importance. In Fig. 3(b), the developed gate stack allows internal gain of up to



**Fig. 4** (a) Schematic of the NC-MoS<sub>2</sub> FET with a HfO<sub>2</sub>/Al:HfO<sub>2</sub> bilayer stack with Ni used as the intermediate metal. (b) Schematic of a reference regular MoS<sub>2</sub> FET with a HfO<sub>2</sub> gate dielectric. In both (a) and (b), a highly doped Si wafer is used as the back gate. (c) Transfer characteristics and transconductance of the NC-MoS<sub>2</sub> FET and reference MoS<sub>2</sub> FET at room temperature. (d) Comparison of the SS of the NC-MoS<sub>2</sub> FET with that of the reference MoS<sub>2</sub> FET. (e), (f) Output characteristics of NC-MoS<sub>2</sub> FET and control MoS<sub>2</sub> FET, respectively. The inset in (f) shows an AFM image of a MoS<sub>2</sub> FET. The scale bar is 4  $\mu\text{m}$ .

1.25 times. This internal gain can improve the subthreshold swing of the transistor and allows the  $60 \text{ mV} \text{ dec}^{-1}$  limit to be overcome without changing the transport mechanism. As shown in Fig. 4(d), the reference device has  $\text{SS}_{\text{min}} = 67 \text{ mV} \text{ dec}^{-1}$  at a current level of  $\sim 10 \text{ pA} \mu\text{m}^{-1}$  (Fig. 4(d)). The deviation of the SS from  $60 \text{ mV} \text{ dec}^{-1}$  is attributed to trapped charges at the MoS<sub>2</sub>/oxide interface, which lower the gate efficiency and thus increase the SS. The NC FET with the same MoS<sub>2</sub> channel length and thickness as well as the same MoS<sub>2</sub>/oxide interface shows significantly improved SS with a minimum of  $57 \text{ mV} \text{ dec}^{-1}$ , which is in agreement with the observed voltage amplification in Fig. 3(b). For the NC-MoS<sub>2</sub> FET, this allows transistor operation with  $I_{\text{off}} < 10 \text{ pA} \mu\text{m}^{-1}$  and  $I_{\text{on}}/I_{\text{off}} > 10^5$  with a  $\Delta V_{\text{gs}}$  of 0.5 V compared with a required  $\Delta V_{\text{gs}}$  of  $\sim 0.65$  V to achieve the same performance in the reference MoS<sub>2</sub> FET. Because the channel/oxide interface can degrade the SS, the SS can be further reduced by developing an ultraclean MoS<sub>2</sub>/oxide interface. Moreover, further optimization of the capacitance matching in the bilayer stack can improve the voltage amplification factor and thus further increase the gate efficiency.

It should be noted that NC technology is in its infancy and several aspects need to be experimentally investigated. In

particular, for targeted applications in state-of-the-art high-performance CMOS technology, the switching performance of NC FETs and the effects of dielectric thickness scaling and the gate-leakage current at high gate switching speeds on the amplification capability of the NC bilayer stack need to be extensively investigated.

## Conclusions

In summary, NC-MoS<sub>2</sub> FETs were fabricated using a CMOS compatible Al:HfO<sub>2</sub> ferroelectric thin film. ALD Al:HfO<sub>2</sub> films with different Al:Hf ratios were deposited on Si substrates and characterized by XPS, which showed excellent Al:Hf ratio control and dopant homogeneity in the host lattice. Gate voltage amplification up to 1.25 times was then demonstrated by incorporation of 10 nm 7.3% Al:HfO<sub>2</sub> in the dielectric stack of a MoS<sub>2</sub> FETs with 10 nm HfO<sub>2</sub> as a positive linear dielectric. The NC-MoS<sub>2</sub> FET showed a significant enhancement of the SS to 57 mV dec<sup>-1</sup>, while the absence of hysteresis showed the effective stabilization of the NC by using the HfO<sub>2</sub>/Al:HfO<sub>2</sub> bilayer.

## Experimental

All the electrical measurements were performed under vacuum with a Keysight B1500A Semiconductor Device Analyzer with a mid-power SMUs with a sweep measurement resolution of 100 μs. Briefly, the MoS<sub>2</sub> devices were prepared by the commonly used pickup and dry transfer method. The MoS<sub>2</sub> was mechanically exfoliated to obtain isolated flakes of a few layers, from commercially available bulk MoS<sub>2</sub> crystals on pre-cleaned (piranha solution, oxygen plasma, and solvent) substrates. A polydimethylsiloxane (PDMS) sheet was placed on a pre-cleaned glass slide. A 6%-solution of polypropylene carbonate (PPC, Sigma-Aldrich) in chloroform was then spin coated on the glass/tape/PDMS stack. This transfer slide was loaded into the probe arm of the transfer setup and brought into contact with the desired flake at room temperature. The stage was heated to 90 °C and maintained at that temperature for 1 min. After cooling the stage, the transfer slide was then slowly disengaged. The picked-up flake was transferred to the target substrate and heated to 155 °C to release the polymer. The polymer was dissolved in chloroform and the structure was cleaned with solvent and annealed in an Ar atmosphere at 360 °C for 3 hours.

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