2D-3D integration of hBN and a high-κ dielectric for ultrafast graphene-based electro-absorption modulators

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Electro-absorption (EA) waveguide-coupled modulators are essential building blocks for on-chip optical communications. Compared to state-of-the-art silicon (Si) devices, graphene-based EA modulators promise smaller footprints, larger temperature stability, cost-effective integration and high speeds. However, combining high speed and large modulation efficiencies in a single graphene-based device has remained elusive so far. In this work, we overcome this fundamental trade-off by demonstrating the first 2D-3D dielectric integration in a high-quality encapsulated graphene device. We integrated hafnium oxide (HfO2) and two-dimensional hexagonal boron nitride (hBN) within the insulating section of a double-layer (DL) graphene EA modulator. This novel combination of materials allows for a high-quality modulator device with record high performances: a ∼ 39 GHz bandwidth (BW) with a three-fold increase in modulation efficiency compared to previously reported high-speed modulators. This 2D-3D dielectric integration paves the way to a plethora of electronic and opto-electronic devices with enhanced performance and stability, while expanding the freedom for new device designs.

Broadband optical modulators with ultra-high speed, low-drive voltage and hysteresis-free operation are key devices for next-generation datacom transceivers. Although Si photonics is nowadays a prime candidate to fulfill these requirements,1,2 graphene is rapidly becoming a major contender in several optoelectronic applications, such as ultrafast modulators4,5 and silicon-integrated photodetectors6,7. Graphene-based modulators have already proven broadband optical bandwidth1, high-speed8,9, relatively high modulation efficiencies10 and temperature stability8. These devices are all based on CMOS compatible materials7,10-13, where CMOS design and fabrication techniques can be further leveraged to decrease costs. However, graphene-based modulators have yet to demonstrate all operation requirements at once. More specifically, EA graphene modulators struggle to show high-speed and high modulation efficiencies simultaneously14. This bottleneck is mostly due to the weak graphene/dielectric combination and the limited quality of the graphene.

Unlike Si technology, where high-κ dielectrics lie at the core of its success, 2D dielectrics are hindering the development of graphene- and other 2D-based electronics and optoelectronic devices1,13,15 and are clearly outperformed by traditional 3D high-κ dielectrics. This under-performing 2D-dielectric/graphene combination deepens even further the fundamental trade-off between speed and modulation efficiency inherent to the DL modulators14. In the DL architecture, the overlapped top and bottom graphene electrodes act as a capacitor (C). The larger the C, the higher the modulation efficiency. On the other hand, the speed of the modulator defined as f3dB = 1/(2πRC) is inversely proportional to C (R being the total resistance). In this framework, the quality of graphene appears as a valid turnaround to overcome this fundamental limitation. A high electron mobility is expected to minimize the overall resistance and reduce the insertion loss (IL)1,9, thus increasing the bandwidth and the extinction ratio (ER). However, the quality of graphene is very sensitive to its environment, e.g. the dielectric to encapsulate it. Indeed, no graphene/dielectric combination has been able to ensure high charge carrier mobilities and low levels of residual doping in existing graphene waveguide-coupled modulators.16. The growth of non-layered (i.e. 3D) dielectrics, e.g. aluminum oxide (Al2O3), silicon nitride (SiN) or HfO2 directly on top of graphene leads to low electronic mobility17-18 and/or inhomogeneous doping19.

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In this work, we demonstrate the 2D-3D integration of hBN and HfO2 within the dielectric section of a DL graphene EA modulator. This dielectric combination enhances the capacitance of the EA modulators without compromising its robustness against high voltages and preserves the high mobility and low doping of intrinsic graphene. As a result, we achieved a static and dynamic (at 40 Gbps) modulation efficiency as high as 2.2 dB/V and 1.49 dB/V, respectively, a f3dB bandwidth of ~39 GHz and a device footprint of 60 µm x 0.45 µm = 27 µm² (neglecting the metal pads and graphene leads). Moreover, the hBN-HfO2-hBN based devices show a symmetric and nearly hysteresis-free operation. The larger breakdown voltage of this 2D-3D dielectric, even beyond the full transparency regime (i.e., Pauli blocking), increases the ER and reduces the IL of the modulators.

I. RESULTS AND DISCUSSIONS

The EA modulators were fabricated on top of a photonic structure²⁰ formed by two gratings couplers²¹ feeding light in and out of an optical waveguide (Fig. 1a). The 750 nm-wide waveguide for the device in Fig. 1 was designed to support a single transverse-magnetic (TM) optical mode²⁰ (see sections III in SI). The presented DL graphene modulators were built, for the very first time, with hBN-encapsulated graphene top and bottom electrodes (Fig. 1b). The hBN-graphene-hBN stacks have been fabricated following state-of-the-art fabrication techniques²²,²³. This ensured low levels of doping and high charge carrier mobilities. We characterized the quality of the resulting modulators (sections II and VI in SI) and extracted a carrier density-independent mobility as high as 30,000 cm²/(Vs) at room temperature²³ (section II in SI).

Although hBN-encapsulated graphene devices have allowed for device designs with unprecedented functionalities²⁴–²⁶ and improved performance²³, such layered dielectric material typically contains impurities and/or crystal defects leading to low breakdown voltages²⁷,²⁸. Moreover, the dielectric permittivity of hBN is rather low compared to existing high-κ dielectrics²⁹, with a value close to that of SiO2 (εr ~ 4). This low dielectric constant and reduced breakdown voltage (see section V in SI) compromises not only the power consumption and the ability to reach high modulation efficiencies at reasonably low drive voltages but also limits the IL and the ER of the modulators.²⁹,³⁰ We thus integrate HfO2, a high-κ dielectric material, within the hBN-encapsulated graphene electrodes (see the sketch in Fig. 1b).

With such hBN-HfO2-hBN dielectric arrangement, graphene remains isolated from HfO2, shielded away from any possible out-of-plane dangling bonds of the 3D oxide material (see inset of Fig. 1b for the molecular represent-
tation of the 2D-3D dielectric interface). More importantly, the hBN-graphene interfaces remain atomically sharp and clean\textsuperscript{22,23,30}. This nanoscale control of the interfaces brings further advantages to real-world EA graphene modulators, like a symmetric and hysteresis-free operation. This is directly visible in the transmission curves as a function of the applied voltage $V_{\text{BT}}$, or, alternatively, as a function of the Fermi energy $E_F$ at the graphene electrodes (see bottom and top axis in Fig. 1c and section IX in SI). Both forward and backward voltage sweeps (black and blue traces, respectively) show minor hysteresis and appear symmetric with respect to the charge neutrality point. For comparison, a device fabricated with a HfO$_2$-hBN dielectric shows no overlap between the forward and backward sweeps (inset of Fig. 1c). This strong hysteresis is nonetheless expected for this HfO$_2$-hBN modulator since, in that case, the top graphene electrode is in direct contact with HfO$_2$. The HfO$_2$-HfO$_2$-hBN modulator device exhibits a modulation efficiency as high as $\sim 2.2\,\text{dB/V}$ within a 0.5 V voltage span (see red line linear fit to the data in Fig. 1c).

Considering the length of our modulator ($\sim 60\,\mu\text{m}$), we obtain a normalized static modulation efficiency of $\sim 0.037\,\text{dB/V}\mu\text{m}$, a three-fold increase compared to previously reported high-speed graphene EA modulators\textsuperscript{9}.

With such a high static modulation efficiency (Fig. 1), one might expect the device speed to be compromised\textsuperscript{14}. However, the high mobility of the hBN-encapsulated graphene is expected to increase the bandwidth. This is visible in Fig. 2a, where we calculated the $f_{\text{3dB}}$ bandwidth as a function of the charge carrier-dependent mobility ($\mu$) and the contact resistivity ($\rho_c$) for a graphene modulator with the same geometry and dielectric combination as the device in Fig. 1 (section XI in SI). As observed, the graphene mobility and the contact resistivity have a major influence on the modulator speed. Considering the mobility $\mu \approx 12,000\,\text{cm}^2/(\text{Vs})$ (evaluated at $V_{\text{BT}} = 10.4\,\text{V}$) and the contact resistivity $\rho_c \approx 800\,\Omega\cdot\mu\text{m}$ achieved experimentally (sections IV and XI in SI), we expect a bandwidth of $f_{\text{3dB}} \approx 46\,\text{GHz}$ (dashed lines in Fig. 2a). To confirm this value experimentally, we measured the electro-optical (EO) bandwidth of the device in Fig. 1 at a DC voltage $V_{\text{BT}} = 10.4\,\text{V}$ and a peak-to-peak voltage $V_{\text{AC}} = 200\,\text{mV}$ (Fig. 2b). The bandwidth of the measured device attains $f_{\text{3dB}} \approx 39\,\text{GHz}$ (without de-embedding, section XIII in SI). This value is close to the capabilities of our setup, limited to 40 GHz by the vector network analyzer (VNA) and the RF probes (section XII in SI). Even tough the measured $f_{\text{3dB}}$ does not reach the expected $f_{\text{3dB}} \approx 46\,\text{GHz}$ (Fig. 2a), possibly due to an increased contact resistivity of the measured device (section XI in SI), this is still the highest $f_{\text{3dB}}$ bandwidth among all graphene-based modulators reported so far\textsuperscript{8,9,11,12,24,35}.

The high-speed operation of our modulator device is also supported by non-return to zero (NRZ) eye diagram measurements. The data were obtained through an electrical pattern generator (PG) driving the modulator with a 2$^{31}$-1 pseudo-random binary sequence (PRBS)
FIG. 3. **Dielectric breakdown and Pauli blocking operation.** a. Maximum Fermi energy, noted $E_F^{\text{max}}$, expected at the graphene electrodes of a graphene modulator with a dielectric’s relative permittivity $\varepsilon_r$ and dielectric strength $E_{BD}$. All points lying inside the blue-colored region represent a dielectric allowing for Pauli blocking operation ($E_F^{\text{max}} > 0.5 \text{ eV}$, refer to section I in SI). The red-colored region indicates otherwise ($E_F^{\text{max}} < 0.5 \text{ eV}$). The white band represents the Pauli blocking boundary condition, defined as $E_F^{\text{max}} = 0.5 \text{ eV}$. The expected $E_F^{\text{max}}$ for HfO$_2$ and hBN are represented by the red and green squares respectively, taking the values of $E_{BD}$ and $\varepsilon_r$ from literature$^{28,31-33}$ (marked with dots) and our dielectric characterization (marked with stars, see sections V and X in SI). The black star represents the $E_F^{\text{max}} = 0.57 \text{ eV}$ expected for the hBN-HfO$_2$-hBN modulator in Fig. 1c (section X in SI). b and c, Normalized transmission as a function of $E_F$ and $V_{BT}$ for modulators with hBN (b) and hBN-HfO$_2$-hBN (c) dielectric. The data points are measurements and the solid curves simulations (see sections II-III and X in SI). The vertical dashed lines indicate the $E_F^{\text{max}}$ achieved at the dielectric breakdown. The orange-shaded regions show the full transparency range, i.e. Pauli blocking. The top $V_{BT}$ axis in panel b is for the 42 µm-long device only (see section VII in SI for the other hBN devices). The graphene Dirac cones in panel b show the absorption and Pauli blocking processes at low and high Fermi energies, respectively.

at 28 and 40 Gbps bit-rate (section XII in SI). The signal was driven by a 3.5 V peak-to-peak voltage while the DC bias was set to 11 V. The device was terminated with a 50 Ω load to avoid reflections due to the impedance mismatch between the PG electrical output and the modulator (when measured at 40 Gbps). Open eye-diagrams at 28 Gbps and 40 Gbps are shown in Fig. 2c, with an ER as high as 5.2 dB and a signal-to-noise ratio (SNR) of 2.28 dB for the latter (see section XIV in SI for an eye-diagram at 10Gbps). These results confirm the large modulation efficiency of our hBN-HfO$_2$-hBN-based modulator device, even at high speeds, with a record-high dynamic modulation efficiency of 1.49 dB/V at 40 Gbps$^9$.

Like the speed of the modulator, the power consumption, understood as the switching energy per bit, also benefits from the small footprint of the device. Ignoring the parasitic pad capacitance, we obtain for the modulator in Fig. 1 an energy per bit of $C(V_{AC})^2/4 \approx 160 \text{ fJ/bit}$, where $C = 52 \text{ fF}$ is the capacitance between the top and bottom graphene electrodes and $V_{AC} = 3.5 \text{ V}$ the voltage swing$^{12}$. This value of energy per bit is on par with state-of-the-art SiGe technologies$^{36,37}$.

To directly compare modulators with different dielectrics, it is more convenient to compare the transmission as a function of $E_F$ (see the $E_F$-axis in Fig. 1c and Fig. 3b and c) since $E_F$ already considers the thickness and the relative permittivity of the dielectric (section VII in SI). Operating the modulators at high $E_F$ enhances both ER and IL, with the ER (IL) increasing (decreasing) as a function of $E_F$.$^9$. In the full transparency regime (Pauli blocking, see section I in SI), the ER is maximized and the IL is expected to become nearly zero for high-quality graphene$^{3,9}$ (section X in SI). It is thus crucial to determine which dielectric materials facilitate Pauli blocking operation. Fig. 3a illustrates the expected maximum $E_F$,

$$E_F^{\text{max}} = h\nu_F \sqrt{\pi\varepsilon_0\varepsilon_r E_{BD}/q},$$

as a function of the relative permittivity ($\varepsilon_r$) and dielectric strength ($E_{BD}$) of any given dielectric. The square boxes in Fig. 3a enclose the expected $E_F^{\text{max}}$ for the HfO$_2$- and hBN-based modulators (in red and green, respectively) and the black star represents the $E_F^{\text{max}} = 0.57 \text{ eV}$ expected for the hBN-HfO$_2$-hBN modulator of Fig. 1c (section X in SI). The boundaries of the boxes are taken from literature$^{28,31-33}$ (marked with dots) and from our dielectric characterization (marked with stars, sections V and X in SI). All dielectric materials fulfilling $E_F^{\text{max}} > 0.5 \text{ eV}$ (see white fringe in Fig. 3a) allow full transparency, i.e. Pauli blocking. The comparison in Fig. 3a highlights the advantages of the hBN-HfO$_2$-hBN dielectric (black star), achieving higher $E_F$ values than the hBN.
Dynamic modulation efficiency [10^2 dB/(Vµm)]

Static modulation efficiency [10^2 dB/(Vµm)]

**FIG. 4. Comparison graph.** The black and red data points and axis represent the static modulation efficiency as a function of the f_{3dB} bandwidth and the dynamic modulation efficiency (extracted from eye-diagrams) as a function of the modulation speed, respectively. The red, blue and green data clouds enclose single-11,12,34,38 and double-layer8-10,35 graphene and silicon-39-41 state-of-the-art modulators operating at λ = 1.55 µm. Refer to sections XV and XVI in SI for a more detailed comparison of graphene-based modulators.

Although material platforms like Lithium Niobate42 (LiNbO_3) or hybrid technologies like Si/Indium Phosphide43 (InP), Si/Ge44 or InGaAlAs44 offer outstanding performances in modulator applications, those are either not scalable42,45 (LiNbO_3) or their integration with a CMOS fabrication line remains challenging44,46. Nowadays, Si and graphene are envisaged as the most scalable, cost-effective and CMOS compatible materials for amplitude modulator applications. To compare our results with state-of-the-art graphene and Si amplitude modulators, both EA and Mach-Zehnder interferometer configurations included, we summarize our results in Fig. 4 and in sections XV and XVI of SI. Fig. 4 shows the dynamic modulation efficiency (extracted from the eye-diagrams and normalized by the device length and drive voltage) as a function of the modulation speed (red axis and red data point in Fig. 4) and the static modulation efficiency (measured in DC and normalized by the device length), as a function of the f_{3dB} bandwidth (black axis and black data point in Fig. 4). To avoid discrepancies due to the different extraction methods, we determine the static modulation efficiency of the compared literature8-12 using the same method as in Fig. 1c, i.e. by applying a linear fit within a 0.5 V voltage span. Results highlight the trade-offs between speed and modulation efficiency and stresses the advantages of a hBN-HfO_2-hBN dielectric to obtain large static and dynamic modulation efficiencies even at high speed. As observed, the modulation efficiency typically drops for devices with high speed8-9, being our device the only modulator able to operate at high speed with a large static and dynamic modulation efficiency (Fig. 4). These results outperform state-of-the-art graphene and not yet commercial silicon-based electro-absorption modulators39-41 (see blue/red and green data clouds, respectively in Fig. 4) when considering the modulation efficiency normalized by the length (i.e. footprint). This figure-of-merit is rather an important one since for many envisaged applications (e.g. chip interconnects) multiple
modulator devices are expected to coexist on the same chip.

II. CONCLUSION

With this work, we demonstrated the advantages of integrating hBN with a 3D high-\( k \) dielectric for high-quality graphene-based EA modulators. Compared to traditional oxide sputtering or ALD-growth on top of graphene, the integration of HfO\(_2\) in between hBN prevented any damage of the underlying graphene and allowed clean graphene-hBN interfaces. These clean interfaces yielded a symmetric and nearly hysteresis-free operation. Moreover, this 2D-3D integration enabled full transparency while maintaining the high mobility and low doping of intrinsic graphene. More importantly, the hBN-HfO\(_2\)-hBN based EA modulators were able to reach high modulation speeds with strong modulation efficiencies, overcoming the fundamental limitations of the DL graphene configuration and outperforming state-of-the-art graphene and Si technologies. The compatibility of this hBN-HfO\(_2\)-hBN dielectric with Si and other 2D materials might allow for considerable scaling improvements and greater device functionality in a broad range of graphene- and 2D-based electronic and optoelectronic applications, even beyond graphene-based modulators.

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CONTRIBUTIONS

B.T., H.A., and F.H.L.K. conceived the idea. H.A., and B.T. fabricated the devices. L.O., B.T. did the simulations. B.T., H.A. performed the measurements and data analysis. A.M., V.S. performed high frequency measurements under the supervision of M.R.. M.P., and D.V.T. provided Si waveguides. K.W., and T.T. synthesized the h-BN crystals. F.H.L.K., and B.T., supervised the project. B.T., H.A., and F.H.L.K. wrote the manuscript with input from all authors.

DATA AVAILABILITY

The data that support the plots within this paper and other findings of this study are available from the corresponding author upon reasonable request.

COMPETING FINANCIAL INTERESTS

The authors declare no competing financial interests.

METHODS

The Si photonic waveguide with a core cross-section of 750 nm × 220 nm was prepared on the IMEC iSiPP25G silicon on insulator (SOI) platform. For the fabrication of the electro-absorption modulator (EAM), the graphene and hBN flakes were exfoliated from highly oriented pyrolytic graphite (HOPG) and hBN crystals, respectively. The bottom hBN-graphene-hBN stacks was prepared by the van der Waals assembly technique and transferred directly onto the Si waveguide separated by a 10 nm spacer of high-quality thermal SiO₂. The bottom hBN flake (separating the graphene and SiO₂ layer) thickness of ~5 nm was chosen to enhanced the graphene absorption while isolating the graphene from the rough SiO₂ substrate. The top hBN has a thickness of ~10 nm. The stack has been etched by reactive ion etching (RIE) in an oxygen (O₂) and Trifluoromethane (CHF₃) (4:40 sccm) environment to expose the graphene edge. The bottom stack was then contacted by a 3/15/30 nm Cr/Pd/Au metal combination. The 10 nm hafnium oxide film has been deposited at 250°C prior deposition of a 2 nm sputtered SiO₂ seed layer by atomic layer deposition (ALD). Tetrakis(dimethylamido) hafnium (TDMAH) (0.4 sec purge time) and water vapor (5 sec purge time) as precursors have been used in a Savannah G1 system from Cambridge Nanotech. The top hBN-graphene-hBN stack with a 7 nm- and 21 nm-thick bottom and top hBN layers has followed the same fabrication steps as the bottom stack.