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Article

# Integrated Waveguide and microring Polarizers Incorporating Reduced 2D Graphene Oxide Thin Films

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Abstract Optical polarizers, which selectively transmit light with specific polarization states, are essential components in modern optical systems. Here, we experimentally demonstrate integrated waveguide and microring resonator (MRR) polarizers incorporating reduced graphene oxide (rGO). 2D graphene oxide (GO) films are integrated onto silicon photonic devices with precise control over their thicknesses and sizes, followed by GO reduction via two different methods including uniform thermal reduction and localized photothermal reduction. We measure devices with different lengths, thicknesses, and reduction degrees of the GO films. The results show that the devices with rGO exhibit better performance than those with GO, achieving a polarization-dependent loss of ~47 dB and a polarization extinction ratio of ~16 dB for the hybrid waveguides and MRRs with rGO, respectively. By fitting the experimental results with theory, it is found that rGO exhibits more significant anisotropy in loss, with an anisotropy ratio over 4 times that of GO. In addition, rGO shows higher thermal stability and greater robustness to photothermal reduction than GO. These results highlight the strong potential of rGO films for implementing high-performance polarization selective devices in integrated photonic platforms.

Keywords: integrated optics; 2D materials; graphene oxide; optical polarizers

# Introduction

In modern optical systems, controlling light polarization is of fundamental importance and underpins a variety of advanced optical technologies [1–3]. Optical polarizers, which allow the transmission of light with a specific polarization orientation and block light with the orthogonal polarization, are key components for controlling light polarization [4]. To date, a wide range of optical polarizers have been implemented based on refractive prisms [5,6], birefringent crystals [7,8], fiber components [9,10], and integrated photonic devices [11–13]. However, these polarizers based on bulk materials typically face challenges in achieving effective polarization selection across broad wavelength ranges [1,14,15], despite the growing demand for broadband optical polarizers driven by rapid advancements in photonic technologies and systems [16,17].

Since the groundbreaking isolation of graphene in 2004 [18], there has been an enormous surge in research on two-dimensional (2D) materials with atomic-scale thicknesses, which exhibit many extraordinary properties unattainable for conventional bulk materials [19–21]. With highly anisotropic properties across wide optical bands, 2D materials such as graphene [15,22,23], graphene oxide (GO) [24–26], and transition metal dichalcogenides (TMDCs) [27–29] have been incorporated into bulk material device platforms to realize high-performance optical polarizers. As a common derivative of graphene, GO has facile solution-based synthesis processes and transfer-free film coating with precise control over the film thickness, making it well-suited for large-scale on-chip integration to implement hybrid devices [30–32]. In addition, the properties of GO can be easily

changed through various reduction methods, providing a high flexibility to optimize the performance of hybrid devices [33–35].

Previously, we demonstrated integrated optical polarizers incorporating 2D GO films on both doped silica and silicon device platforms [24,25]. In this work, we integrate 2D reduced GO (rGO) films onto integrated photonic devices to realize waveguide and MRR polarizers with improved performance. We fabricate hybrid integrated devices with precise control over the thicknesses and lengths of the GO films. The reduction of GO is realized by using two methods: uniform thermal reduction, achieved by heating the integrated chip on a hot plate, and localized photothermal reduction, induced by high power of input light. Detailed measurements are carried out for devices with different lengths, thicknesses, and reduction degrees of the GO films. The results show that the devices with rGO exhibit better polarization selectivity than comparable devices with GO. Up to ~47-dB polarization-dependent loss (*PDL*) and ~16-dB polarization extinction ratio (*PER*) are achieved for the hybrid waveguides and MRRs with rGO, respectively. By fitting the experimental results with theoretical simulations, we find that rGO exhibits significantly improved loss anisotropy, with an anisotropy ratio more than 4 times that of GO. Compared to GO, rGO also exhibits stronger thermal stability and lower sensitivity to photothermal reduction. These results verify the effectiveness of onchip integration of 2D rGO films to realize high performance optical polarizers.

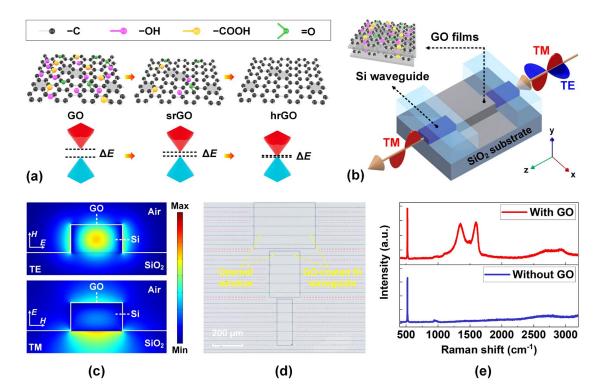
## **Results and Discussion**

Device Design and Fabrication

As an oxidized derivative of graphene, GO consists of carbon networks decorated with various oxygen functional groups (OFGs), such as hydroxyl, epoxide, carbonyl, and carboxylic groups [33,36,37]. **Figure 1(a)** illustrates the atomic structures and bandgaps of graphene oxide (GO), semi-reduced GO (srGO), and highly reduced GO (hrGO). Due to the presence of isolated  $sp^2$  domains within the  $sp^3$  carbon-oxygen matrix, unreduced GO is a dielectric material with an opened bandgap of ~2.1 – 3.6 eV [36,38]. This bandgap is larger than the energy of two photons at ~1550 nm (*i.e.*, ~1.6 eV), allowing for both low linear light absorption and two-photon absorption at infrared wavelengths. The reduction of GO breaks the chemical bonds between the OFGs and the carbon network. Compared to pristine GO, reduced GO (rGO) has a decreased bandgap [39,40], resulting in alterations to material properties such as refractive index, optical absorption, and electrical conductivity. Practically, the reduction of GO films can be achieved by using different methods, such as thermal reduction, chemical reduction, and photoreduction [41–43]. As the degree of reduction increases, the fraction of  $sp^2$ -hybridized carbon atoms increases. For hrGO with minimal remaining OFGs, the bandgap and material properties closely resemble those of graphene, which exhibits a zero bandgap and metallic behaviour [44,45].

Figure 1(b) shows the schematic of an integrated waveguide polarizer based on a silicon photonic waveguide coated with a 2D GO film. The cross section of the silicon waveguide is 400 nm × 220 nm. The GO film has a thickness of 4 nm, which corresponds to 2 layers of GO fabricated using a solution-based self-assembly method (as discussed later in this section). Figure 1(c) shows the corresponding transverse electric (TE) and transverse magnetic (TM) mode profiles for the hybrid waveguide in Figure 1(b), which were simulated using a commercial mode-solving software (COMSOL Multiphysics). The TE- and TM polarized effective indices (at 1550 nm) for the hybrid waveguide were  $2.093 + 1.244 \times 10^{-4}i$  and  $2.093 + 4.784 \times 10^{-5}i$ , respectively. In our simulation, the refractive index (n) and extinction coefficient (k) of GO for TE polarization were  $n_{TE} = \sim 1.969$  and  $k_{TE}$ = ~0.0098, respectively. For TM polarization, the corresponding values were  $n_{TM}$  = ~ 1.898 and  $k_{TM}$  = ~0.0022. The n, k values were obtained from our previous measurements in Ref. [34] and the experimental results in following sections. The large difference between  $k_{TE}$  and  $k_{TM}$  is due to the significant anisotropy in the light absorption of 2D GO films, where the in-plane absorption is much stronger than the out-of-plane absorption [15,24]. As a result, TE-polarized (in-plane) light experiences a higher loss compared to TM-polarized (out-of-plane) light as it propagates through the hybrid waveguide, allowing the hybrid waveguide to function as a TM-pass optical polarizer.





**Figure 1.** I (a) Schematics of atomic structures and bandgaps of graphene oxide (GO), semi-reduced GO (srGO), and highly reduced GO (hrGO). (b) Schematic illustration of a GO-coated silicon waveguide as an optical polarizer. Inset illustrates the layered GO film structure fabricated by self-assembly. (c) TE and TM mode profiles for the hybrid waveguide with 2 layers of GO. (d) Microscopic image of the fabricated devices on a GO-coated silicon-on-insulator (SOI) chip. (e) Measured Raman spectra of the SOI chip in (d) without GO and with 2 layers of GO.

**Figure 1(d)** shows a microscopic image of the fabricated devices on a silicon-on-insulator (SOI) chip. The silicon waveguides were patterned via 248-nm deep ultraviolet lithography followed by inductively coupled plasma etching. After this, a 1.5-μm-thick silica layer was deposited by plasma enhanced chemical vapor deposition to cover the SOI chip as an upper cladding. To enable the interaction between the GO films and the evanescent field from the silicon waveguides, windows were opened on the silica upper cladding to allow the coating of 2D GO films onto the silicon waveguides. In our fabricated devices, the length of all silicon waveguides was ~3.0 mm, and the lengths of the opened windows ranged between ~0.1 mm and ~2.2 mm.

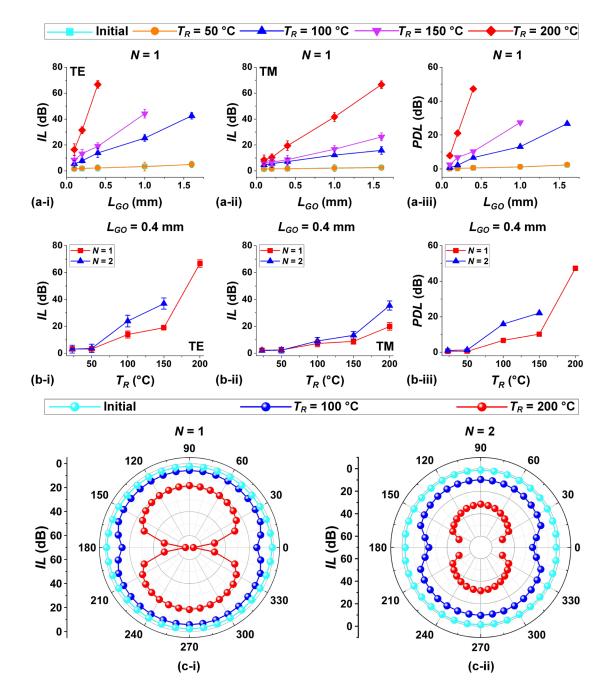
The coating of the GO film, with a thickness of ~2 nm per layer, was realized by using a solution-based self-assembly method that enabled transfer-free and layer-by-layer film coating [24,30]. During the coating process, a multilayered film structure composed of alternating GO layers and oppositely charged polymer layers was constructed, with the GO layers formed through the self-assembly of exfoliated GO nanoflakes. As compared with the complicated film transfer methods employed for other 2D materials such as graphene and transition-metal dichalcogenides (TMDCs) [46,47], this coating method allows for transfer-free film coating with precise control of the film thickness. In addition, it enables conformal coating of 2D GO films onto silicon waveguides with minimal air gaps [48]. In **Figure 1(d)**, the coated GO film shows high transmittance and good morphology without any noticeable wrinkling or stretching, confirming excellent film attachment onto the silicon waveguides.

**Figure 1(e)** shows the measured Raman spectra of the SOI chip in **Figure 1(d)** before and after coating 2 layers of GO, which were measured using a  $\sim$ 514-nm pump laser. The GO films had a thickness of  $\sim$ 4.0 nm, which was characterized using atomic force microscopy measurement. In the Raman spectrum for the GO-coated chip, the existence of the representative D ( $\sim$ 1345 cm $^{-1}$ ) and G ( $\sim$ 1590 cm $^{-1}$ ) peaks [49,50] provides evidence for successful on-chip integration of 2D GO film.

In **Figure 2**, we show the measured insertion losses (IL's) of our fabricated devices for input continuous-wave (CW) light in different polarization states. We measured devices with different GO film lengths ( $L_{GO}$ ) and GO layer numbers (N), after being subjected to various reduction temperatures ( $T_R$ ) ranging from ~50 to ~200 °C. Here we chose the temperature range of  $T_R \le 200$  °C because the polymer layers in the self-assembled films cannot withstand temperatures beyond this range. For all the devices, the cross section of the uncoated silicon waveguides was ~400 nm × 220 nm. In our measurements, lensed fibers were employed to butt couple a CW light at ~1550 nm into and out of the fabricated devices with inverse-taper couplers at both ends. The fiber-to-chip coupling loss was ~5 dB per facet. For comparison, we measured the IL's by using the same input power of ~0 dBm. Unless otherwise specified, the input power ( $P_{In}$ ) and IL in our following discussion refers to those measured after excluding the fiber-to-chip coupling loss.

**Figures 2(a-i)** and **2(a-ii)** show the measured TE- and TM-polarized IL versus Lco for the hybrid waveguides with 1 layer of GO (N = 1), respectively. Before the IL measurement, the SOI chip was heated on a hot plate for 15 minutes at various temperatures  $T_R$ . For comparison, the results corresponding to different  $T_R$  are plotted together with those measured at room temperature prior to heating (which are labeled as 'initial'). In each figure, the data points represent the average values from measurements of three duplicate devices, and the error bars reflect the variations across different samples. We do not show results for IL > 70 dB in these and subsequent figures because it exceeds the detection range of the optical power meter used in our measurements.





**Figure 2.** I (a) Measured (i) TE- and (ii) TM-polarized insertion loss (*IL*) versus GO coating length (LGO) for the hybrid waveguides coated with a monolayer GO film (N =1) after the chip was heated at various temperatures  $T_R$ . (iii) shows the polarization dependent loss (PDL) calculated from (i) and (ii). (b) Measured (i) TE- and (ii) TM- polarized IL versus  $T_R$  for the hybrid waveguides with 1–2 layers of GO (N =1, 2). (iii) shows the PDL calculated from (i) and (ii). (c) Polar diagrams for the measured IL of devices with different GO layer numbers of (i) N = 1 and (ii) N = 2 after the chip was heated at various temperatures  $T_R$ . The polar angle represents the angle between the input polarization plane and the substrate. In (a) – (c), the input continuous-wave (CW) power and wavelength were ~0 dBm and ~1550 nm, respectively. In (a) and (b), the data points illustrate the average of measurements on three duplicate devices and the error bars depict the variations among the different devices. In (b) and (c), LGO = ~0.4 mm.

In **Figure 2(a-i)** and **2(a-ii)**, the *IL* increases with  $L_{GO}$  for both polarizations, with the TE polarization exhibiting a more dramatic increase than the TM polarization. At  $T_R = ~50$  °C, both the TE- and TM-polarized *IL* did not show any significant difference as compared with that at the initial

unheated status. These results suggest that there were no significant changes in the GO film properties at  $T_R = ~50$  °C, indicating that the reduction of GO did not occur at this temperature. In contrast, when  $T_R \ge ~100$  °C, the IL increases with  $T_R$  for both polarizations. This reflects the loss increase due to the reduction of GO at high temperatures. As  $T_R$  increases, a higher degree of reduction was achieved, leading to a more significant increase in the IL.

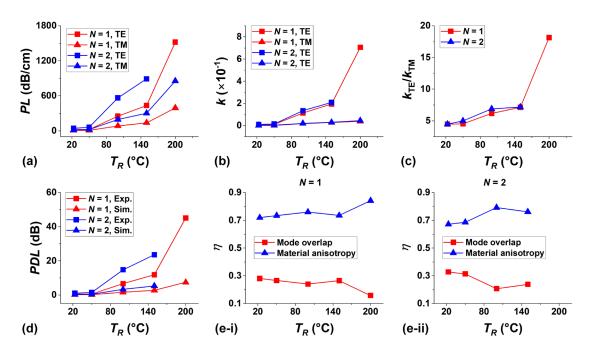
**Figure 2(a-iii)** shows the corresponding *PDL* (dB) obtained by subtracting the TM-polarized *IL* in **Figure 2(a-ii)** from the TE-polarized *IL* in **Figure 2(a-i)**. The *PDL* increases with  $L_{GO}$ , and it also increases with  $T_R$  when  $T_R \ge 100$  °C. For the device with  $L_{GO} = \sim 0.4$  mm and at  $T_R = \sim 200$  °C, a maximum *PDL* value of ~47 dB was obtained. In contrast, the *PDL* exhibited no significant difference between the initial status and at  $T_R = 50$  °C, achieving a *PDL* of ~1 dB for the devices with the same  $L_{GO}$ . By further increasing  $L_{GO}$  for devices with hrGO (*i.e.*, at  $T_R = \sim 150$  and  $\sim 200$  °C), a *PDL* exceeding ~47 dB can be achieved (not shown in this figure due to limited detection range of the optical power meter), at the expense of a higher additional *IL* induced by GO.

**Figures 2(b-i)** and **2(b-ii)** show the measured IL versus  $T_R$  for TE and TM polarizations, respectively. Here we show the results for the hybrid waveguides with 1 and 2 layers of GO. For comparison, all the waveguides had the same  $L_{GO} = \sim 0.4$  mm. Both the TE- and TM-polarized IL remains unchanged when  $T_R \le \sim 50$  °C. For  $T_R \ge \sim 100$  °C, the IL for TE polarization shows a more significant increase with  $T_R$  than that for TM polarization, following a trend similar to that in **Figure 2(a-i)** and **2(a-ii)**. Compared to the devices with 1 layer of GO (N = 1), higher IL was achieved for the devices with 2 layers of GO (N = 2), reflecting a higher loss induced by a thicker GO film. **Figure 2(b-iii)** shows the corresponding PDL extracted from **Figure 2(b-i)** and **2(b-ii)**, where higher PDL values were also achieved for the devices with thicker GO films. For the 2-layer device at  $T_R = \sim 150$  °C, the PDL was  $\sim 24$  dB, in contrast to  $\sim 10$  dB for a comparable 1-layer device. At  $T_R = \sim 200$  °C, it is anticipated that the 2-layer device can achieve a high PDL exceeding 60 dB, we were not able to measure it due to the limited detection range of our optical power meter.

**Figures 2(c-i)** and **2(c-ii)** show the polar diagrams for the measured IL of devices with 1 and 2 layers of GO (N = 1, 2), respectively. In each figure, we plot three curves corresponding to different  $T_R$ . For comparison, all the hybrid waveguides had the same  $L_{GO}$  = ~0.4 mm. The polar diagrams show variations in IL values across different polarization angles, which reflects the polarization selectivity of the hybrid waveguides. For the hybrid waveguides with 1 layer of GO, the PDL values at the initial unheated status,  $T_R$  = ~100 °C, and  $T_R$  = ~200 °C are ~1 dB, ~7 dB, and ~47 dB, respectively. These results indicate that the polarization selectivity is improved as the degree of GO reduction increases. At  $T_R$  = ~100 °C, the PDL values for N = 1 and N = 2 are ~7 dB and ~15 dB, respectively. This reflects that improved polarization selectivity can also be achieved for hybrid devices with thicker GO films.

## Analysis of GO Film Properties

Based on the measured results in **Figure 2**, we further analyze the properties of 2D GO films by fitting the experimental results with theoretical simulations. **Figure 3(a)** shows the waveguide propagation loss (PL) versus  $T_R$  for the hybrid devices with 1 and 2 layers of GO (i.e., N = 1, 2), which was extracted from the measured IL in **Figure 2(b-i)** and **2(b-ii)**. The excess propagation loss (EPL) induced by the GO films was further calculated by excluding the PL for the uncoated silicon waveguide (i.e., ~3.4 dB/cm for TE polarization and ~3.1 dB/cm for TM polarization). The TE-polarized EPL induced by 1 layer of rGO at  $T_R$  = 200°C was ~1520 dB/cm, in contrast to ~20 dB/cm for 1 layer of unreduced GO at the initial status. This reflects the substantial increase in loss for highly reduced GO films. We also note the value of ~1520 dB/cm is lower than the typical values of EPL induced by monolayer graphene (i.e., ~2000 dB/cm [51,52]). This suggests that, although the GO film was highly reduced at  $T_R$  = 200°C, it was not yet fully reduced.



**Figure 3.** I (a) TE- and TM-polarized waveguide propagation loss (PL) versus  $T_R$  for the hybrid waveguides with 1 and 2 layers of GO (N = 1, 2). (b) Extinction coefficients (k's) of 2D GO films versus  $T_R$  obtained by fitting the results in (a) with optical mode simulations. (c) Anisotropy ratios of k values for TE and TM polarizations ( $k_{TE} / k_{TM}$ ) extracted from (b). (d) Measured (Exp.) and simulated (Sim.) PDL versus  $T_R$  for the hybrid waveguides with 1-2 layers of GO (N = 1, 2). The simulated PDL values were obtained by using the same k value for both TE and TM polarizations. (e) Fractional contributions ( $\eta$ 's) to the overall PDL from polarization-dependent mode overlap and material loss anisotropy, which were extracted from (d). (i) and (ii) show the results for N = 1 and 2, respectively.

**Figure 3(b)** shows the extinction coefficients (k's) of 2D GO films obtained by fitting the results in **Figure 3(a)** with optical mode simulations (at 1550 nm) for the hybrid waveguides. For 1 layer of rGO at  $T_R = \sim 200$  °C, the value of k is  $\sim 0.7057$  for TE polarization, which is about 75 times that of comparable unreduced GO. For all different N and  $T_R$ , the GO films exhibited higher values of k for TE polarization than TM polarization, reflecting the intrinsic anisotropy in the loss of 2D GO films. We also note that, for both polarizations, slightly higher k values were obtained with thicker GO films. This is likely due to the increased scattering loss caused by film unevenness and accumulation of imperfect contact between adjacent layers in thicker films.

In **Figure 3(c)**, we further plot the anisotropy ratios defined as the ratios of the corresponding k values for TE- and TM- polarizations ( $k_{TE} / k_{TM}$ ) in **Figure 3(b)**. Compared to unreduced GO, higher values of the anisotropy ratio are obtained for rGO at  $T_R \ge 100$  °C, with the anisotropy ratio increasing as the degree of reduction increases. For 1 layer of rGO at  $T_R = ~200$  °C, the anisotropy ratio is ~18 - over 4 times higher than that of 1 layer of unreduced GO. These results highlight an interesting phenomenon that the 2D GO films exhibit more significant loss anisotropy as the degree of reduction increases. This is probably because the reduction of GO leads to the removal of OFGs and hence a decrease in the film thickness. We also note that in Ref. [15] monolayer graphene (with a thickness of ~0.5 nm, in contrast to ~2 nm for monolayer unreduced GO) exhibits a higher anisotropy ratio of ~30. This suggests that highly reduced GO exhibits loss anisotropy close to that of graphene.

Compared to GO, the higher anisotropy ratio of rGO leads to more significant difference between the absorption of in-plane and out-of-plane light waves, making it better suited for implementing optical polarizers with high polarization selectivity. In addition, unlike the intricate film transfer methods needed for on-chip integration of graphene, GO offers advantages for large-scale manufacturing due to its facile synthesis processes and transfer-free film coating. Hybrid integrated devices with rGO can be readily fabricated by further reducing GO within the hybrid

devices. Therefore, the GO fabrication techniques can be leveraged for large-scale manufacturing of hybrid integrated devices with rGO.

In **Figure 3(c)**, slightly increased anisotropy ratio is also achieved for thicker GO films. For unreduced GO, the anisotropy ratios are ~4.4 and ~4.5 for the films including 1 and 2 layers of GO, respectively. For 1 layer of rGO at  $T_R = \sim 100$  °C, the anisotropy ratio is ~6, in contrast to ~7 for 2 layers of rGO at that same  $T_R$ . These results reflect that the thicker film exhibits more significant anisotropy in loss for both GO and rGO.

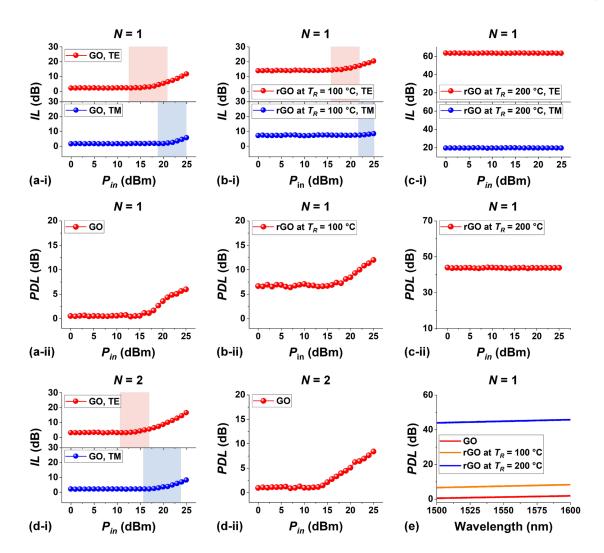
In **Figure 3(d)**, we compare the measured *PDL* values with those obtained from optical mode simulations. We show the results at different  $T_R$  for the hybrid devices with 1 and 2 layers of GO. In our simulations, we assumed that the GO films were isotropic with the same values of k (*i.e.*,  $k_{TE}$  in **Figure 3(b)**) for both TE and TM polarizations. As a result, the simulated *PDL* values represent the polarization selectivity caused by the polarization-dependent mode overlap with the GO films, and the variation between the simulated and measured *PDL* values characterizes the extra polarization selectivity enabled by the loss anisotropy of 2D GO films. For all different  $T_R$  and N, the simulated *PDL*'s exhibited positive values, reflecting that the polarization-dependent mode overlap with GO contributes to the overall *PDL*.

In **Figure 3(e-i)** and **3(e-ii)**, we further calculated the fractional contributions to the overall *PDL* from the polarization-dependent mode overlap and the material loss anisotropy (where the sum of these two fractions equals 1). For all different  $T_R$  and N, over 65% of the polarization selectivity is attributed to the loss anisotropy of 2D GO films. This highlights its dominance in enabling the functionality of the optical polarizer. It is also interesting to note that the fractional contribution from the loss anisotropy increases as  $T_R$  increases. This is mainly due to the fact there is more significant loss anisotropy for rGO, as we discussed in **Figure 3(c)**.

### Dependence on Input Power and Wavelength

For the experiments in **Figure 2**, the *IL* was measured at a low input CW power of  $P_{in} = \sim 0$  dBm, to ensure that the GO films remained unaffected by photothermal reduction induced by the CW light. In **Figure 4**, we further increase the input CW power  $P_{in}$  to induce photothermal reduction of GO and characterize the changes in the polarization selectivity of the hybrid waveguides. Compared to GO reduction caused by heating the entire chip on a hot plate (as we did for the experiments in **Figure 2**), using input CW power can trigger localized photothermal reduction of GO in the hybrid waveguides, along with dynamic changes in the GO film properties.

**Figure 4(a-i)** shows the measured IL versus  $P_{in}$  for the hybrid waveguide with 1 layer of unreduced GO (*i.e.*, we directly measured the IL without heating the GO-coated chip on a hot plate, unlike what we did later in **Figure 4(b)** and **4(c)**). The GO film length in the hybrid waveguide was  $L_{GO} = \sim 0.4$  mm, and the wavelength of the input CW light was  $\sim 1550$  nm. We measured the IL for both TE and TM polarizations, and the results were recorded only when a steady thermal equilibrium state with stable output power was achieved. We chose an input power range of  $P_{in} \leq \sim 25$  dBm because the polymer layers in the self-assembled films cannot withstand input powers beyond this range.



**Figure 4.** I (**a**) Measured (**i**) TE- and TM-polarized *IL* and (**ii**) calculated *PDL* versus input power ( $P_{in}$ ) for the hybrid waveguide with 1 layer of GO. (**b**) – (**c**) Measured (**i**) TE- and TM-polarized *IL* and (**ii**) calculated *PDL* versus  $P_{in}$  for the hybrid waveguide with 1 layer of rGO after heating at  $T_R = \sim 100$  °C and  $\sim 200$  °C, respectively. (**d**) Measured (**i**) TE- and TM-polarized *IL* and (**ii**) calculated *PDL* versus  $P_{in}$  for the hybrid waveguide with 2 layers of GO. In (**a**) – (**d**), the red and blue shaded areas in (**i**) indicate the power ranges associated with reversible GO reduction for TE and TM polarizations, respectively. (**e**) Measured *PDL* versus input CW wavelength for the hybrid waveguide with 1 layer of unreduced GO, rGO at  $T_R = \sim 100$  °C, and rGO at  $T_R = \sim 200$  °C. In (**a**) – (**e**), the GO film length was  $\sim 0.4$  mm. In (**a**) – (**d**), the input CW wavelength was  $\sim 1550$  nm. In (**e**), the input CW power was  $P_{in} = \sim 0.4$  Bm.

In **Figure 4(a-i)**, the TE-polarized IL remained constant at ~2 dB when  $P_{in} \le$  ~13 dBm, indicating that the GO film was not reduced in this power range. For  $P_{in} \ge$  ~13 dBm, the TE-polarized IL increased with  $P_{in}$ , and reached ~12 dB at  $P_{in} =$  ~25 dBm. This reflects that there was loss increase induced by photothermal reduction of GO at high light powers. We also note that the reduction of GO exhibited reversibility within a power range of ~13 dBm  $\le P_{in} \le$  ~21 dBm, as indicated by the red shaded area. In this power range, after turning off the high-power input and remeasuring the IL with a low input power of  $P_{in} =$  ~0 dBm, the IL returned to ~2 dB (*i.e.*, the IL for unreduced GO when  $P_{in} \le$  ~13 dBm). This reversibility indicates that the photothermally reduced GO was unstable in nature, which reverted to the unreduced status after cooling down in an oxygen-containing ambient. As  $P_{in}$  continued to rise above ~21 dBm, there was permanent increase in the IL after turning off the high-power input and remeasuring at  $P_{in} =$  ~0 dBm. This reflects that there was permanent reduction of GO induced by the high CW power in this range, where the chemical bonds between the OFGs and

the carbon network were irreversibly broken, resulting in a lasting alteration in GO's atomic structure and material properties. The photothermal reduction of GO in GO-Si waveguides is more significant as compared to that observed for GO-silicon nitride and GO-doped silica waveguides [31,53], mainly due to the stronger GO mode overlap in the GO-Si waveguides.

In **Figure 4(a-i)**, the TM-polarized IL increased when  $P_{in} \ge \sim 19$  dBm, reaching  $\sim 6$  dB at  $P_{in} = \sim 25$  dBm. Compared to TE polarization, the power threshold for initiating photothermal reduction of GO was higher for TM polarization. This can be attributed to weaker photo-thermal effects for TM polarization that result from lower absorption for out-of-plane light waves in the anisotropic 2D GO films. **Figure 4(a-ii)** shows the corresponding PDL versus  $P_{in}$  extracted from **Figure 2(a-i)**. The PDL exhibited no significant changes when  $P_{in} \le \sim 13$  dBm. However, when  $P_{in}$  exceeded  $\sim 13$  dBm, there was an obvious increase in the PDL as  $P_{in}$  increased. This indicates that the polarization selectivity was enhanced by increasing the input power.

**Figure 4(b)** and **4(c)** show the corresponding results for the hybrid waveguides with 1 layer of rGO after heating at  $T_R = \sim 100$  °C and  $\sim 200$  °C, respectively. For comparison, the GO film length was the same as that of the hybrid waveguide in **Figure 4(a)**. Before measuring the *IL*, the GO-coated chip was heated on a hot plate for 15 minutes, as we did in **Figure 2**. According to the results in **Figure 2**, the GO films in the hybrid waveguides were reduced after heating at  $T_R = \sim 100$  °C and  $\sim 200$  °C. For rGO at  $T_R = \sim 100$  °C in **Figure 4(b)**, loss increase induced by photothermal reduction was observed for TE polarization when  $P_{in} \geq \sim 16$  dBm. The power threshold of  $\sim 16$  dBm was higher than that for unreduced GO (*i.e.*,  $\sim 13$  dBm in **Figure 4(a)**). This indicates that unreduced GO was more easily reduced by the applied CW power, and higher power is required to trigger photothermal reduction of rGO.

For rGO at  $T_R = \sim 200$  °C in **Figure 4(c)**, increasing  $P_{in}$  did not result in any significant variations in the IL and PDL. These results further confirm that the photothermal reduction behaviour of GO becomes less obvious as the degree of reduction increases. According to Ref. [34], rGO exhibits higher thermal conductivity compared unreduced GO, and the thermal conductivity increases with the degree of reduction. The relatively high thermal conductivity of rGO leads to a lower heat accumulation efficiency, which in turn diminishes the photothermal effects and the power-dependent response. In addition to exhibiting a higher anisotropy ratio in **Figure 3(c)**, rGO shows better thermal stability and stronger immunity to photothermal reduction than GO, making it attractive for implementing optical polarizers operating at high temperatures and input powers.

**Figure 4(d)** shows the corresponding results for the hybrid waveguide with 2 layers of unreduced GO (*i.e.*, without heating the GO-coated chip on a hot plate). Loss increase induced by photothermal reduction was observed for TE polarization when  $P_{in} \ge -11$  dBm, and reversible GO reduction was observed when  $\sim 11$  dBm  $\leq P_{in} \leq \sim 17$  dBm. Compared to the results in **Figure 4(a)**, the hybrid waveguide with a thicker GO film exhibited a lower power threshold for initiating photothermal reduction and a smaller power range for reversible GO reduction. These reflect more significant photo-thermal effects in thicker GO films.

**Figure 4(e)** shows the measured *PDL* versus input CW wavelength for the hybrid waveguides with 1 layer of GO, rGO at  $T_R = \sim 100$  °C, and rGO at  $T_R = \sim 200$  °C. For comparison, the input CW power was kept the same as  $P_m = \sim 0$  dBm. For all three waveguides, the *PDL* exhibited a very small variation (< 1 dB) across the measured wavelength range of  $\sim 1500 - 1600$  nm. This reflects the broad operation bandwidth for these waveguide polarizers. We also note that there was a slight increase in the *PDL* as the wavelength increased, mainly due to a minor change in the mode overlap with GO induced by dispersion.

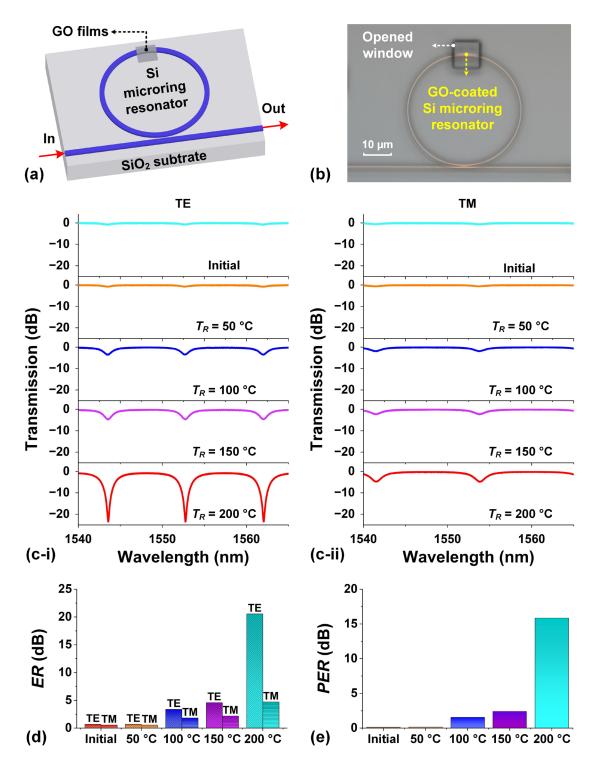
### Polarization-Selective Microring Resonators

In addition to waveguide polarizers, we also investigate polarization-selective MRRs by integrating 2D GO films onto silicon MRRs. **Figure 5(a)** shows the schematic of a silicon MRR coated with 1 layer of GO, and a microscopic image of the fabricated device is provided in **Figure 5(b)**. The silicon MRR had a radius of ~20  $\mu$ m, and the length of the opened windows (i.e., the length of the coated GO film) was ~10  $\mu$ m. The ring and the bus waveguide in the MRR had the same waveguide

cross-section of ~400 nm × 220 nm – identical to that for the waveguide polarizers in **Figure 2**. The hybrid MRR and the hybrid waveguides in **Figure2** were fabricated on the same SOI chip via the same processes.

In **Figure 5(c)**, we compare the TE- and TM- polarized transmission spectra for the hybrid MRRs with 1 layer of GO at different degrees of reduction. All the spectra were measured by scanning the wavelength of an input CW light with a power of  $P_{in} = \sim -10$  dBm (which did not induce any significant photo-thermal effects in the GO films). We first measured the device with unreduced GO in **Figure 5(b)** before heating it on a hot plate (the results are labeled as 'initial'). Then, we measured the same device after heating it on a hot plate at various temperatures  $T_R$  ranging from  $\sim 50$  to  $\sim 200$  °C (for 15 minutes, as we did in **Figure 2**).

**Figure 5(d)** shows the extinction ratios (*ER*'s) of the hybrid MRRs extracted from **Figure 5(c)**. As can be seen, the *ER* of the hybrid MRR after heating at  $T_R = ~50$  °C showed negligible difference as compared to that of the unheated MRR. This shows agreement with the results in **Figure 2** and provides further evidence that the reduction of GO did not occur at  $T_R = ~50$  °C. For  $T_R \ge 100$  °C, an increase in the *ER* was observed as  $T_R$  increased, particularly for TE polarization. This was because the uncoated silicon MRR we chose was over-coupled [54,55]. As  $T_R$  increased, the degree of reduction for GO also increased, leading to higher loss of the GO film. As the loss induced by GO increased, the difference between the round-trip loss and the coupling strength in the hybrid MRR became smaller, resulting in a higher *ER* (*i.e.*, more approaching the critical coupling condition [56]).

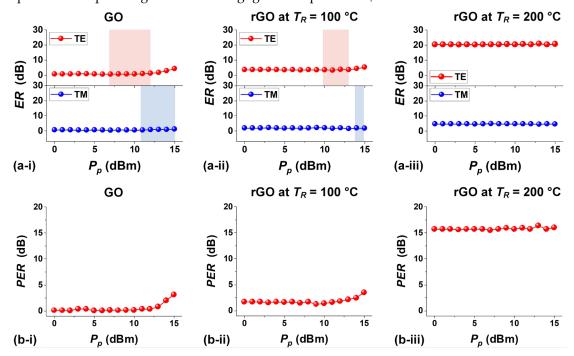


**Figure 5.** I (a) Schematic illustration of a GO-coated silicon microring resonator (MRR) as a polarization-selective MRR. (b) Microscopic image of a fabricated silicon MRR coated with 1 layer of unreduced GO. (c) Measured (i) TE- and (ii) TM-polarized transmission spectra of the hybrid MRR with 1 layer of GO at different degrees of reduction. The same hybrid MRR underwent heating at temperatures  $T_R$  ranging from ~50 to 200 °C prior to the measurement. The corresponding results measured at room temperature before heating (initial) are also shown for comparison. (d) Extinction ratios (*ER*'s) for the MRRs extracted from (c). (e) Polarization extinction ratios (*PER*'s) extracted from (d). In (c) – (e), the CW input power was  $P_{in} = \sim$ -10 dBm.

**Figure 5(e)** shows the *PER* obtained by subtracting the TM-polarized *ER* from the TE-polarized *ER* in **Figure 5(d)**. As can be seen, the hybrid MRR with unreduced GO exhibited a low polarization

selectivity, with its *PER* being less than ~1 dB. In contrast, The *PER* increased with  $T_R$  when  $T_R \ge 100$  °C. After heating at  $T_R = \sim 200$  °C, the hybrid MRR exhibited a high *PER* of ~16 dB, highlighting its excellent polarization selectivity.

In **Figure 6**, we characterize the power-dependent response for the polarization-selective MRRs in **Figure 5** by increasing the input CW power to induce photothermal reduction of GO. In **Figure 5**, we measured the MRRs' transmission spectra by using a single input CW light with a low power of  $P_{in} = \sim -10$  dBm. In **Figure 6**, we employed two CW inputs in our measurements. The first one with a power of  $P_p$  was employed as a pump injecting into one of the MRR's resonances near  $\sim 1550$  nm. The wavelength of this input CW light was slightly tuned around the resonance until a steady thermal equilibrium state with stable output power was achieved. After this, the second CW light, with a power of  $\sim -10$  dBm (*i.e.*, the same as that in **Figure 5**), was employed as a low-power probe to scan the MRR's transmission spectrum. Compared to directly using a high-power CW light to scan the spectrum, this approach would not induce significant asymmetry in the measured resonance spectral lineshape caused by optical bistability [57,58], thus allowing for a higher accuracy in characterizing the MRR's extinction ratio. In our measurements, the CW pump power  $P_p$  was  $\leq \sim 15$  dBm to prevent damages to the polymer layers in the self-assembled films. We also chose  $P_p \geq \sim 0$  dBm to ensure that the power of the probe light remained negligible compared to  $P_p$ .

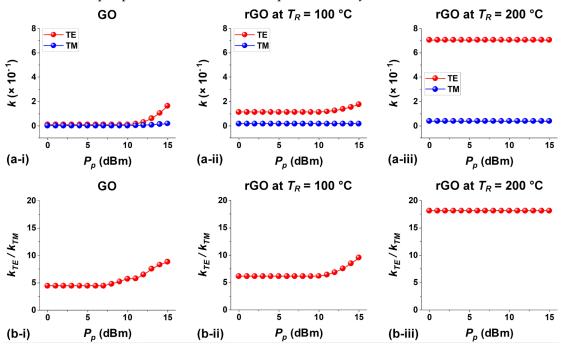


**Figure 6.** I (**a**) Measured TE- and TM-polarized *ER* versus input CW pump power  $P_p$  for the hybrid MRR with 1 layer of GO at different degrees of reduction. (**i**) – (**iii**) show the results measured for the same hybrid MRR with 1 layer of GO, rGO after heating at  $T_R = \sim 100$  °C, and rGO after heating at  $\sim 200$  °C, respectively. (**b**) *PER*'s extracted from (a). In (**a**) – (**b**), the red and blue shaded areas indicate the power ranges associated with reversible GO reduction for TE and TM polarizations, respectively.

In **Figure 6(a)**, we plot TE- and TM-polarized *ER* versus input CW pump power  $P_p$ . We first measured a hybrid MRR with unreduced GO. As shown in **Figure 6(a-i)**, the TE-polarized *ER* exhibited no significant variations when  $P_p \le \sim 7$  dBm. When  $P_p \ge \sim 7$  dBm, it increased with  $P_p$ , indicating that there was increased loss induced by localized photothermal reduction of GO. The power threshold of  $\sim 7$  dBm for the hybrid MRR was much lower than that for a comparable hybrid waveguide (*i.e.*,  $\sim 13$  dBm in **Figure 4(a)**), reflecting more significant photothermal effects in the hybrid MRR enabled by the resonance enhancement effect. Compared to TE polarization, a higher power threshold of  $\sim 11$  dB was observed for TM polarization, further indicating the anisotropy of the 2D GO film. For both polarizations, reversible GO reduction behaviour was also observed within specific power ranges – similar to the results in **Figure 4(a)**.

**Figures 6(a-ii)** and **6(a-iii)** show the corresponding results for the hybrid MRR with 1 layer of rGO after heating at  $T_R = \sim 100$  °C and  $\sim 200$  °C, respectively. For the device with rGO at  $T_R = \sim 100$  °C, the increase in ER caused by localized photothermal reduction of GO was observed when  $P_p \geq \sim 10$  dBm for TE polarization and  $P_p \geq \sim 14$  dBm for TM polarization. These power thresholds are higher than those in **Figure 6(a)** for the device with unreduced GO, further confirming that a higher CW power is needed to induce photothermal reduction of rGO. For the device with rGO at  $T_R = \sim 200$  °C, no significant variations in the ER were observed for both polarizations within the measured input pump power range. This highlights that the highly reduced GO exhibited even less noticeable photothermal reduction behaviour, showing agreement with the results in **Figure 4c**.

**Figure 6(b)** shows the *PER* calculated from **Figure 6(a)**. In **Figure 6(b-i)**, the hybrid MRR with unreduced GO exhibited a low *PER* < ~1 dB when  $P_p$  < ~7 dBm, and the *PER* increased when  $P_p \ge$  ~7 dBm, reaching ~3 dB at  $P_{in}$  = ~15 dBm. For the hybrid MRR with rGO at  $T_R$  = ~100 °C, the *PER* increased from ~2 dB in the low-power state without photothermal reduction to ~4 dB at  $P_p$  = ~15 dBm. For rGO at  $T_R$  = ~200 °C, the *PER* remained unchanged at ~16 dB as  $P_p$  increased from ~0 dBm to ~15 dBm. These results further confirms that the hybrid MRR with highly reduced GO is less susceptible to variations in the input power and shows a better power stability.



**Figure 7.** I (**a**) Extinction coefficients (k's) of 2D GO films versus  $P_p$  obtained by fitting the results in **Figure 6(a)** with optical mode simulations. (**i**) – (**iii**) show the results for GO, rGO after heating at  $T_R = -100$  °C, and rGO after heating at  $\sim 200$  °C, respectively. (**b**) Anisotropy ratios of k values for TE and TM polarizations ( $k_{TE}/k_{TM}$ ) extracted from (**a**).

In **Figure 6**, the variations in the ER of the hybrid MRRs cannot directly indicate changes in the properties of the GO films. To address this, we further extracted the extinction coefficients (k's) of 2D GO films by fitting the results in **Figure 6(a)** with theory and plotted them in **Figure 7(a)**. In our fitting process, we first obtained the GO-induced EPL by fitting the measured transmission spectrum of the hybrid MRR based on the scattering matrix method [59]. After that, the k of 2D GO film was extracted from the obtained EPL by using the same method as we used in **Figure 3(b)**. Note that the photothermal changes in GO films coated on integrated waveguides or MRRs actually exhibit nonuniform behavior along the direction of light propagation [33]. This occurs because, as the light power diminishes along the 2D film, the photothermal effects become weaker, resulting in a smaller difference in properties between the photothermally reduced GO and the unreduced GO. For simplification, in our fitting process we regarded the 10- $\mu$ m-long GO or rGO films in the hybrid MRRs as uniform films with consistent loss. In principle, this approximation can lead to slight

deviations in the fit k values, particularly at a high  $P_p$ . Despite this, the fit k can still be regarded as an average value reflecting the over-all loss performance of the GO films at different  $P_p$ .

For unreduced GO in **Figure 7(a-i)**, the k values at low pump powers (e.g.,  $P_p = ~0$  dBm) are ~0.0088 and ~0.0017 for TE and TM polarizations, respectively. These values obtained from the MRR experiment show good agreement with those obtained from the waveguide experiment in **Figure 3(b)**, reflecting the consistency of our GO film fabrication process. The k for TE polarization increases when  $P_p \ge ~7$  dBm and reaches ~0.1634 at  $P_m = ~15$  dBm - ~17 times of the k at  $P_p < ~7$  dBm. This suggests that the change in k induced by localized photothermal reduction of GO is quite significant, even though the variation in ER shown in **Figure 6(a)** is not very noticeable. This is mainly due to the fact the ER in **Figure 6(a)** was plotted on a dB scale, which results in less significant change for the ER with a lower value.

**Figures 7(a-ii)** and **7(a-iii)** show the corresponding results for the hybrid MRR with 1 layer of rGO after heating at  $T_R = \sim 100$  °C and  $\sim 200$  °C, respectively. For rGO at  $T_R = \sim 100$  °C, the TE-polarized k increases from  $\sim 0.1013$  at  $P_p < \sim 10$  dBm to  $\sim 0.1670$  at  $P_p = \sim 15$  dBm. Whereas the k for TM polarization slightly increases from  $\sim 0.0183$  at  $P_p < \sim 15$  dBm to  $\sim 0.0197$  at  $P_p = \sim 15$  dBm. In contrast, the k of rGO at  $T_R = \sim 200$  °C remains constant for both polarizations (*i.e.*,  $k = \sim 0.7022$  for TE polarization and  $k = \sim 0.0367$  for TM polarization) as  $P_{in}$  increases from  $\sim 0$  dBm to  $\sim 15$  dBm.

**Figure 7(b)** shows the anisotropy ratios calculated from **Figure 7(a)**. In **Figure 7(b-i)**, the anisotropy ratio for unreduced GO remains constant at ~4.5 when  $P_p$  < 7 dBm, showing agreement with the results in **Figure 3(c)**. For  $P_p \ge 7$  dBm, the anisotropy ratio increases with  $P_p$ , achieving a maximum value of ~8.8 at  $P_{in}$  = ~15 dBm. For rGO at  $T_R$  = ~100 °C, the anisotropy ratio remains unchanged at ~6.2 when  $P_p$  < ~10 dBm before experiencing a gradual increase to ~9.6 at  $P_p$  = ~15 dBm. For rGO at  $T_R$  = ~200 °C, the anisotropy ratio remains unchanged at ~18.1 within the measured input pump power range. These results further confirm that 2D GO films exhibit more significant loss anisotropy as the degree of reduction increases. This will work be aided with the use of other novel 2D materials [60–90] that will be extremely useful for all forms of microcombs [91–155] with the use of novel designs [156–164] for a wide range of applications to classical and quantum nonlinear optics. [165–223]

### **Conclusions**

In summary, we experimentally demonstrate integrated waveguide and MRR polarizers incorporating rGO. We integrate 2D GO films onto silicon photonic devices with precise control over their thicknesses and sizes, and use two methods – uniform thermal reduction and localized photothermal reduction – to reduce the GO films. Detailed measurements are performed for devices with different lengths, thicknesses, and reduction levels of the GO films. The results show that the devices with rGO exhibit better polarizer performance than those with GO. A maximum *PDL* of ~47 dB is achieved for the hybrid waveguide with rGO, and the hybrid MRR with rGO achieves a maximum *PER* of ~16. By fitting the experimental results with theory, it reveals that rGO exhibits more significant loss anisotropy, with an anisotropy ratio more than 4 times that of GO. In addition, rGO also exhibits enhanced thermal stability and lower sensitivity to photothermal reduction. Our work opens up new opportunities for implementing high-performance polarization-selective devices through on-chip integration of 2D rGO films.

**Competing interests:** The authors declare no competing financial interests.

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