Fundamental limits and near-optimal design of graphene modulators and non-reciprocal devices

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The potential of graphene for photonic applications was evidenced by recent demonstrations of modulators, polarization rotators and isolators. These promising yet preliminary results raise crucial questions: what is the optimal performance achievable by more complex designs and how can this optimum be achieved in practice? We answer by first demonstrating that the relevant figures of merit for the devices above are subject to absolute theoretical upper bounds. Strikingly, these limits are related only to the conductivity tensor of graphene; thus, we can provide essential roadmap information such as the best possible device performance versus wavelength and graphene quality. Second, based on the theory developed, physical insight and detailed simulations, we demonstrate how structures closely approaching these fundamental limits can be designed, demonstrating the possibility of significant improvement. These results are believed to be of paramount importance for the design of modulators, rotators and isolators using graphene or other two-dimensional materials.

In recent years there has been a surge of interest in graphene-based devices for various photonic and optoelectronic applications. Among other very promising passive devices (including amplitude modulators), Faraday and Kerr polarization rotators and non-reciprocal isolators have been demonstrated. These essential photonic blocks might constitute some of the most important applications of graphene. Thus, there is a significant and growing effort to transform the above-mentioned initial demonstrations into real capabilities. However, this prospect still requires drastic performance improvements. For this purpose, researchers have now started to develop graphene-based devices with multiple degrees of freedom by considering several technological solutions (Fig. 1e), including multilayer structures, graphene patternings, substrate engineering and combining metal and graphene.

Except for very particular cases, structures as complex as those listed above cannot be modelled analytically, so the analysis of a single design variation entails intensive numerical electromagnetic simulations. Moreover, the best performance will typically be achieved by combining several of the above approaches, resulting in a very large optimization space. Consequently, today, the design process is left to trial-and-error optimization procedures that are based on heavy electromagnetic simulations. Such procedures are not only time-consuming, but are also bound to yield suboptimal performance or unnecessarily complex structures. Furthermore, the high sensitivity of the conductivity of graphene to fabrication or applied bias fields is a graphene-specific issue and, given the lack of analytical solutions available, it is very impractical to distinguish potential performance improvements that are due to higher graphene quality from those due to improved device design.

Our results constitute a disruptive and far-reaching solution to the problems above. First, we demonstrate that there are absolute theoretical limits to the performance of graphene-based modulators, non-reciprocal polarization rotators, and isolators. Remarkably, these upper bounds depend only on the conductivity of the graphene used to implement the devices. Thus, the minimum graphene quality, doping and bias fields required to enable a given desired performance can be determined before any design or simulation. Similarly, for each application of interest, we calculate the best possible performance achievable as a function of frequency or any parameter influencing graphene conductivity, providing crucial ‘roadmap’ information for graphene photonics.

The above fundamental results take on their full practical relevance if one can design devices for which the performance closely approaches the theoretical limit. Here, we demonstrate that such near-optimal devices can indeed be designed, which also validate our theoretical results. Furthermore, based on physical insight and concrete examples, we provide guidelines for designing low-complexity devices with quasi-optimal performance.

The results are based on rigorous theory and are applicable to virtually all graphene-based linear reconfigurable or non-reciprocal photonic devices, based on either guided or free-space waves, under the assumption that the used materials have local constitutive equations. Additionally, the methodology can be readily applied to other two-dimensional materials, such as MoS$_2$, or to two-dimensional electron gases (2DEGs) in semiconductors. For illustration purposes, four representative applications of significant interest are addressed in detail (Fig. 1a–d), including amplitude modulation in reflection and transmission, magneto-optic Kerr rotation, and non-reciprocal isolation based on Faraday rotation.

Electro-optical graphene amplitude modulators

Electro-optic modulation is the most studied application based on the dynamic reconfiguration of graphene conductivity. The practical feasibility of these devices has been verified at different frequencies, ranging from the infrared to kilohertz. In particular, graphene demonstrates a remarkable potential for modulation at terahertz frequencies, where alternative technologies are subject to significant limitations.

Graphene modulators, either in guided-wave systems or as metasurfaces for free-space beams, can be rigorously described using scattering matrix formalism, which relates incident waves to reflected waves with $b = 2a$. The scattering matrix of the

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Figure 1 | Graphene-based capabilities used for a detailed illustration of the method and ‘design space’ dimensions. The device capabilities are classified according to the direction (reflection or transmission) and framework (non-reciprocity or reconfigurability). a, Amplitude modulation in reflection. b, Amplitude modulation in transmission. c, Magneto-optic Kerr rotation. d, Isolation. e, Main independent degrees of freedom for the design of the devices.

A modulator takes two distinct values, \( S_A \) and \( S_B \), for the two scalar conductivities \( \sigma_A \) and \( \sigma_B \) of graphene, obtained by applying different electrostatic bias fields. Here, we consider amplitude modulators both in reflection (for example, ref. 32) and in transmission (for example, ref. 16). A modulator in reflection is a one-port device (Fig. 1a) whose scattering matrix is a scalar with values \( S_A = |T_A| \) and \( S_B = |T_B| \). For modulators in transmission, the input and output signals flow through different ports, and the scattering matrices are \( 2 \times 2 \) and symmetrical:

\[
\begin{bmatrix}
S_A & \frac{1}{|T_A|^2} + \sigma_A \\
\frac{1}{|T_A|^2} + \sigma_A & S_B
\end{bmatrix}
\]

where \( T_A \) and \( T_B \) represent the transmission coefficients responsible for the modulation and the other terms represent undesired reflections. In the following we assume that the variation of graphene conductivity is much slower than the time oscillation of the optical electromagnetic fields.

Applying the mathematical formulation described in the Supplementary Methods to the scattering matrices defined above, the following inequalities are obtained:

\[
\gamma_m = \frac{|\sigma_A - \sigma_B|^2}{4Re(\sigma_A)Re(\sigma_B)}
\]

Inequalities (2) and (3) are hereafter referred to as ‘amplitude modulation inequalities’. The left term \( \gamma_{m,T} \) is only a function of the two reflection/transmission coefficients, whereas \( \gamma_m \) is a positive real scalar that depends only on the graphene conductivities \( \sigma_A \) and \( \sigma_B \), as shown in expression (4).

Helpful intuition regarding the inequality introduced is obtained by considering the Cartesian plane \((|T_A|^2, |T_B|^2))\), as depicted in Fig. 2a. For brevity, only the case of reflection is considered here, but the illustration applies equally to transmission modulators due to the similarity of inequalities (2) and (3). Inequality (2) states that there are some areas in the space of \((|T_A|^2, |T_B|^2))\) that are strictly forbidden; these regions are highlighted in yellow in Fig. 2a. The boundaries of these forbidden regions are determined by \( \gamma_m \) and thus solely by graphene conductivity. In this plot, an ideal modulator corresponds to the red squares, that is, \( |T_A|^2 = 1 \) and \( |T_B|^2 = 0 \), or vice versa. However, inequality (2) readily shows that such an ideal modulator is not practically realizable. For example, if the modulator is designed to achieve perfect absorption in its ‘off’ state, such that \( |T_B|^2 = 0 \), then \( |T_A|^2 \) cannot exceed the value shown by the red circle in Fig. 2a.

A more practical view is obtained by representing the same data in the Cartesian plane defined by insertion loss and modulation depth (Fig. 2b) is representation is accomplished by assuming, without loss of generality, that \( |T_A|^2 \geq |T_B|^2 \) and by defining the modulation depth as \( h = (|T_A|^2 - |T_B|^2)/(|T_A|^2 + |T_B|^2) \) and the insertion loss a \( |T_A|^2 \). In this case, inequality (2) can be rewritten as

\[
\gamma_{m,T}(|T_A|^2, h) = \frac{2h|T_A|^2}{1 - |T_A|^2}\left(1 - |T_A|^2\right) \leq \gamma_m
\]

For example, the best possible modulator with 100% modulation depth will have a loss of \( |T_A|^2 = \sqrt{\gamma_m/(1 + \gamma_m)} \), which is obtained by introducing \( h = 1 \) in expression (5). This case is represented by the red circle in Fig. 2b, which corresponds exactly to the red circle in the alternative representation in Fig. 2a.

In summary, the theory indicates that a given target value for modulation depth cannot be reached without a minimum value for insertion loss. The general trend is intuitive and related to the loss in graphene. More precisely, a high modulation originates from a strong interaction of the fields with graphene, which in turn increases the loss. However, here, we rigorously demonstrate that although arbitrarily complex designs might potentially allow the modulation depth to be increased without limit, such unbounded increases will always come at the cost of a minimum amount of loss. Importantly, this minimum amount of loss is a known function of the parameters of graphene alone, which allows us to fully utilize the developed theory for practical purposes, as detailed below.

The bound \( \gamma_m \) is a function of graphene conductivity (see expression (4)), and Fig. 2c presents the frontiers corresponding to different values of \( \gamma_m \). Modulators with different amplitudes that are available in the literature are also reported on the graph, where possible with their corresponding frontiers. The performance achieved in these initial concept demonstrations is typically significantly below the theoretical upper bound. This result suggests the important potential for improvement if devices approaching the theoretical limit can indeed be designed in practice.

To provide our first answers to this question and subsequently to validate the theoretical prediction, we simulated a very large number of randomly generated modulators. All modulators use graphene of...
represents a single simulated device. The frequency considered is 1 THz and the graphene parameters are $g$ (here, $g_{\text{M}} = 0.6$ is used as an example, see expression (4)). The squares represent ideal modulators, and the circles denote the best possible modulators with 100% modulation depth. Forbidden areas (yellow) are delimited by the boundary curve, where $\gamma_{\text{mod}} = \gamma_{\text{M}}$. As in a, but using the insertion loss and modulation depth coordinates. c, Upper bounds for different values of $\gamma_{\text{M}}$. The available designs in the literature are represented by coloured symbols and, where possible, the corresponding bound is represented using the same colour. d-f, Simulations of randomly generated reflection modulators. Each red point represents a single simulated device. The frequency considered is 1 THz and the graphene parameters are $T = 300$ K, $\mu_{\text{h}} = 0.1$ eV, $\mu_{\text{d}} = 0.8$ eV and $\tau = 66$ fs (leading to $\gamma_{\text{M}} = 1.76$). g-i, Random simulations of different device topologies for transmission modulation. g and h represent random sequences of graphene sheets (patterned in h) and dielectric layers.

Figure 2 | Performances of electro-optical modulators. a, Graphical representation of the amplitude modulation inequality in the Cartesian plane ($|F_a|, |F_b|$) (here, $\gamma_{\text{M}} = 0.6$ is used as an example, see expression (4)). The squares represent ideal modulators, and the circles denote the best possible modulators with 100% modulation depth. Forbidden areas (yellow) are delimited by the boundary curve, where $\gamma_{\text{mod}} = \gamma_{\text{M}}$. b, As in a, but using the insertion loss and modulation depth coordinates. c, Upper bounds for different values of $\gamma_{\text{M}}$. The available designs in the literature are represented by coloured symbols and, where possible, the corresponding bound is represented using the same colour. d-f, Simulations of randomly generated reflection modulators. Each red point represents a single simulated device. The frequency considered is 1 THz and the graphene parameters are $T = 300$ K, $\mu_{\text{h}} = 0.1$ eV, $\mu_{\text{d}} = 0.8$ eV and $\tau = 66$ fs (leading to $\gamma_{\text{M}} = 1.76$). g-i, Random simulations of different device topologies for transmission modulation. g and h represent random sequences of graphene sheets (patterned in h) and dielectric layers.

Therefore, the logical next step is to consider the simplest modulators that provide additional design flexibility, namely, graphene patterns (Fig. 2e) or the use of an additional dielectric layer before graphene (Fig. 2f). The distribution of the computed results (red dots) provides evidence that these minimal increments in device complexity allow an almost arbitrary approach to the absolute theoretical upper bound, which is of considerable practical importance. The numerical results presented correspond to the conductivity of the given selected graphene, but the conclusions apply to any conductivity. The detailed parameters and performance of designs close to the optimal performance are provided in the Supplementary Methods. The method is bandwidth agnostic, so different near-optimal solutions can be compared and selected according to the bandwidth requirement.

The simulations provide convincing evidence of the bound validity. The frontiers of the performance clouds for the randomly generated modulators almost exactly match the theoretical upper bound. Importantly, many more simulations of complex randomly generated set-ups were carried out, including combinations of a multilayer substrate, patterned graphene and the addition of metal. No single result was found to exceed the theoretical limit.

The optimal performances for modulators in transmission are more difficult to attain. Despite closely approaching the bound for
most modulation depth, non-patterned and patterned multilayer structures are suboptimal when 100% modulation is desired (Fig. 2g,h). The physical interpretation is that, in contrast to modulators in reflection, a good high-transmission modulation state not only requires limiting loss in graphene but also must independently impedance match the system. The operation of a polarization twist upon transmission, which is achieved in Fig. 2i by using highly anisotropic hybrid graphene–metal patterns, offers a new degree of freedom. The hybrid metasurfaces modulate one polarization, while the other polarization is left largely unaffected, allowing for feedback in the structure. However, the improvement is only useful for a 100% modulation depth and comes with increased complexity, so a simpler unpatterned multilayer structures as in Fig. 2g is in general the best solution (see Supplementary Methods for designs close to the optimal performance).

Here, we have just confirmed that it is possible to design modulators with performances closely approaching the theoretical limit. Therefore, because the upper bound itself depends only on the conductivity of graphene, we can predict a priori the performance that optimized graphene modulators will achieve as a function of frequency and all other parameters that influence graphene conductivity. This prediction provides crucial ‘roadmap’ information regarding the applicability of graphene modulators at different wavelengths and the required graphene quality to achieve a given desired performance. Figure 3 presents the theoretical upper bound as a function of different parameters influencing the conductivity of graphene in the two states of the modulator (namely \( \gamma_M(f, T, \mu_{BA}, \mu_{BB}, \tau) \), where \( f \) is the frequency, \( T \) the temperature, \( \mu_{BA} \) and \( \mu_{BB} \) are the chemical potentials in the two states and \( \tau \) is the carrier scattering time), leading to the following conclusions. First, the best modulators can be designed between 10 and 100 THz and, evidently, for larger dynamic variations of the chemical potential. The performance sharply decreases at shorter wavelengths due to the well-known universal conductivity of graphene at optical frequencies (\( \mu_{cA} = 0.1 eV, \mu_{cB} = 0.8 eV \) and \( \tau = 66 fs \)).

In all plots, the quantities that are not swept or otherwise specified have values of \( f = 1 THz, T = 300 K, \mu_{cA} = 0.1 eV, \mu_{cB} = 0.8 eV \) and \( \tau = 66 fs \). a-d, Frequency dependence of \( \gamma_M \) for several values of temperature (a), \( \mu_{cA} \) (b), \( \mu_{cB} \) (c) and \( \tau \) (d). e, Parametric level curves of \( \gamma_M \) for different values of \( \mu_{cA} \) and \( \mu_{cB} \).
Non-reciprocal isolation

The second major class of graphene passive photonic devices is based on the presence of a magnetostatic field bias and the resulting off-diagonal terms in the conductivity tensor of graphene \( \sigma \). This class includes Kerr and Faraday polarization rotators, as well as isolators, which are non-reciprocal devices. These devices are considered in the remainder of this Article. A procedure similar to that used for the modulators can be followed for the mathematical derivation, its verification and the practical exploitation of the results. Accordingly, only the key methodology difference and practical results are reported here.

Non-reciprocal isolators display different transmission coefficients depending on the direction of wave propagation and can be completely described by a \( 2 \times 2 \) scattering matrix:

\[
S = \begin{pmatrix}
S_{11} & S_{12} \\
S_{21} & S_{22}
\end{pmatrix}
\]

Assuming, without loss of generality, that \( |S_{12}| \geq |S_{21}| \) and observing that here \( S_{11} = S_{22} = S \), the following ‘isolation inequality’ can be derived (see Supplementary Methods for the complete derivation):

\[
\gamma_{\text{isol}}(|S_{12}|, |S_{21}|) = \frac{(|S_{12}| - |S_{21}|)^2}{(1 - |S_{21}|^2)(1 - |S_{12}|^2)} \leq \gamma_{\text{NR}}
\]

where \( \gamma_{\text{NR}} \) is given by

\[
\gamma_{\text{NR}} = \frac{|\sigma_2|}{\text{Re}^2(\sigma_4) - \text{Im}^2(\sigma_0)} \quad \text{with} \quad \sigma = \begin{pmatrix} \sigma_4 \\ -\sigma_0 \end{pmatrix}
\]

Notice that using the approximate Drude–Lorentz model, expression (8) becomes

\[
\gamma_{\text{NR}} = \left( \frac{\omega_c \gamma}{\mu B_0} \right)^2 = \left( B_0 e \gamma T / \mu \right)^2
\]

where \( \mu \) is the carrier mobility, \( \omega_c \) is the cyclotron frequency, \( \gamma \) is the Fermi velocity, \( e \) is the elementary charge and \( B_0 \) is the magnetostatic biasing.

Similar to the case of the modulator (expression (2)), the left side of inequality (7) is an intrinsic figure of merit of the isolator and is written as \( \gamma_{\text{isol}} \). The right side is the graphene non-reciprocity upper bound \( \gamma_{\text{NR}} \), which now takes the form in expression (8). Figure 4a,b represent the isolation inequality and the ideal and best possible optima. The isolation is defined as \( I = |S_{12}|/|S_{21}| \). Figure 4c presents the variation in the best possible trade-off between isolation and insertion loss for different values of \( \gamma_{\text{NR}} \). The minimum insertion loss for an isolator having perfect isolation \( (I \rightarrow \infty) \) is given by

\[
|S_{12}| \leq \sqrt{\gamma_{\text{NR}}/(1 + \gamma_{\text{NR}})}
\]

Figure 4d–f illustrates that the best possible performance can be reached with simple planar structures encapsulated between two polarizers. In the simplest case of a graphene sheet placed between two dielectric slabs (Fig. 4d), optimality is achieved everywhere except for a small degradation when perfect isolation is required \( (I \rightarrow \infty) \). Increasing the number of dielectric layers between the graphene and polarizers does not solve this issue (Fig. 4e). However, a structure composed of three dielectric slabs and two graphene sheets in an alternating pattern can reach optimal performances with moderate complexity (Fig. 4f). Unlike additional dielectric layers, a second graphene layer allows for decorrelation rotation and loss in the system, approaching the theoretical upper bound.

As done earlier for modulators, we now study the ultimate performance that optimally designed isolators can achieve as a function
of graphene parameters, via $\gamma_{NR}(f, T, \mu_C, B_0, \tau)$. Figure 5a illustrates that the optimal performance improves with larger magnetostatic biasing, as expected. A less obvious observation is the fact that isolators perform better at low $\mu_C$. This finding can also be inferred by inspection of expression (9) and can be explained semiclassically by observing that the effective mass of the carriers decreases (or similarly, the mobility increases) for low $\mu_C$. Thus, the bending of their trajectories due to the magnetic field increases (higher cyclotron frequency). Figure 5b illustrates that temperature is influential at low chemical potentials, where the presence of thermal carriers of both polarities degrades the performance. High values of $\tau$, that is, high graphene quality, can lead to very high $\gamma_{NR}$, as shown in Fig. 5c. Figure 5c,d illustrates that performance is relatively frequency invariant until the mid-infrared region. This can be explained by the independence of $\gamma_{NR}$ on the imaginary part of $\sigma_T$ in expression (8). This behaviour contrasts with that of $\gamma_C$ in the modulation case, which degrades significantly towards low-terahertz and microwave frequencies (Fig. 3). Similarly to the modulator case, impedance matching at both ports is essential to achieve optimal performance.

**Kerr rotation**

An important effect associated with non-reciprocal surfaces is magneto-optical Kerr rotation, which is a property of a linearly polarized wave upon reflection at a surface. The following Kerr rotation inequality is obtained (see Supplementary Methods):

$$\gamma_{Kerr}(M, \varphi) \triangleq \frac{|2M \sin \varphi |^2}{(1 - M^2)} \leq \gamma_{NR}$$

(10)

where $\varphi$ is the Kerr rotation angle, $M$ is the magnitude of the reflected linear polarization and $\gamma_{NR}$ is given by expression (8). $M$ represents the magnitude of the major ellipse axis when the polarization of the reflected wave is elliptical. The inequality is graphically represented in Fig. 6a–c using the same conventions as in Figs 2 and 4. The 90° Kerr rotator is particularly important because it acts as a gyrator between vertically and horizontally polarized waves. The ideal and best possible Kerr rotators are identified by green markers in the figures.

Figure 6d presents the Kerr rotation of uniform graphene on a metal-backed substrate. Although optimal performance is obtained in a small region, the lack of degrees of freedom again prevents optimal performance over the full optimal frontier. The insertion of a single superstrate of a dielectric provides an additional degree of freedom but has no particular effect on the performance (Fig. 6e). However, two different superstrates allow for greatly enhanced Kerr rotations thanks to Fabry–Pérot resonances (Fig. 6f), leading to optimal performances in a technologically simple structure. This enhancement is also in agreement with a similar effect reported for Faraday rotation29,30,39. Finally, a significant number of devices obtained by randomized combinations of the different strategies of Fig. 6d–f were also simulated, all satisfying the bound expressed in (10). The detailed parameters and results of the near-optimal designs are shown in the Supplementary Information.

**Conclusions**

The performance of graphene-based modulators and non-reciprocal devices is bounded by absolute upper limits, which solely depend on the conductivity of graphene. This relationship allows the ultimate performance that will be achieved by such devices to be predicted as a function of frequency and the other parameters that influence graphene conductivity. Simple technological implementations
allow for very close approaches to the upper limit for the metasurface implementation of an amplitude modulator in reflection and transmission, as well as Kerr rotators and isolators. The observed influence of graphene parameters on the upper bounds, as well as the device topologies allowed to approach them, can be confirmed by physical insight. The developed theory applies to any passive linear structure that can be described in terms of a scattering matrix and can thus be extended to graphene guided devices and other 2DEGs. The developed methodology and practical results thereby obtained are believed to constitute an essential milestone towards the optimal operation of numerous future photonic devices. It is nevertheless clear that the practical design and implementation of any such devices will also entail considering parameters other than those bounded by the present theory, such as speed, footprint or cost.

Methods
Mathematical derivations. The theoretical bounds are derived in two steps. First, a general scattering bound based on a generic scattering matrix is demonstrated using a proof inspired by a previous formulation for three-dimensional materials used in guided devices\(^1\). Then, the relevant inequalities for actual graphene modulators, isolators and Kerr rotators are obtained by algebraically manipulating the general bound. See the Supplementary Methods for more information and a complete proof of the different graphene device inequalities.

Numerical simulations. The simulations of biperiodic graphene structures have been performed using a periodic method of the moment electromagnetic solver. See the Supplementary Methods for more information. Graphene conductivity is evaluated using the Kubo formalism\(^3\).

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Figure 6 | Performances of magneto-optical Kerr rotators. a. Graphical representation of the Kerr rotation inequality on the Cartesian plane (M, φ) (here, γNR = 0.6 is used as an example). The squares represent ideal 90° rotators and the circles denote the best possible 90° rotators. Forbidden areas (cyan) are delimited by the boundary curve, where γKerr = γNR. b. As in a, with the major axis expressed in dB. c. Theoretical bound curve for different values of γNR. d–f. Random simulations for different device topologies. Each red point represents a single simulated device. The frequency considered is 1 THz and the graphene parameters are T = 3 K, μC = 0.2 eV, B0 = 4 T and τ = 66 fs (γNR = 1.78). The topology shown in f reaches optimal performance values for every rotation in the range 0°–90°.
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Author contributions
M.T. and J.P.-C. conceived the idea of the bounds on the performance of graphene devices, as well as their practical exploitation (with comments from J.R.M.). M.T. developed the detailed mathematics of the theoretical bounds and A.F. developed the numerical electromagnetic solver. M.T. performed the simulations. M.T. and J.P.-C. wrote the manuscript (with comments from A.F.). J.P.-C. led the project.

Additional information
Supplementary information is available in the online version of the paper. Reprints and permissions information is available online at www.nature.com/reprints. Correspondence and requests for materials should be addressed to J.P.-C.

Competing financial interests
The authors declare no competing financial interests.