2D-3D integration of hexagonal boron nitride 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 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 combination of materials allows for a high-quality modulator device with 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, graphene is rapidly becoming a major contender in several optoelectronic applications, such as ultrafast modulators and silicon-integrated photodetectors. Graphene-based modulators have already proven broadband optical bandwidth, high speed, relatively high modulation efficiencies, and temperature stability. These devices are all based on complementary metal–oxide–semiconductor (CMOS)-compatible material, where CMOS design and fabrication techniques can be further leveraged to decrease costs. However, graphene-based modulators are yet to demonstrate all operation requirements at once. More specifically, EA graphene modulators struggle to show high-speed and high modulation efficiencies simultaneously. This bottleneck is mostly due to the weak graphene/dielectric combination and the limited quality of 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 devices and are clearly outperformed by traditional 3D high-κ dielectrics. This underperforming 2D-dielectric/graphene combination deepens even further the fundamental trade-off between speed and modulation efficiency inherent to the double-layer (DL) modulators. In the DL architecture, the overlapped top and bottom graphene electrodes act as a capacitor. The larger the C, the higher the modulation efficiency. On the other hand, the speed of the modulator defined as \( f_{\text{mod}} = 1/(2\pi RC) \) 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. High electron mobility is expected to minimize the overall resistance and reduce the insertion loss and the extinction ratio (ER). However, the quality of graphene is very sensitive to its environment, e.g., the dielectric to encapsulate it.
As observed, the graphene mobility and the contact resistivity have a major influence on the modulator speed. Considering the mobility $\mu \approx 12,000 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ (evaluated at $V_{BT} = 10.4 \text{ V}$) and the contact resistivity $r_c \approx 800 \Omega \mu m$ achieved experimentally (see Supplementary Notes 4 and 11), we expect a bandwidth of $f_{3dB} \sim 46 \text{ GHz}$ (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 $V_{BT} = 10.4 \text{ V}$ and a peak-to-peak voltage $V_{AC} = 200 \text{ mV}$ (Fig. 2b). The bandwidth of the measured device attains $f_{3dB} \approx 39 \text{ GHz}$ (without de-embedding, see Supplementary Note 13). This value is close to the capabilities of our setup, limited to 40 GHz by the vector network analyzer and the RF probes (see Supplementary Note 12). Even though the measured $f_{3dB}$ does not reach the expected $f_{3dB} \sim 46 \text{ GHz}$ (Fig. 2a), possibly due to an increased contact resistivity of the measuring device (see Supplementary Note 11), this is still the highest $f_{3dB}$ bandwidth among all graphene-based modulators reported so far.30,11,12,31,32.

With such a high static modulation efficiency (Fig. 1), one might expect the device speed to be compromised. 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 $f_{3dB}$ bandwidth as a function of the charge carrier-dependent mobility ($\mu$) and contact resistivity ($\rho_c$) for a graphene modulator with the same geometry and dielectric combination as the device in Fig. 1 (see Supplementary Note 11). As observed, the graphene mobility and the contact resistivity have a major influence on the modulator speed. Considering the mobility $\mu \approx 12,000 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ (evaluated at $V_{BT} = 10.4 \text{ V}$) and the contact resistivity $\rho_c \approx 800 \Omega \mu m$ achieved experimentally (see Supplementary Notes 4 and 11), we expect a bandwidth of $f_{3dB} \sim 46 \text{ GHz}$ (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 $V_{BT} = 10.4 \text{ V}$ and a peak-to-peak voltage $V_{AC} = 200 \text{ mV}$ (Fig. 2b). The bandwidth of the measured device attains $f_{3dB} \approx 39 \text{ GHz}$ (without de-embedding, see Supplementary Note 13). This value is close to the capabilities of our setup, limited to 40 GHz by the vector network analyzer and the RF probes (see Supplementary Note 12). Even though the measured $f_{3dB}$ does not reach the expected $f_{3dB} \sim 46 \text{ GHz}$ (Fig. 2a), possibly due to an increased contact resistivity of the measuring device (see Supplementary Note 11), this is still the highest $f_{3dB}$ bandwidth among all graphene-based modulators reported so far.30,11,12,31,32.

The high-speed operation of our modulator device is also supported by non-return-to-zero 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 at 28 and 40 Gbps bit-rate (see Supplementary Note 13). 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 and 40 Gbps are shown in Fig. 2c, with an ER as high as 5.2 dB and a signal-to-noise ratio of 2.28 dB for the latter (see Supplementary Note 14 for an eye diagram at 10 Gbps). These results confirm the large modulation efficiency of our hBN–HfO2–hBN-based modulator device, even at high speeds, with a dynamic modulation efficiency of 1.49 dBV$^{-1}$ at 40 Gbps.8.
Fig. 3 Dielectric breakdown and Pauli blocking operation. a Maximum Fermi energy, noted $E_F^{max}$, 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 ($E_F^{max}$ > 0.5 eV, refer to Supplementary Note 1). The red-colored region indicates otherwise ($E_F^{max}$ < 0.5 eV). The white band represents the Pauli blocking boundary condition, defined as $E_F^{max}$ = 0.5 eV. The expected $E_F^{max}$ 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 (marked with dots) and our dielectric characterization (marked with stars, see Supplementary Notes 5 and 10). The black star represents the $E_F^{max}$ = 0.57 eV expected for the hBN–HfO$_2$–hBN modulator in Fig. 1e (see Supplementary Note 10). b, c Normalized transmission as a function of $E_F$ and $V_{g2}$ for modulators with hBN (b) and hBN–HfO$_2$–hBN (c) dielectric. The data points are measurements and the solid curves simulations (Supplementary Notes 1–3 and 10). The vertical dashed lines indicate the $E_F^{max}$ 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 Supplementary Note 7 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.

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 $C(V_{AC})_F^2/2 \approx 160 \frac{fJ}{bit}$, where $C = 52 \mu F$ is the capacitance between the top and bottom graphene electrodes and $V_{AC} = 3.5 \text{ V}$ the voltage swing. This value of energy per bit is on par with state-of-the-art SiGe technologies.

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 Figs. 1e and 3b and c) since $E_F$ already considers the thickness and the relative permittivity of the dielectric (see Supplementary Note 7). Operating the modulators at high $E_F$ enhances both ER and IL, with the ER (IL) increasing (decreasing) as a function of $E_F$. In the full transparency regime (Pauli blocking, see Supplementary Note 1), the ER is maximized and the IL is expected to become nearly zero for high-quality graphene (see Supplementary Note 10). It is thus crucial to determine which dielectric materials facilitate Pauli blocking operation. Figure 3a illustrates the expected maximum $E_F$,

$$E_F^{max} = \frac{\hbar v_F}{\pi \varepsilon_r \varepsilon F_{BD}} / q,$$

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 $E_F^{max}$ for the HfO$_2$—and hBN-based modulators (in red and green, respectively) and the black star represents the $E_F^{max}$ = 0.57 eV expected for the hBN–HfO$_2$–hBN modulator of Fig. 1e (see Supplementary Note 10). The boundaries of the boxes are taken from literature (marked with dots) and from our dielectric characterization (marked with stars, see Supplementary Notes 5 and 10). All dielectric materials fulfilling $E_F^{max}$ > 0.5 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 dielectric while equally preserving the intrinsic qualities of graphene. These results are confirmed by the transmission traces in Fig. 3b, c. None of the hBN-based modulators were able to withstand Pauli blocking operation (orange-shaded region Fig. 3b), all breaking their hBN dielectric at a similar $E_F^{max} \approx 0.4 \text{ eV}$ (see vertical dashed lines in Fig. 3b and Supplementary Notes 7 and 8). Even though these hBN-based modulators were too fragile, we obtained modulation efficiencies as high as 0.3, 1.3, and 2 dB·V$^{-1}$ for device lengths $L = 12$, 24, and 42 μm, respectively. Once normalized by its length, we obtain 0.025, 0.054, and 0.047 dB·V$^{-1}$·μm$^{-1}$. These results exceed the state-of-the-art modulation efficiency of 0.038 dB·V$^{-1}$·μm$^{-1}$ still. The prematurity hBN breakdown compromises the ER and the IL. Indeed, the measured ER = 0.75, 2.3, and 4.9 dB (data points in Fig. 3b) is far from the simulated ER = 1.8, 4.4, and 7.9 dB (solid traces in Fig. 3b) expected for the 12, 24, and 42 μm-long modulators, respectively (for simulations, refer to Supplementary Notes 1–3).

Likewise, the measured IL = 1, 2.2, and 3.4 dB are higher than IL = 0 dB expected for high-mobility graphene modulators (see the minimum 0 dB normalized transmission, i.e., neglecting the losses from grating couplers and Si waveguide, achieved by the simulation traces in Fig. 3b and Supplementary Note 10).

On the other hand, the second hBN–HfO$_2$–hBN modulator device attains the Pauli blocking regime (Fig. 3c), in agreement with the dielectric characterization of hBN–HfO$_2$–hBN (Fig. 3a and Supplementary Notes 5 and 10), reaching a maximum Fermi energy of $E_F^{max}$ ≈ 0.54 eV. The ER and IL improve accordingly, with an ER = 7.8 dB almost twice the value obtained by the hBN-based modulator of comparable length (compare the black and red traces of Fig. 3c, b, respectively) and an IL reaching nearly zero (IL = 0.04 dB in Fig. 3c and Supplementary Note 10). However, being shorter ($L = 44 \mu m$) than the device in Fig. 1e ($L = 60 \mu m$), the modulation efficiency is lower (1.3 dB·V$^{-1}$ in a 0.5 V span, see Fig. 3c). We note that the hBN–HfO$_2$–hBN device of Fig. 1e has a relatively weak measured ER = 4.4 dB and IL = 7.8 dB (see Supplementary Note 10) due to an overcautious $V_{BT}$ = 12.1 V applied voltage (or alternatively $E_F = 0.41 \text{ eV}$).
In this work, we demonstrated the advantages of integrating hBN with a 3D high-κ dielectric for high-quality graphene-based EA modulators. Compared to traditional oxide sputtering or atomic layer deposition (ALD) growth on top of graphene, the integration of HfO$_2$ in-between hBN prevented any damage to 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, over- coming 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.

**Methods**

**Device fabrication.** The Si photonic waveguide with a core cross-section of 750 nm × 220 nm was prepared on the IMEC iSiPP25G silicon-on-insulator platform. For the fabrication of the electroabsorption modulator, the graphene and hBN flakes were exfoliated from highly oriented pyrolytic graphite and hBN crystals, respectively. The bottom hBN–graphene–hBN stacks were 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 SiO$_2$. The bottom hBN flake (separating the graphene and the SiO$_2$ layer) thickness of ~5 nm was chosen to enhance the absorption while isolating the graphene from the rough SiO$_2$ substrate. The top hBN has a thickness of ~10 nm. The stack has been etched by reactive ion etching in an oxygen (O$_2$) and trifluoromethane (CHF$_3$) (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 depositions of a 2 nm sputtered SiO$_2$ seed layer by ALD. Tetraakis(dimethylamido) hafnium (TDMH) (0.4 s purge time) and water vapor (5 s 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.

**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.

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