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*Article*

# Graphene Oxide 2D Films for Integrated Thermo-Optic Devices

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**Abstract:** Efficient heat management and control in optical devices, facilitated by advanced thermo-optic materials, is critical for many applications such as photovoltaics, thermal emitters, mode-locked lasers, and optical switches. Here, we investigate the thermo-optic properties of 2D graphene oxide (GO) films by precisely integrating them onto microring resonators (MRRs) with control over the film thicknesses and lengths. We characterize the refractive index, extinction coefficient, thermo-optic coefficient, and thermal conductivity of for the GO films with different layer numbers and degrees of reduction, including reversible reduction and enhanced optical bistability induced by photo-thermal effects. Experimental results show that the thermo-optic properties of 2D GO films vary widely with the degree of reduction, with significant polarization anisotropy, enabling efficient polarization sensitive devices. The versatile thermo-optic response of 2D GO substantially expands the scope of functionalities and devices that can be engineered, making it promising for a diverse range of thermo-optic applications.

**Keywords:** integrated photonics; 2D materials; thermo-optics

## 1. Introduction

The management and control of heat [1] is important for tackling issues like global warming, energy shortages, and device overheating. Thermo-optic effects have underpinned the functionality and efficiency of many optical devices and systems such as photovoltaics [2,3], thermal emitters [4,5], mode-locked lasers [6,7], optical switches [8,9], logic gates [10,11], power limiters [12,13], optical memories [14,15], and sensors [16,17].

Over the past two decades, the rapid advances in material science have provided powerful tools to efficiently manipulate heat transfer in optical devices [18,19]. This has been particularly facilitated by the emergence of many novel 2D materials with atomically thin film thicknesses, such as graphene, transition metal dichalcogenides (TMDCs), and hexagonal boron nitride (hBN) [20,21]. These 2D materials exhibit many remarkable thermo-optic properties, including the ability to tune the refractive index, absorption, conductivity and anisotropy. These have enabled a range of high performance devices with new functionality compared to conventional bulk materials [22,23].

As a common derivative of graphene, graphene oxide (GO) is a rising star in the 2D material family [24–26]. It exhibits many distinctive optical properties, such as a broadband response, high optical nonlinearity, and significant anisotropy [27–29]. There is also a high degree of flexibility in engineering GO's properties through reduction and doping processes [17,30,31], offering a flexible platform for engineering a wide range of functionalized materials. Moreover, facile solution-based synthesis processes and transfer-free film coating with precise control have been developed for 2D GO films [32–34], with a high degree of compatibility with integrated device platforms.

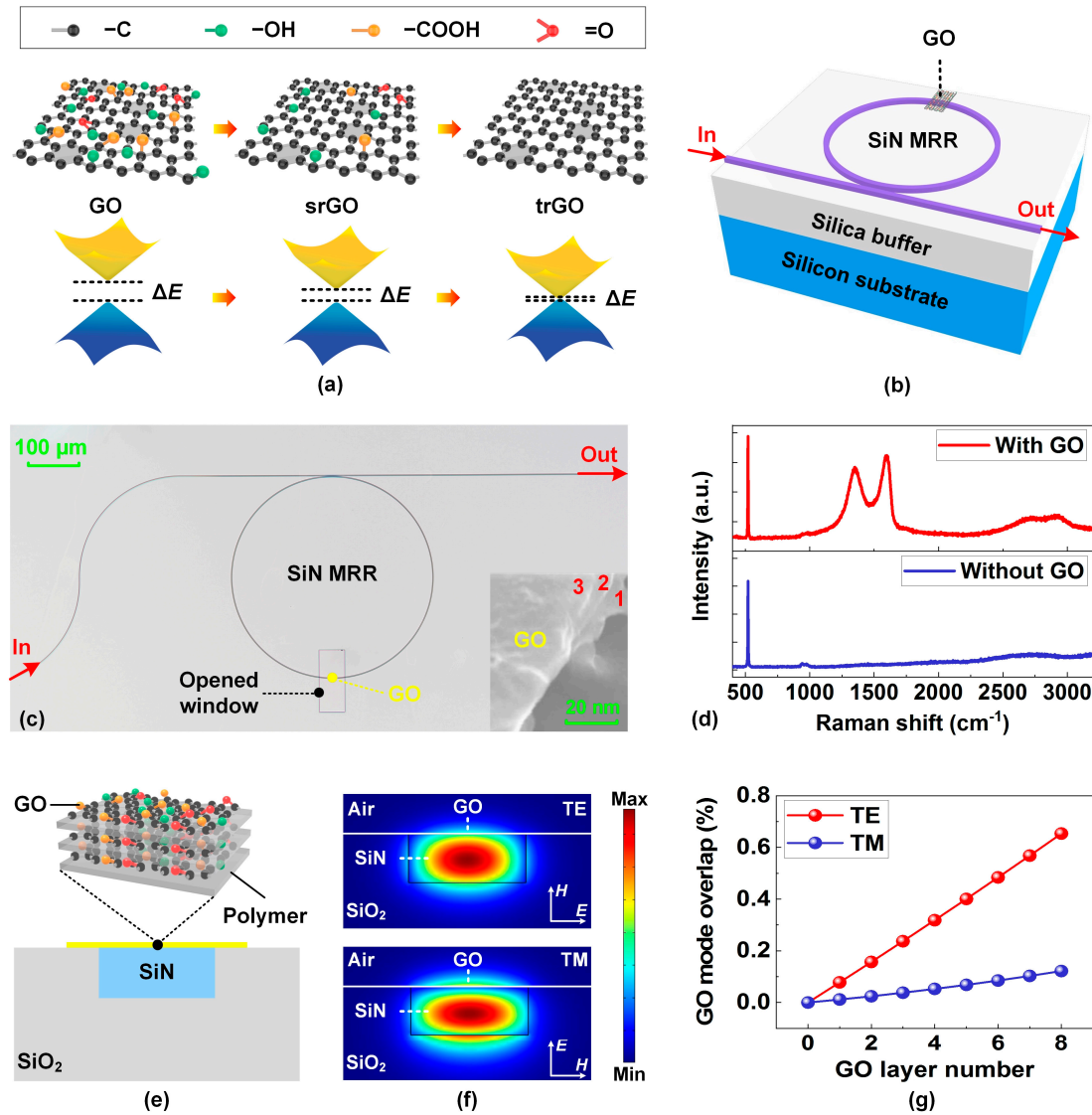
In this paper, we comprehensively investigate a range of thermo-optic properties of 2D GO films by integrating them onto silicon nitride (SiN) microring resonators (MRRs) with precise control over the film thicknesses and lengths. We characterize the refractive index, extinction coefficient, thermo-optic coefficient, and thermal conductivity of 2D layered GO films for different layer numbers and degrees of reduction. Experimental results show that the properties, including the fundamental

thermo-optic response itself, of 2D GO films vary widely with temperature as the reduction degree increases, including an increase of  $\sim 0.2280$  in the refractive index, a more than 36-fold increase in the extinction coefficient, a transition from a positive thermo-optic coefficient to a negative one, and a  $\sim 48$  times improvement in the thermal conductivity. In addition, the GO films shows a markedly anisotropic response for light in transverse electric (TE) and transverse magnetic (TM) polarizations, including a difference of  $\sim 0.14$  for the refractive index, a ratio of  $\sim 4$  for the extinction ratio, a ratio of  $\sim 6$  for the thermo-optic coefficient, and a ratio of  $\sim 18$  for the thermal conductivity. We also characterize reversible GO reduction and optical bistability in the hybrid MRRs induced by photo-thermal effects. Experimental findings reveal that reversible GO reduction can be initiated by light power within a specific range, and that the hybrid MRRs exhibit significantly enhanced optical bistability compared to uncoated MRRs. These results offer interesting insights for the versatile thermo-optic properties of 2D layered GO films, which offer significant potential for thermo-optic applications.

## 2. Device Design and Fabrication

Figure 1a illustrates the atomic structures and bandgaps of GO, semi-reduced GO (srGO), and totally reduced GO (trGO). As a derivative of graphene, GO contains various oxygen functional groups (OFGs), such as hydroxyl, epoxide, carbonyl, and carboxylic groups, located either on the carbon basal plane or at the sheet edges [24]. Unlike graphene, which possesses a zero bandgap, the presence of the OFGs in GO results in a large optical bandgap  $\Delta E$  typically ranging between  $\sim 2.1$  eV and  $\sim 3.6$  eV [26,35]. Due to its large optical bandgap, GO exhibits low linear optical absorption at infrared wavelengths, which is about 2 orders of magnitude lower than that of graphene [36]. The reduction of GO leads to the dissociation of OFGs, along with the decreasing of  $\Delta E$  and the changing of the material properties such as refractive index, optical absorption, and thermal conductivity [25]. In practical, the reduction of GO films can be realized by using various thermal reduction, chemical reduction, or photoreduction methods [24]. The bandgap and optical properties of trGO are similar to graphene, with slight differences arising from the defects within the carbon networks [37].

Figure 1b shows a schematic of an integrated SiN microring resonator (MRR) coated with a 2D GO film. Figure 1c shows a microscopic image of a fabricated device coated with a film including 3 layers of GO, with the inset showing a scanning electron microscopy (SEM) image of the layered GO film. The SiN MRR was fabricated via CMOS compatible and crack-free processes, as reported previously [29,38]. A  $2.3\text{-}\mu\text{m}$ -thick silica layer was deposited on the top of the fabricated SiN MRRs as an upper cladding. To facilitate the interaction between the GO film and the evanescent field of the SiN MRR, lithography and dry etching processes were employed to open a window in the silica cladding, extending down to the top surface of the SiN MRR. The coating of the 2D layered GO film was realized by using a solution-based method that allowed transfer-free and layer-by-layer film deposition [32,39]. During the coating process, four steps were repeated to assemble a multilayered film consisting of alternating GO monolayers and polymer layers with oppositely charged surfaces. As can be seen from Figure 1c, the coated GO film exhibits high transmittance and good morphology. The film also shows a high uniformity without any noticeable wrinkling or stretching. The thickness for the self-assembled GO film increases almost linearly with the number of GO layers, with an average thickness of  $\sim 2$  nm for each layer. Our GO coating method enables large-area film coating with precisely controlled film thickness, which was also used for on-chip integration of 2D GO films to realize other functional devices [17,27,29,31,34].



**Figure 1.** (a) Schematics of atomic structures and bandgaps of graphene oxide (GO), semi-reduced GO (srGO), and totally reduced GO (trGO). (b) Schematic of a GO-coated silicon nitride (SiN) microring resonator (MRR). (c) Microscopic image of a fabricated SiN MRR coated with 3 layers of GO. Inset shows a scanning electron microscopy (SEM) image of the layered GO film, where numbers (1–3) refer to the number of layers for that part of the image. (d) Measured Raman spectra of a SiN chip without GO and coated with 1 layer of GO. (e) Schematic illustration of cross section and (f) corresponding TE and TM mode profiles for the hybrid waveguide with 3 layers of GO. Inset in (e) illustrates the layered GO film fabricated by self-assembly. (g) Mode overlap with GO versus GO layer number for both TE and TM polarizations of the hybrid waveguides.

Figure 1d shows the measured Raman spectra for the SiN chip before and after coating 1 layer of GO, which were measured using a ~514-nm pump laser. The presence of the representative D ( $1345\text{ cm}^{-1}$ ) and G ( $1590\text{ cm}^{-1}$ ) peaks of GO in the Raman spectrum for the GO-coated chip confirms the successful integration of the GO film onto the chip.

In our fabricated SiN MRRs, the cross section for the waveguides forming the MRR (includes both the ring and the bus waveguide) was  $\sim 1.60\text{ }\mu\text{m} \times \sim 0.72\text{ }\mu\text{m}$ , as illustrated in Figure 1e. The inset shows a schematic of the layered GO film fabricated by self-assembly. Figure 1f shows the TE and TM mode profiles for the hybrid waveguide with 3 layers of GO in Figure 1e, which were simulated using a commercial mode solving software. The corresponding effective refractive indices at 1550 nm were  $n_{\text{eff, TE}} \sim 1.784$  and  $n_{\text{eff, TM}} \sim 1.713$ . In our simulation, the refractive indices of GO and SiN at 1550 nm were  $n_{\text{GO}} = 1.972$  and  $n_{\text{SiN}} = 1.990$ , respectively. These values were obtained from our experiments



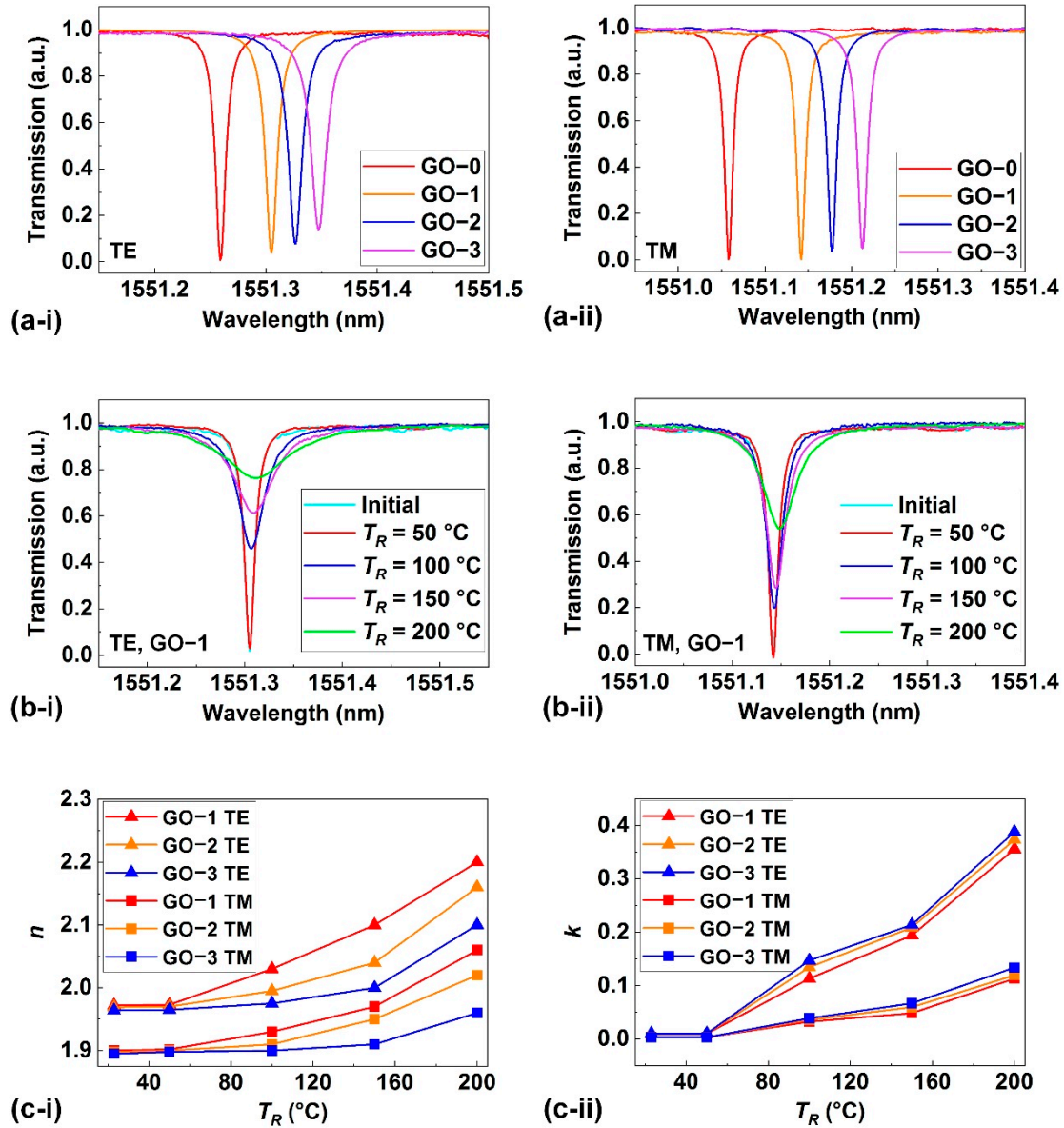
introduced in the following sections. Figure 1g shows GO mode overlap versus GO layer number for both TE and TM polarizations, which were calculated based on mode simulations of the hybrid waveguide at 1550 nm. Due to the difference in volume between the bulk SiN waveguide and the ultrathin 2D GO films, most light power is confined within the SiN waveguide ( $> 88\%$ ), and the GO mode overlap is  $< 1\%$ . For both TE and TM polarizations, the GO mode overlap increases with the GO layer number, mainly resulting from the increase in the GO film thickness. The GO mode overlap for TE polarization is higher than that for the TM polarization. Given that the GO film has much higher light absorption compared to the SiN waveguide, a stronger mode overlap with GO leads to a higher propagation loss for the GO-SiN hybrid waveguide.

### 3. Refractive Indices and Extinction Coefficients

By using the fabricated MRRs coated with 2D GO films, we first characterize the refractive indices ( $n$ 's) and extinction coefficients ( $k$ 's) of the GO films with different thicknesses or after experiencing different degrees of reduction. For all the experiments performed in this section, we employed SiN MRRs with a radius of  $\sim 200\ \mu\text{m}$ . The length of the coated GO films, which approximately equalled to the width of the opened window in the silica cladding, was  $\sim 50\ \mu\text{m}$ . Light coupling to the fabricated devices was achieved by using lensed fibres that were butt-coupled to inverse-taper couplers located at both ends of the bus waveguides. The fibre-to-chip coupling loss was  $\sim 4\ \text{dB}$  per facet. We also employed a polarization controller (PC) to adjust the polarization of the input light and characterized the  $n$ ,  $k$  values for both TE and TM polarizations.

Figure 2a shows the measured transmission spectra of a SiN MRR coated with 1–3 layers of GO for both TE and TM polarizations. The corresponding results for the uncoated SiN MRR (GO-0) are also shown for comparison. In order to minimize the wavelength shift induced by thermo-optic effects, the transmission spectra were measured by scanning the wavelength of an input continuous-wave (CW) light with a low power of  $\sim 0\ \text{dBm}$ . Unless otherwise specified, the input power in our following discussion refers to the power coupled into the bus waveguide after excluding the fibre-to-chip coupling loss. Compared to the uncoated SiN MRR, the GO-coated MRRs exhibited resonance redshifts, which became more significant as the GO layer number increases. This indicates the variation in the waveguide effective refractive index caused by the GO films. On the other hand, the GO-coated MRRs exhibited decreased extinction ratios ( $ER$ s, defined as the ratio of the maximum to the minimum transmission) compared to the uncoated MRR, and the  $ER$  decreased for increasing GO layer numbers. This reflects that the presence of the GO films also resulted in changes to the waveguide propagation loss.

Figure 2b shows the measured transmission spectra of a SiN MRR coated with 1 layer of GO for both TE and TM polarizations. We measured the transmission spectra for the same MRR by scanning a CW light with a power of  $\sim 0\ \text{dBm}$  (*i.e.*, the same as that in Figure 2a). Before measuring the spectra, the integrated chip was heated on a hot plate for 15 minutes at various temperatures  $T_R$  ranging from  $\sim 50$  to  $200\ ^\circ\text{C}$ . We did not further increase  $T_R$  beyond  $200\ ^\circ\text{C}$  because the polymer layers within the GO films could not withstand temperatures in this range. The corresponding results for the unheated MRR (initial) are also shown for comparison. At  $T_R = 50\ ^\circ\text{C}$ , the spectrum of the heated MRR showed negligible difference compared to that of the unheated MRR. Whereas for  $T_R \geq 100\ ^\circ\text{C}$ , the heated MRR exhibited a significantly decreased  $ER$ . Given that reduced GO has higher light absorption than unreduced GO [24], these results indicate that the 2D GO film did not undergo reduction at a relatively low  $T_R$ . However, as the temperature continued to rise above a certain threshold, thermal reduction of GO occurred, and the degree of reduction increased with  $T_R$ . On the other hand, we observed very slight redshifts in the resonance wavelength for  $T_R \geq 100\ ^\circ\text{C}$ . This reflects that the reduction of GO also resulted in slight changes in the waveguide effective refractive index.



**Figure 2.** (a) Measured (i) TE- and (ii) TM-polarized transmission spectra of the hybrid MRRs with 1–3 layers of GO. The corresponding results for the uncoated SiN MRR (GO-0) are also shown for comparison. (b) Measured (i) TE- and (ii) TM-polarized transmission spectra of a hybrid MRR with 1 layer of GO. The GO-SiN chip underwent heating at various temperatures  $T_R$  prior to the measurement of its transmission spectra. The corresponding results for the chip before heating (initial) are also shown for comparison. (c) Extracted (i) refractive index  $n$  and (ii) extinction coefficient  $k$  of the films including 1–3 layers of GO versus  $T_R$  for both TE and TM polarizations.

Figure 2c shows  $n, k$  values of the layered GO films, which were obtained by using the scattering matrix method [40] to fit the measured transmission spectra in Figure 2a,b. The refractive index of SiN was  $\sim 1.9902$  at 1550 nm, which was obtained by fitting the measured spectra for the uncoated SiN MRR. To improve the accuracy, the slight decrease in the film thicknesses (*i.e.*,  $\sim 0.5$  nm for 1 layer of GO at  $T_R = 200$  °C, measured by atomic force microscopy) was considered when we calculated the  $n, k$  values for reduced GO. In Figure 2c,  $n$  slightly decreases as the GO layer number increases, but  $k$  shows an opposite trend. The former could be attributed to the presence of more air voids in a thicker GO film, which arose from imperfections during the GO film's fabrication processes. Whereas the latter was resulting from an increase in scattering loss due to film unevenness and accumulation of imperfect contact between adjacent layers in a thicker film.

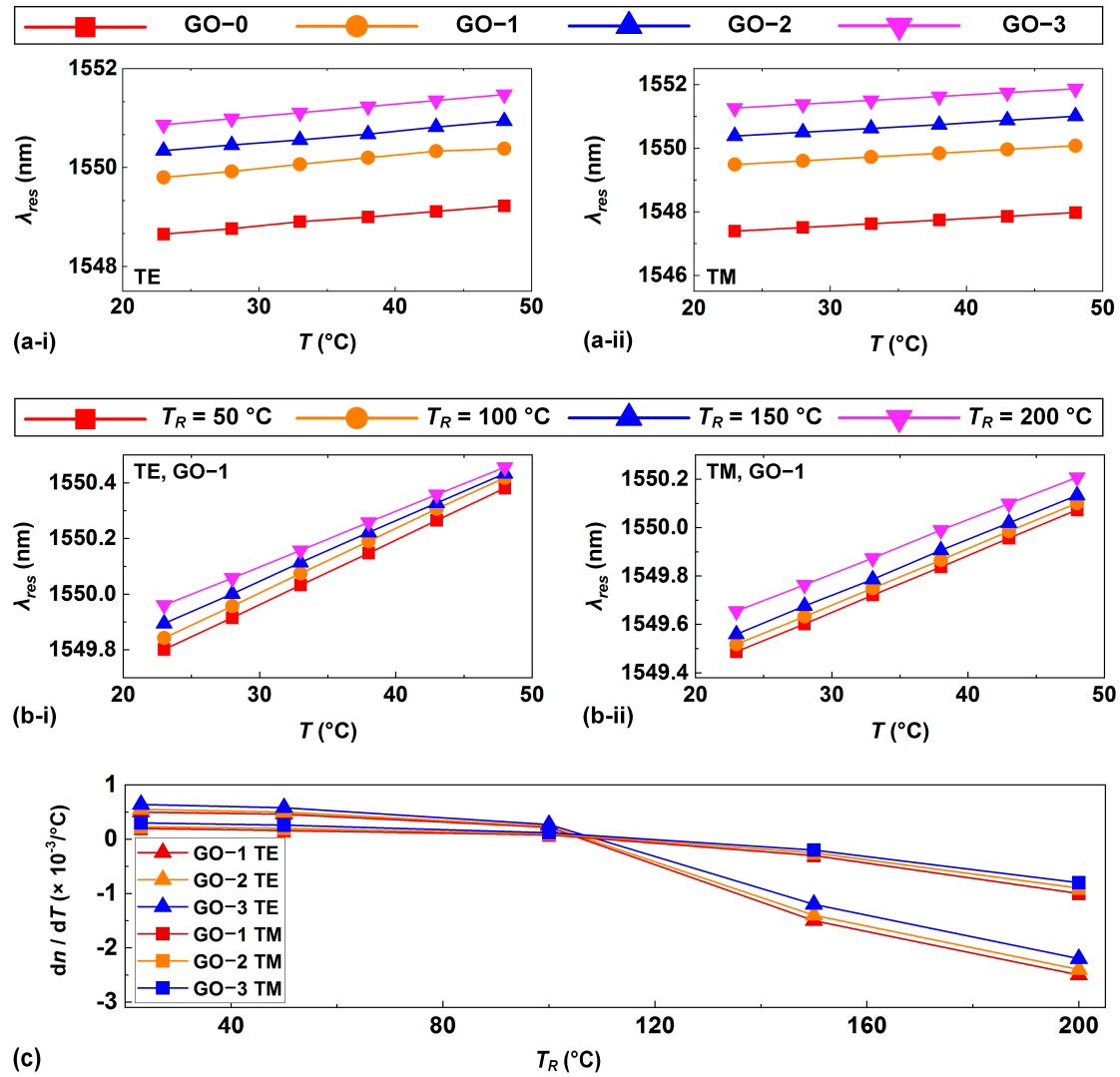
In Figure 2c, both  $n$  and  $k$  initially exhibited no significant changes for  $T_R \leq 50$  °C, but then show a more noticeable increase with  $T_R$  for  $T_R \geq 100$  °C. These results reflect the changes in GO's  $n$ ,  $k$  induced by thermal reduction. Since the CW light power used to scan the spectra was not sufficient to induce any significant thermo-optic effects, the changes in  $n$ ,  $k$  were mainly induced by changes resulting from the heating treatment before the measurement. The  $n$ ,  $k$  values for unreduced GO were  $\sim 1.9720$  and  $\sim 0.0097$ , respectively. In contrast, the corresponding values for the reduced GO at  $T_R = 200$  °C were  $\sim 2.2000$  and  $\sim 0.3557$ . The change in  $n$ , *i.e.*,  $\Delta n = \sim 0.2280$ , is over 1 order of magnitude larger than conventional bulk refractive materials [41]. In addition,  $k$  also changes from  $\sim 0.0097$  to  $\sim 0.3557$  – an increase of over 36-fold. The broad variation ranges for both  $n$ ,  $k$  are critical for engineering and manipulation of the phase and amplitude response of many GO devices, such as optical lenses and holographic displays [31,42].

The results in Figure 2c also reveal the differences in the  $n$ ,  $k$  values between TE and TM polarizations. For all different GO layer numbers and  $T_R$ 's, the GO film exhibits higher values of  $n$ ,  $k$  for TE polarization than TM polarization. This is mainly caused by the intrinsic material anisotropy of the 2D GO films with atomically thin thicknesses, which leads to distinctive response for light in different polarization states. In the GO-SiN MRR, the TE polarization supports in-plane light-GO interaction, which is much stronger as compared to the out-of-plane interaction supported by TM polarization. For 1 layer of GO, the  $n$ ,  $k$  values for TE polarization were  $\sim 1.9720$  and  $\sim 0.0097$ , respectively. In contrast, the corresponding values for TM polarization were  $\sim 1.9000$  and  $\sim 0.0032$ . The large difference in the  $n$ ,  $k$  values between TE and TM polarizations reflect the significant material anisotropy of the 2D GO films, which is useful for realizing liquid crystal display devices and optical polarizers [28,43].

#### 4. Thermo-Optic Coefficients

In this section, we use the fabricated GO-SiN MRRs to characterize the thermo-optic coefficients of the GO films with different thicknesses or after experiencing different degrees of reduction. The thermo-optic coefficient is a fundamental parameter that indicates the change in the refractive index of a material with variations in environmental temperature. For the experiments performed in this section, we employed SiN MRRs with a radius of  $\sim 56$   $\mu\text{m}$  that was smaller than those used in Section 3. In addition, the silica upper cladding of the integrated chip was removed by using highly selective chemical-mechanical polishing (CMP) and the entire MRR was coated with GO films. We chose these MRRs in order to magnify the difference in measured wavelength shifts caused by GO films. We measured the transmission spectra of the GO-SiN MRRs by scanning the wavelength of a CW input with a power of  $\sim 0$  dBm. To adjust the temperature of the GO-SiN MRRs, the fabricated chip was placed on a temperature controller with a minimum resolution of 0.1 °C.

Figure 3a shows the measured resonance wavelength  $\lambda_{\text{res}}$  versus chip temperature  $T$  for the SiN MRRs coated with 1–3 layers of GO. We show the results for both TE and TM polarizations, and the corresponding results for the uncoated SiN MRR (GO-0) are also shown for comparison. To prevent changes in GO film properties caused by thermal reduction, the chip temperature was not raised beyond 50 °C. As can be seen, all the MRRs exhibited a redshift in their resonance wavelengths, albeit at slightly different rates. For instance, the TE-polarized  $\lambda_{\text{res}}$  of the uncoated MRR redshifted at a rate of  $\sim 22.9$  pm/°C, in contrast to  $\sim 23.3$  pm/°C for the TE-polarized  $\lambda_{\text{res}}$  of the hybrid MRR with 1 layer of GO. Figure 3b shows the measured  $\lambda_{\text{res}}$  versus  $T$  for a hybrid MRR with 1 layer of GO. Similar to that in Figure 2b, before conducting the measurement, the GO-SiN chip was heated at various  $T_R$  on a hot plate for 15 minutes. As a result, the measured redshifts in Figure 3b can be used to calculate the thermo-optic coefficients of reduced GO at various reduction degrees.



**Figure 3.** (a) Resonance wavelength  $\lambda_{res}$  versus chip temperature  $T$  (changed via a temperature controller) for the hybrid MRRs with 1–3 layers of GO. (i) and (ii) show the results for TE and TM polarizations, respectively. The corresponding results for the uncoated SiN MRR (GO-0) are also shown for comparison. (b)  $\lambda_{res}$  versus  $T$  for the hybrid MRR with 1 layer of GO. The GO-SiN chip underwent heating at temperatures  $T_R$  ranging from ~50 to 200 °C prior to the measurement. (i) and (ii) show the results for TE and TM polarizations, respectively. (c) Extracted thermo-optic coefficient  $dn/dT$  of the films including 1–3 layers of GO versus  $T_R$  for both TE and TM polarizations.

Figure 3c shows thermo-optic coefficient  $dn/dT$  of the films including 1–3 layers of GO versus  $T_R$  for both TE and TM polarizations. The values of  $dn/dT$  were calculated based on the results in Figure 3a,b, using the relationship between the resonance wavelengths and the waveguide effective refractive index given by:

$$n_{eff} \times 2\pi / \lambda_m \times L = m \times 2\pi \quad (1)$$

where  $n_{eff}$  is the effective refractive index of the hybrid waveguide,  $L$  is the circumference of the SiN MRR, and  $\lambda_m$  is the resonance wavelength of the  $m$ th resonance.

In Figure 3c, as  $T_R$  increases, the value of  $dn/dT$  changes from positive to negative for all the three GO layer numbers. For 1 layer of unreduced GO in TE polarization, the value of  $dn/dT$  is  $\sim 0.5 \times 10^{-3}/^{\circ}\text{C}$ , in contrast to  $\sim -2.5 \times 10^{-3}/^{\circ}\text{C}$  for reduced GO at  $T_R = 200$  °C with the same GO layer number and polarization. These results reflect interesting changes in GO's thermo-optic coefficient induced by thermal reduction. The thermo-optic coefficient of unreduced GO is higher than silicon ( $\sim 1.8 \times 10^{-4}/^{\circ}\text{C}$  [44]). Given that most materials have positive values of thermo-optic coefficients, the negative



thermo-optic coefficients exhibited by reduced GO can be utilized to mitigate thermal drift induced by temperature variation and develop athermal devices [40,45]. The large variation range for the thermo-optic coefficient also increases the range of functionalities and devices that can be developed.

We also note that thicker GO films showed higher values of  $dn/dT$  in Figure 3c, which can be attributed to diminished thermal dissipation in them, especially considering the presence of polymer layers between the GO layers. Similar to that in Figure 2c, the GO film exhibits significant difference in the values of  $dn/dT$  for TE and TM polarizations. For unreduced GO with positive values of  $dn/dT$ , the  $dn/dT$  for TE polarization is higher than that for TM polarization. Whereas for reduced GO at  $T_R = 200$  °C with negative values of  $dn/dT$ , the  $dn/dT$  for TE polarization is lower than that for TM polarization. These phenomena can also be attributed to the stronger in-plane light-GO interaction supported by TE polarization compared to the out-of-plane interaction supported by TM polarization. We also calculated the  $dn/dT$  values of SiN based on the measured results for an uncoated SiN MRR, which were  $\sim 2.5 \times 10^{-5}/^{\circ}\text{C}$  and  $\sim 2.4 \times 10^{-5}/^{\circ}\text{C}$  for TE and TM polarizations, respectively. These values show agreement with those reported in Ref. [46], and the close resemblance between the coefficients for TE and TM polarizations reflects that SiN does not exhibit significant anisotropy in terms of its thermo-optic coefficient.

## 5. Thermal Conductivities

Thermal conductivity is a parameter that defines material's ability to conduct heat, which is of fundamental importance in modelling heat transfer for thermal management [18]. In this section, we characterize the thermal conductivities of the layered GO films with different thicknesses and reduction degrees by using the fabricated GO-SiN MRRs.

For the experiments performed in this section, we employed SiN MRRs with a radius of  $\sim 200$   $\mu\text{m}$ , and the length of the coated GO films was  $\sim 50$   $\mu\text{m}$  (*i.e.*, the same as those used in **Section 3**). We used two CW lights to measure the transmission spectra of the GO-coated MRRs. The first one served as a pump injecting into one of the MRR's resonances. The wavelength of this input CW light was slightly tuned from blue to red around the resonance until it reached a steady thermal equilibrium state with a stable output power. After this, the second CW light, with a constant power of  $\sim 0$  dBm, was employed as a low-power probe to scan the MRR's transmission spectrum. Compared to directly employing a high-power CW light to scan the spectra, our approach can minimize the asymmetry in the measured resonance lineshape resulting from optical bistability (which will be discussed in Section 7), thus allowing more precise measurement of the resonance wavelength shifts. We measured the resonance wavelength shifts of the fabricated GO-SiN MRRs for various input pump powers, and extracted the thermal conductivity by fitting the wavelength shift with theoretical simulations.

Figure 4a shows the measured resonance wavelength shift  $\Delta\lambda$  versus the power of the CW pump  $P_p$  for the uncoated SiN MRR and the hybrid MRR with 1 layer of GO. We chose the range of  $P_p \leq \sim 40$  mW because, within this range, there were no observable changes in the *ER* of the hybrid MRR compared to that measured at  $P_p = 0$ . This implies that GO film did not experience thermal reduction induced by the CW pump power. For both the uncoated and the hybrid MRRs,  $\Delta\lambda$  increases gradually for smaller values of  $P_p$ , and then increases more significantly as  $P_p$  increases. As  $P_p$  increases, the difference in  $\Delta\lambda$  between the uncoated and hybrid MRRs also becomes more significant.

In our modelling, the steady-state temperature distributions in the waveguide cross section were simulated using commercial finite-element multi-physics software (COMSOL Multiphysics) to solve the heat equation described by the *Fourier's law* as [18]:

$$-\nabla \cdot (K\nabla T) = q \quad (2)$$

where  $q$  is the heat flux density,  $K$  is the material's thermal conductivity,  $\nabla T$  is the temperature gradient, and  $\nabla$  acting on the vector function  $K\nabla T$  is the divergence operator. The heat flux intensity  $q$  in the MRR was calculated by [47]:

$$q = \frac{P \times BUF \times R}{w \times h \times l} \quad (3)$$

where  $P$  is the input CW power that equals to  $P_p$  (neglecting the small difference induced by the low-power probe),  $R$  is the conversion efficiency that determines the amount of light power converted into heat,  $w$ ,  $h$ , and  $l$  are the waveguide width, height, and length, respectively, and  $BUF$  is the MRR's intensity build-up factor that can be expressed as [48]:

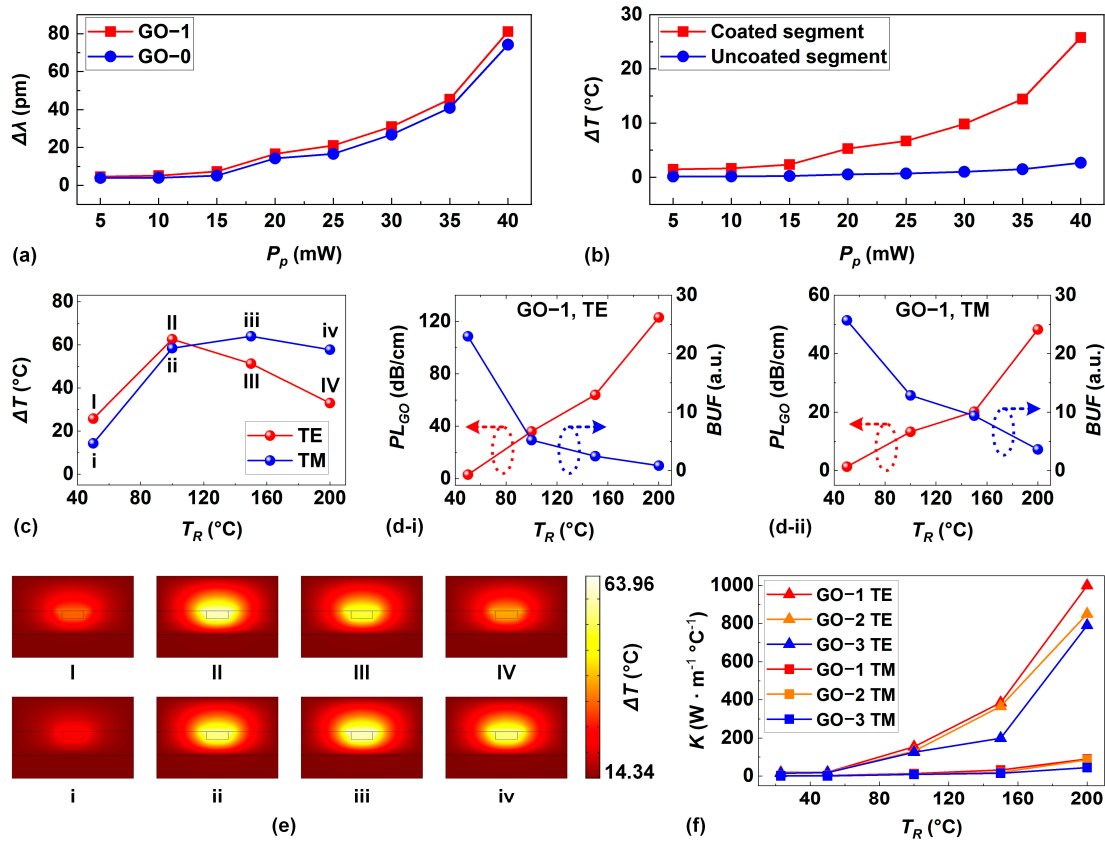
$$BUF = \frac{(1 - t^2)a^2}{1 - 2ta + t^2a^2} \quad (4)$$

where  $a$  is the round-trip amplitude transmission, and  $t$  is the field transmission coefficient of the directional coupler. The values of  $a$  and  $t$  were obtained by fitting the measured spectrum of the MRR.

Based on the measured resonance wavelength shift for the uncoated SiN MRR in Figure 4a, we first calculated the variation in the waveguide effective refractive index, and extracted the change in the refractive index of SiN  $\Delta n_{SiN}$  based on optical mode simulations (e.g.,  $\Delta n_{SiN} = \sim 8.54 \times 10^{-5}$  at  $P_p = 40$  mW). Next, by dividing  $\Delta n_{SiN}$  by the thermo-optic coefficient of SiN (i.e.,  $\sim 2.50 \times 10^{-5}/^\circ\text{C}$ , obtained from the experiments in Section 4), we derived the change in temperature  $\Delta T$  at the waveguide core (e.g.,  $\Delta T = \sim 3.4^\circ\text{C}$  at  $P_p = 40$  mW). Finally, by fitting the  $\Delta T$  value at the waveguide core with simulated temperature distribution, we obtained the heat flux intensity  $q$  and calculated  $R$  for the uncoated SiN MRR based on Eq. (3) (i.e.,  $R = \sim 0.66\%$ ).

On the other hand, we divided the hybrid MRR into uncoated and GO-coated segments. For the uncoated segment, since the presence of GO increased the round-trip loss of the MRR and hence reduced the  $BUF$ , the heat flux density and temperature increase in the uncoated segment were lower compared to those for the uncoated SiN MRR. Assuming that  $R$  for the uncoated segment remains consistent with that obtained from the uncoated MRR, and neglecting the slight temperature variation induced by heat transfer between the uncoated and GO-coated segments, we obtained the  $\Delta T$  for the uncoated segment in the hybrid MRR via thermal simulations (using  $q$  calculated based on Eq. (3)). Using this  $\Delta T$ , we further calculated the additional phase shift along the uncoated segment based on Eq. (1). This, together with the measured resonance wavelength shift for the hybrid SiN MRR in Figure 4a, allows us to calculate the additional phase shift along the GO-coated segment and the corresponding  $\Delta T$  in this segment.

Based on the methods mentioned above, we calculated  $\Delta T$  values for the uncoated and GO-coated segments of a hybrid MRR with 1 layer of GO, and plotted them as functions of  $P_p$  in Figure 4b. It is evident that, owing to the presence of GO, the two segments experienced different temperature changes at the SiN waveguide core. Given that the hybrid MRR with 1 layer of GO has a  $BUF$  value of  $\sim 23.0$  that is lower than the  $BUF$  of  $\sim 28.2$  for the uncoated MRR, the uncoated segment within the hybrid MRR has a reduced heat flux intensity than that in the uncoated MRR. This leads to a lower  $\Delta T$  value for the uncoated segment within the hybrid MRR relative to the uncoated MRR (e.g.,  $\sim 2.8^\circ\text{C}$  versus  $\sim 3.4^\circ\text{C}$  at  $P_p = 40$  mW). Even though the lower  $\Delta T$  caused a smaller phase shift along the uncoated segment, in Figure 4a the hybrid MRR exhibited a more significant redshift compared to the uncoated MRR. This indicates that the GO-coated segment experienced a more significant phase shift resulting from the heat generated by the GO films. This is also reflected by the higher values of  $\Delta T$  for the GO-coated segment in Figure 4b.



**Figure 4.** (a) Measured resonance wavelength shift  $\Delta\lambda$  versus input continuous-wave (CW) pump power  $P_p$  for the uncoated SiN MRR (GO-0) and the hybrid MRR with 1 layer of GO (GO-1). (b) Calculated changes at the waveguide core  $\Delta T$  for the uncoated and GO-coated segments in the hybrid MRR with 1 layer of GO versus  $P_p$ . (c) Calculated  $\Delta T$  for the GO-coated segment in the hybrid MRR with 1 layer of GO after the chip was heated at various temperatures  $T_R$ . (d) Calculated GO-induced propagation loss  $PL_{GO}$  and intensity build up factor  $BUF$  of the hybrid MRR with 1 layer of GO versus  $T_R$ . (i) and (ii) show the results for TE and TM polarizations, respectively. (e) Simulated temperature distributions in the waveguide cross section for the hybrid MRR with 1 layer of GO. (I) – (IV) and (i) – (iv) correspond to the different points in (c). (f) Extracted thermal conductivity  $K$  of the films including 1–3 layers of GO versus  $T_R$  for both TE and TM polarizations. In (c) – (e),  $P_p = \sim 40$  mW, at which there was no observable changes in the extinction ratios of the hybrid MRRs as compared to those measured at  $P_p = 0$ . In the thermal simulation, the initial temperature was assumed to be at room temperature of 23 °C.

After measuring the resonance wavelength shifts of a hybrid MRR with 1 layer of GO following the heating of the chip at various  $T_R$ , we employed the same method as in Figure 4b to calculate  $\Delta T$  for the GO-coated segment in these MRRs. Figure 4c shows the calculated  $\Delta T$  versus  $T_R$  for both TE and TM polarizations. For comparison, the input pump power was kept the same as  $P_p = \sim 40$  mW, at which there were no observable changes in the ERs of these MRRs as compared to those measured at  $P_p = 0$ . For both TE and TM polarizations,  $\Delta T$  initially rises with  $T_R$  and then starts to decline after reaching peak values at  $T_R = 100$  °C. To delve deeper into the underlying reasons for this interesting phenomenon, we calculated the GO-induced propagation loss  $PL_{GO}$  and the  $BUF$  of these MRRs, and depicted them in Figure 4d. As can be seen,  $PL_{GO}$  increases with  $T_R$ , while  $BUF$  exhibits an opposite trend. This is not surprising given the fact that the reduced GO at  $T_R \geq 100$  °C has a higher light absorption than unreduced GO. Since the light absorbed by the GO film is converted to heat, the increased light absorption for reduced GO also leads to a higher value of  $R$  in Eq. (3). Hence, the observed phenomenon in Figure 4c can be attributed to the trade-off between a decreased  $BUF$  and an increased  $R$  in Eq. (3) as  $T_R$  increases.

Based on the calculated  $\Delta T$  values for the GO-coated segment, we further extracted the thermal conductivities of the GO films by fitting the  $\Delta T$  values with those obtained from thermal simulations based on the finite-element method (FEM). In our simulations, the GO film in the GO-coated segment was regarded as an additional heat source that absorbed light power and generated heat, which in turn caused a temperature change,  $\Delta T$ , in the underlying SiN waveguide. The heat flux densities in the GO films and the SiN waveguide were calculated individually based on Eq. (3). The values of  $R$  for the GO films were calculated based on the linear loss of the GO films obtained in Section 3, assuming that all the light absorbed by GO – equivalent to the additional loss induced by GO – was converted to heat. The values of  $K$  for each of the material regions were also specified (*e. g.*,  $K_{\text{SiN}} = \sim 29.00 \text{ W}/(\text{m}\cdot^\circ\text{C})$ ,  $K_{\text{SiO}_2} = \sim 1.40 \text{ W}/(\text{m}\cdot^\circ\text{C})$ ,  $K_{\text{polymer}} = \sim 0.25 \text{ W}/(\text{m}\cdot^\circ\text{C})$ , and  $K_{\text{air}} = \sim 0.03 \text{ W}/(\text{m}\cdot^\circ\text{C})$  [47,49]), with the exception of the GO film that needed fitting. The initial temperature  $T_0$  was set to  $23^\circ\text{C}$ , which was the ambient temperature during our experiments.

Figure 4e shows the simulated temperature distributions in the waveguide cross section for the hybrid MRR with 1 layer of GO, where (I) – (IV) and (i) – (iv) correspond to the different points in Figure 4c. Figure 4f shows the thermal conductivity  $K$  of the films including 1–3 layers of GO versus  $T_R$  for both TE and TM polarizations. With the exception of the results for 2 and 3 layers of unreduced GO in TE polarization, where the fitting was based on measurements at  $P_p = \sim 30 \text{ mW}$  due to changes in the  $ER$  observed at  $P_p \geq \sim 35 \text{ mW}$ , all other results were obtained by fitting the measurements at  $P_p = \sim 40 \text{ mW}$ .

In Figure 4f, a thicker GO film exhibits a lower value of  $K$ , further confirming its lower thermal dissipation property. For 1 layer of unreduced GO, the value of  $K$  is  $\sim 20.6 \text{ W}/(\text{m}\cdot^\circ\text{C})$  for TE polarization. This value is higher than that reported in Ref. [50] (*i.e.*,  $\sim 2.0 \text{ W}/(\text{m}\cdot^\circ\text{C})$ ) for GO films with greater thicknesses (*e.g.*,  $\sim 143 \text{ nm}$ ). The decrease in the thermal conductivity with increasing film thickness can be attributed to more significant scattering of phonons from the film surface caused by film unevenness, lattice impurities, and structural defects within thicker films.

Compared to unreduced GO, higher values of  $K$  are obtained for reduced GO at  $T_R \geq 100^\circ\text{C}$ , with  $K$  increasing as the GO reduction degree increases. For 1 layer of reduced GO at  $T_R = 200^\circ\text{C}$ , the value of  $K$  is  $\sim 998 \text{ W}/(\text{m}\cdot^\circ\text{C})$  for TE polarization, which is more than 48 times that of comparable unreduced GO. These results reflect that the reduced GO films possess superior heat conduction property, which can be attributed to the diminished presence of the OFGs that induce phonon-scattering from the film surface. We also note that the value of  $\sim 998 \text{ W}/(\text{m}\cdot^\circ\text{C})$  is about five times lower than the reported value for graphene [51], which is possibly due to the fact that the GO film at  $T_R = 200^\circ\text{C}$  was still not fully reduced and the existence of polymer layers with a much lower thermal conductivity in our GO films.

We also note that the TE polarization exhibited higher values of  $K$  than the TM polarization for all the three GO layer numbers in Figure 4f. This indicates that the GO films also exhibit anisotropic thermal conductivity – similar to the anisotropy observed in their refractive index, extinction coefficient, and thermo-optic coefficient. The ratios between the in-plane and out-of-plane thermal conductivities are  $\sim 6.6$  for 1 layer of unreduced GO and  $\sim 11.1$  for 1 layer of reduced GO at  $T_R = 200^\circ\text{C}$ . We note that in Ref. [52] a considerably higher ratio of  $\sim 675$  was achieved for thick ( $\sim 40 \mu\text{m}$ ) reduced GO films annealed at  $1000^\circ\text{C}$ . In these films, the formation of “air pockets” with limited thermal transport capability between the GO layers strongly hindered out-of-plane thermal transport, but not significantly affecting the in-plane thermal conduction. In contrast to this ratio achieved through engineering the fabrication processes of thick GO films, our calculated ratios more accurately reflect inherent anisotropy in the thermal conductivities of 2D GO films.

## 6. GO Reduction Induced by Localized Photo-Thermal Effects

For the experiments in Section 5, the input CW pump power  $P_p$  was maintained below a specific threshold to ensure that there were no obvious changes in the  $ER$ s of the hybrid MRRs compared to that measured at  $P_p = 0$ . As a result, the GO films remained unaffected by thermal reduction induced by the CW pump power. In this section, we further increase  $P_p$  to be above the threshold and characterize the changes in the  $ER$ s of the hybrid MRRs induced by thermal reduction of the GO



films. Compared to thermal reduction of GO caused by heating the entire chip on a hotplate (at various  $T_R$ ) in previous experiments, using the CW pump power to trigger thermal reduction of GO leads to localized effects, which can cause dynamic changes of the GO film properties. For the experiments in this section, we employed the same experimental method as well as the same MRRs as those used in Section 5.

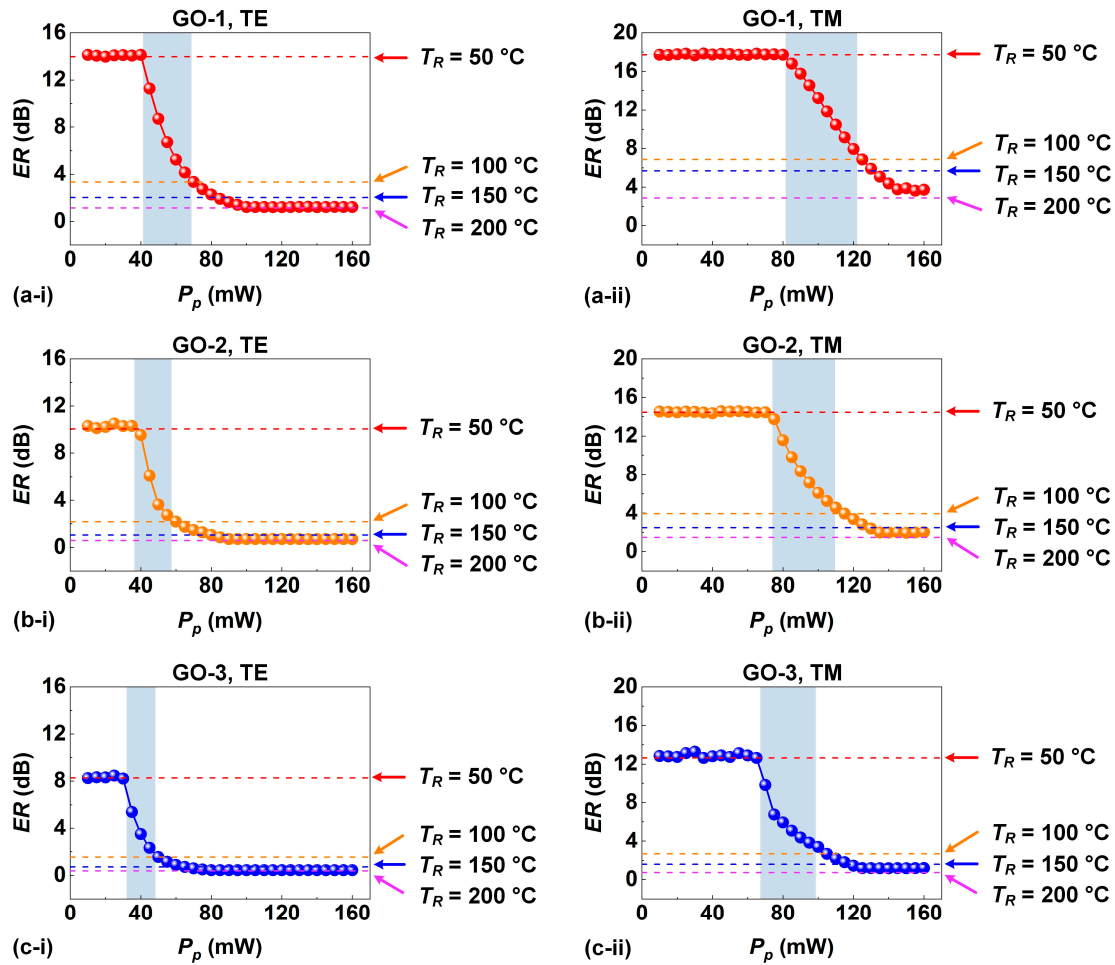
Figure 5a shows the measured  $ER$  versus  $P_p$  for the hybrid MRR with 1 layer of GO. In (i) and (ii), we show the results for TE and TM polarizations, respectively. In Figure 5a-i, the  $ER$  remained constant at  $\sim 14.0$  dB for  $P_p \leq 40$  mW. However, an obvious decrease in the  $ER$  was observed for  $P_p > 40$  mW, indicating increased loss in the hybrid MRR induced by thermal reduction of the GO film. In the hybrid MRR, localized heating occurs due to optical absorption, which leads to a range of photo-thermal effects within the GO film such as self-heating, thermal dissipation, and photo-thermal reduction. Upon reaching the power threshold, the reduction of GO can be initiated when the thermal heating initiates a deoxygenation reaction in GO. The reduced GO with higher light absorption increased the loss in the MRR, consequently lowering its  $ER$ . We also observed an interesting phenomenon – when  $P_p$  exceeded lower power threshold of  $P_{thres1} = \sim 40$  mW but remained below higher power threshold of  $P_{thres2} = \sim 72$  mW (which is highlighted by the blue shaded area), the  $ER$  of the hybrid MRR returned to its initial value of  $\sim 14.0$  dB upon switching off the CW pump. This reversibility reflects the instability of the reduced GO induced by photo-thermal effects, which can easily revert to the original unreduced state once it has cooled down in an oxygen-containing ambient. The reversible GO reduction within the MRR enables dynamic tuning of the MRR's  $ER$ , which can be useful for potential applications such as optical switches and power limiting [53,54].

As  $P_p$  continued to rise above  $\sim 72$  mW, there were permanent changes in the  $ER$  after switching off the CW pump. This occurred because the temperature rise resulting from localized heating caused by the CW pump reached a level where it permanently fractured the chemical bonds between the OFGs and the carbon network. This led to a lasting alteration in the atomic structure of GO and hence its properties. For  $P_p > 110$  mW, the  $ER$  nearly approaches 0, indicating a substantial increase in loss induced by the highly reduced GO. It should be noted that for  $P_p > \sim 72$  mW, there was an increase in the  $ER$  after tuning off the CW pump, although the  $ER$  could not return to the initial value of  $\sim 14.0$  dB. At  $P_p = \sim 160$  mW, there was still a slight increase in the  $ER$  ( $\sim 0.2$  dB) when the CW pump was off, reflecting the fact that the highly reduced GO at this power level was not yet fully reduced.

In Figure 5a-i, the dashed horizontal lines indicate the measured  $ER$ s for the hybrid MRR with 1 layer of GO after the chip was heated on a hotplate at various  $T_R$  (i.e., the same as those in Figure 2b, which were measured by scanning a CW light with a power  $\sim 1$  mW). These lines were plotted to compare the performance between localized heating using a CW pump and uniform heating of entire chip via a hotplate. As can be seen, despite using different methods to reduce the GO film, similar  $ER$ s can be achieved for the hybrid MRRs with GO films reduced to different degrees.

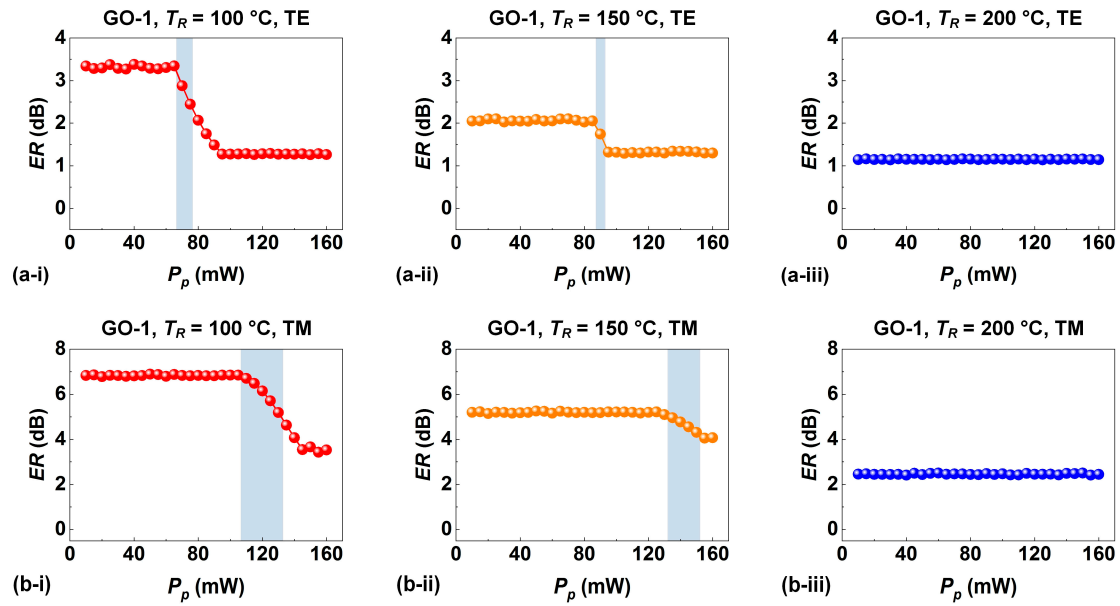
For TM polarization in Figure 5a-ii, reversible GO reduction was observed for  $P_p$  within the range of  $\sim 82 - 123$  mW. Compared to TE polarization, the lower power threshold  $P_{thres1}$  is higher and the power range for reversible reduction is larger for TM polarization. This reflects the relatively weak photo-thermal effects for TM polarization due to the significant anisotropy of the GO film, aligning with the experimental results in previous sections.

Figure 5b,c show the corresponding results for the hybrid MRRs with 2 and 3 layers of GO, respectively. For the hybrid MRR with 2 layers of GO, reversible GO reduction was observed for  $P_p$  within the range of  $\sim 36 - 58$  mW, and for the hybrid MRR with 3 layers of GO, it was observed for  $P_p$  within the range  $\sim 32 - 47$  mW. Compared to the results in Figure 5a, the hybrid MRR with a thicker GO film shows a decreased  $P_{thres1}$  and a smaller power range, reflecting the more significant photo-thermal effects in them.



**Figure 5.** Measured extinction ratio  $ER$  versus input CW pump power  $P_p$  for the hybrid MRRs with (a) 1, (b) 2, and (c) 3 layers of GO. In (a) – (c), (i) and (ii) show the results for TE and TM polarizations, respectively. The blue shaded areas indicate the power ranges associated with reversible GO reduction. The dashed horizontal lines indicate the  $ER$ s for the corresponding MRRs after the chip was heated at various temperatures  $T_R$  and measured by scanning a CW light with a power of ~1 mW.

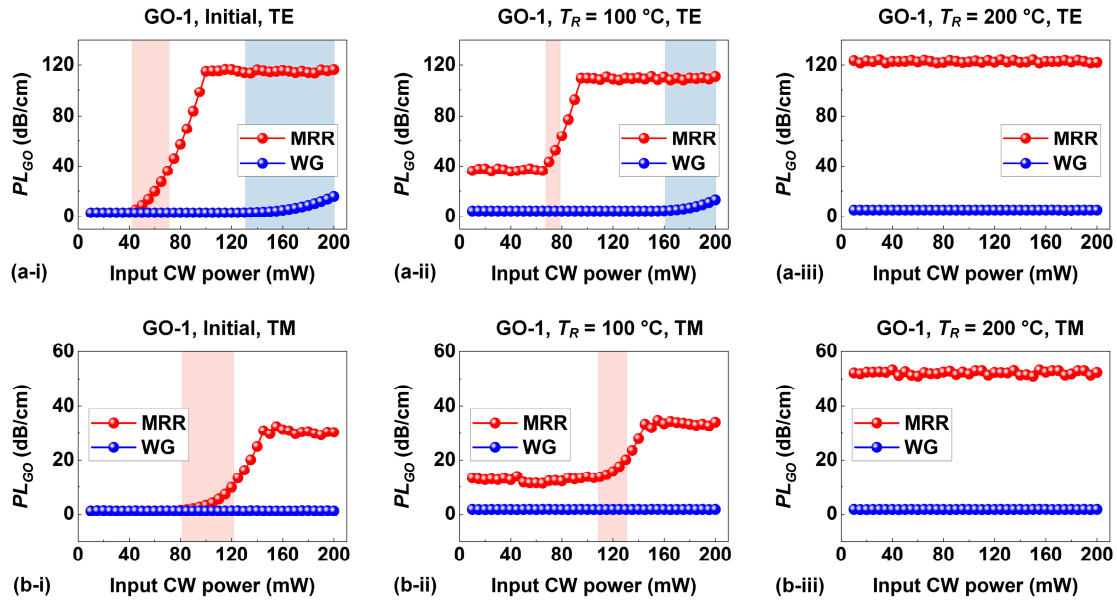
In Figure 5, before injecting the CW pump, the hybrid MRRs were coated with unreduced GO films. In Figure 6, we further characterize the performance for the hybrid MRRs with reduced GO films before injecting the CW pump. In Figure 6a,b, we show the measured  $ER$  versus  $P_p$  for the hybrid MRR with 1 layer of reduced GO for TE and TM polarizations, respectively. Prior to injecting the CW pump and measuring the transmission spectra, the chip was heated on a hotplate to obtain reduced GO at different reduction degrees. In each figure, (i) – (iii) show the results measured after the chip was heated at  $T_R = \sim 100$  °C,  $\sim 150$  °C, and  $\sim 200$  °C, respectively.



**Figure 6.** Measured extinction ratio  $ER$  versus input CW pump power  $P_p$  for the hybrid MRR with 1 layer of reduced GO for (a) TE and (b) TM polarizations. In (a) – (b), (i) – (iii) show the results measured after the chip was heated at temperatures of  $T_R = \sim 100^\circ\text{C}$ ,  $T_R = \sim 150^\circ\text{C}$ , and  $T_R = \sim 200^\circ\text{C}$ , respectively. The blue shaded areas indicate the power ranges associated with reversible GO reduction.

For  $T_R = \sim 100^\circ\text{C}$  in Figure 6a-i, reversible GO reduction was observed for  $P_p$  within the range of  $\sim 65 - 78$  mW. Compared to the results for unreduced GO in Figure 5a-i, the  $P_{thres1}$  is higher, and the power range is smaller. This indicates that unreduced GO was more easily reduced by the CW pump. In Figure 6a-ii, for reduced GO at  $T_R = 150^\circ\text{C}$ , reversible GO reduction was observed with an even higher  $P_{thres1}$  and in an even smaller range of  $\sim 87 - 93$  mW. For the reduced GO at  $T_R = 200^\circ\text{C}$  in Figure 6a-iii, no significant changes in the  $ER$  were observed, indicating that there was no obvious reversible GO reduction for highly reduced GO. These results further confirm that the reversible reduction behaviour becomes less obvious as the degree of reduction increases. Similar to Figure 5, the results for TM polarization in Figure 6b show higher  $P_{thres1}$  values and larger ranges for reversible GO reduction compared to the corresponding results for TE polarization in Figure 6a. At  $T_R = 200^\circ\text{C}$ , there were also no obvious changes in the  $ER$  for TM polarization.

In Figure 7, we compare the reversible GO reduction behaviours in GO-coated SiN MRRs and waveguides. In our measurements, the GO-coated MRRs had a radius of  $\sim 200\ \mu\text{m}$ , and the GO film length was  $\sim 50\ \mu\text{m}$  (*i.e.*, the same as those in Figures 5 and 6). On the other hand, the GO-coated waveguides had a total length of  $\sim 2$  cm, and the GO film length was  $\sim 1.4$  mm. Since the MRRs and the waveguides had different GO film lengths, in Figure 7 we compare the additional propagation loss induced by GO ( $PL_{GO}$ , in unit of dB/cm). For the hybrid MRRs, the  $PL_{GO}$  was calculated by using the scattering matrix method to fit the measured transmission spectra. For the hybrid waveguides, the  $PL_{GO}$  was calculated by dividing the GO-induced insertion loss by the GO film length. The  $PL_{GO}$  in Figure 7 was plotted as a function of input power. For GO-coated MRRs, the input power corresponds to the CW pump power  $P_p$  injected into the MRR's resonance (similar to those in Figures 5 and 6). Whereas for the GO-coated waveguides, the input power corresponds to the input CW power coupled into the waveguide.



**Figure 7.** GO-induced propagation loss  $PL_{GO}$  versus input CW power for a GO-coated SiN waveguide (WG) and a GO-coated SiN MRR for (a) TE and (b) TM polarizations. Both the WG and the MRR were coated with 1 layer of GO. In (a) – (b), (i) – (iii) show the results measured for a chip before heating (initial) and after the chip was heated at  $T_R = \sim 100^\circ\text{C}$  and  $T_R = \sim 200^\circ\text{C}$ , respectively. The blue and red shaded areas indicate the power ranges associated with reversible GO reduction for the WGs and the MRRs, respectively.

Figure 7a-i shows  $PL_{GO}$  versus input CW power for a hybrid MRR and waveguide, both coated with 1 layer of GO. For the hybrid MRR, the trend of  $PL_{GO}$  varying with input power matches the trend of  $ER$  varying with  $P_P$  in Figure 5a-i, with reversible GO reduction being observed within the same input power range of  $\sim 40 - 72$  mW. The  $PL_{GO}$  remained constant at  $\sim 3.0$  dB/cm for a low input power  $< 40$  mW, and at an input power of  $\sim 100$  mW it dramatically increased to  $\sim 115.2$  dB/cm. On the other hand, the  $PL_{GO}$  for the hybrid waveguide remained constant at  $\sim 3.0$  dB/cm for an input power  $< 120$  mW, followed by a gradual increase to  $\sim 15.8$  dB/cm when the input power reached  $\sim 200$  mW. In the input power range of  $\sim 131 - 200$  mW, reversible GO reduction behaviour was also observed. The comparison between the results for the hybrid MRR and waveguide reveals that there were more significant photo-thermal effects in the MRR due to resonant enhancement of optical intensity, which induced more significant changes to the GO film loss.

Figure 7a-ii,a-iii show the corresponding results for the hybrid devices with 1 layer of reduced GO at  $T_R = \sim 100^\circ\text{C}$  and  $\sim 200^\circ\text{C}$ , respectively. For both the hybrid MRR and waveguide, the input power range for reversible GO reduction narrows as the reduction degree increases – similar to the trend observed in Figure 6. At  $T_R = \sim 200^\circ\text{C}$ , no obvious GO reduction was observed for either the MRR or the waveguide, as evidenced by the absence of significant changes in the  $PL_{GO}$ . Figure 7b shows the corresponding results for TM polarization. Similar to that in Figures 5 and 6, the TM-polarized hybrid MRRs showed higher thresholds and larger power ranges for reversible GO reduction than comparable TE-polarized MRRs. On the other hand, all the hybrid waveguides in TM polarization exhibited no significant changes in  $PL_{GO}$  within our measured input power range, reflecting that there was no significant GO reduction even at a high input power of  $\sim 200$  mW.

## 7. Optical Bistability

Optical bistability featured by a steepened asymmetric transitional edge is a phenomenon arising from nonlinear thermo-optic effects. It has found wide applications in optical switches, memories, and logic devices [55–57]. In this section, we characterize the optical bistability in the fabricated GO-SiN MRRs. For the experiments performed in this section, we employed SiN MRRs with a radius of  $\sim 200$   $\mu\text{m}$ , and the length of the coated GO films was  $\sim 50$   $\mu\text{m}$ . We measured the output



power of a CW light with gradually varying input power. The initial wavelength detuning  $\delta$  between the CW light and the resonance wavelength of the MRR was defined as:

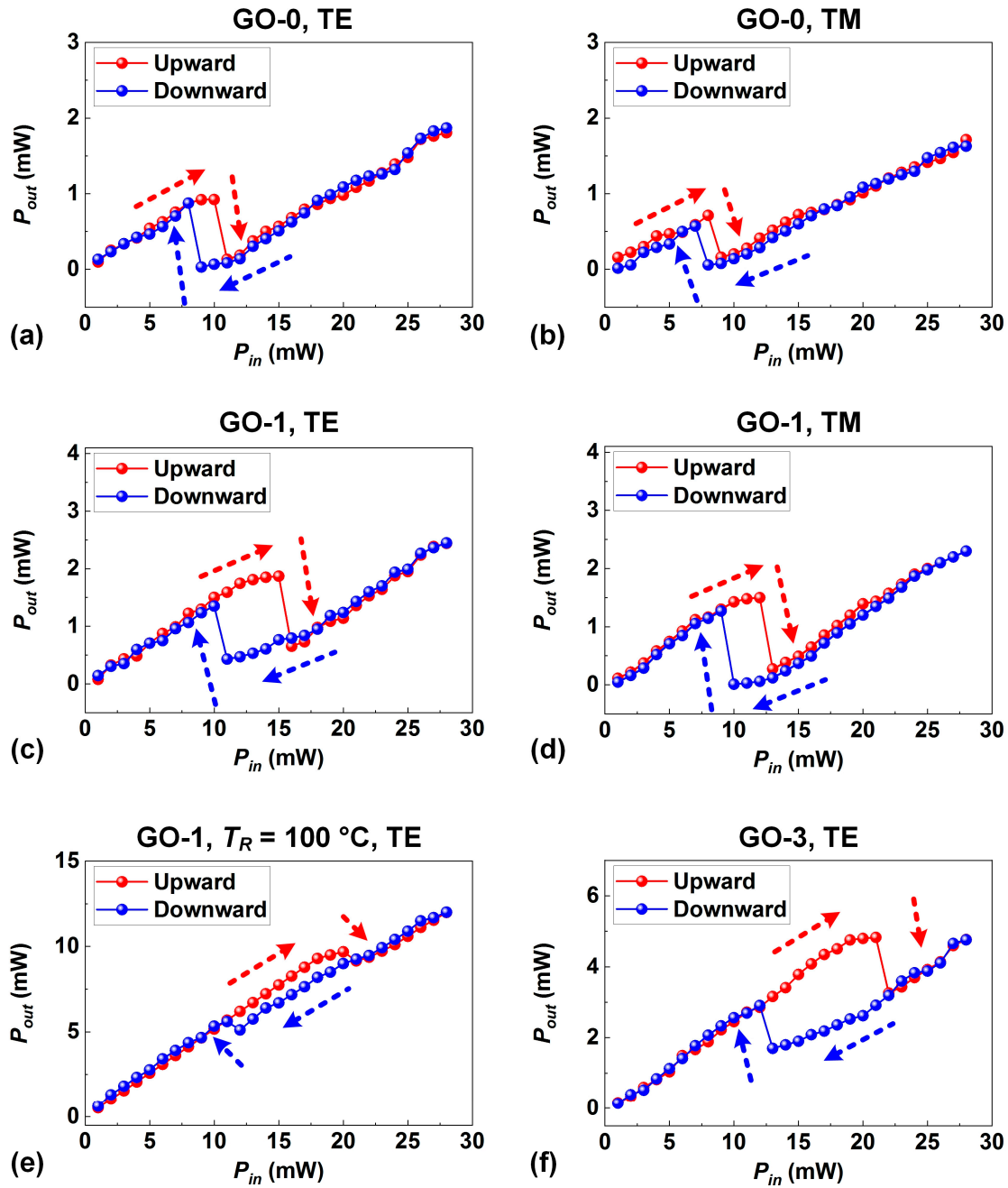
$$\delta = (\lambda_{laser} - \lambda_{res})/\Delta\lambda \quad (5)$$

where  $\lambda_{laser}$  is the wavelength of the input CW light,  $\lambda_{res}$  is the resonance wavelength of the MRR measured at a low input power of  $P_{in} = \sim 1$  mW (*i.e.*, the same as that in Figure 2a), and  $\Delta\lambda$  is the full width at half-maximum (FWHM) of the resonance. For comparison, we selected the same  $\delta = 0.2$  for all the measurements in this section.

Figures 8a and b show the measured output power  $P_{out}$  versus input power  $P_{in}$  for TE- and TM-polarized resonances of the uncoated SiN MRR, respectively. We first performed upward sweeping by gradually increasing  $P_{in}$  from  $\sim 1$  mW to  $\sim 28$  mW, followed by downward sweeping where  $P_{in}$  was gradually decreased within the same range. As can be seen, for both polarizations, the output power first gradually increased before experiencing a sudden drop toward a lower output power during the upward sweeping. In contrast, during the downward sweeping the output power first exhibited a gradual decrease before a sudden jump toward a higher output power. The existence of a hysteresis loop formed by the upward and downward sweeping confirms the presence of optical bistability in the SiN MRR. In principle, optical bistability manifests in the response of resonators when, under certain conditions, the output power produces multiple distinct solutions for a given input power [58]. As a result, the resonator can transition between the states corresponding to different solutions due to the influence of noise.

Figures 8c and d show the measured output power  $P_{out}$  versus input power  $P_{in}$  for TE- and TM-polarized resonances of a hybrid MRR with 1 layer of GO, respectively. The power range for the upward and downward sweeping was the same as those in Figure 8a,b. Compared to the uncoated SiN MRR, the GO-coated MRR exhibited a more open hysteresis loop as well as lower power thresholds for the jump, reflecting the enhanced optical bistability behavior in the hybrid MRR. Such enhancement can be attributed to more significant thermo-optic effects within the GO films, as confirmed by the experimental results in previous sections. Unlike similar hysteresis loops that observed for TE- and TM-polarized resonances of the uncoated MRR, the hybrid MRR showed a more open hysteresis loop for the TE-polarized resonance compared to the TM-polarized resonance. This further confirms that the enhanced optical bistability was induced by the 2D GO film that had significant material anisotropy.

Figures 8e and f show the corresponding results for the TE-polarized resonances of a hybrid MRR with 1 layer of reduced GO (at  $T_R = \sim 100$  °C) and a hybrid MRR with 3 layers of GO, respectively. We did not use the hybrid MRR heated at  $T_R = \sim 200$  °C due to its low  $ER$ , which prevented us from observing obvious power jumps in the hysteresis loop. Compared to the hybrid MRR with unreduced GO, the hybrid MRR with reduced GO exhibited a broader input power range and a smaller output power range within the hysteresis loop. The former indicates higher thermal nonlinearity for reduced GO film, and the latter was mainly resulting from the decreased  $ER$ . Similar trends in the hysteresis loop were also observed for the hybrid MRR with a thicker GO film, albeit with a minor reduction in the output power range due to a smaller decrease in the  $ER$ . This reflects the increased thermal nonlinearity for thicker GO films. These results have wide applications to GO based devices as well as other novel photonic platforms. [59–111] Ultimately this could be useful for both classical and quantum microcomb based applications. [112–183]



**Figure 8.** (a) – (b) Measured output power  $P_{out}$  versus input CW power  $P_{in}$  for TE- and TM-polarized resonances of an uncoated SiN MRR (GO-0), respectively. (c) – (d) Measured  $P_{out}$  versus  $P_{in}$  for TE- and TM-polarized resonances of a hybrid MRR coated with 1 layer of GO (GO-1), respectively. (e) Measured  $P_{out}$  versus  $P_{in}$  for a TE-polarized resonance of a hybrid MRR coated with 1 layer of GO after the chip was heated at  $T_R \approx 100$  °C. (f) Measured  $P_{out}$  versus  $P_{in}$  for a TE-polarized resonance of a hybrid MRR coated with 3 layers of GO. In (a) – (f), the red and blue arrows indicate the increasing and decreasing of the input power, respectively. The initial wavelength detuning was  $\delta = 0.2$ .

## 8. Conclusion

In summary, we systematically investigate a series of thermo-optic properties of 2D layered GO films by precisely integrating them onto SiN MRRs. We characterize the refractive index, extinction coefficient, thermo-optic coefficient, and thermal conductivity of 2D layered GO films with different layer numbers and reduction degrees. Experimental results show broad variation ranges for their thermo-optic properties with increasing reduction degree, including an increase of  $\sim 0.2280$  in the

refractive index, a ~36 times improvement in the extinction coefficient, a transition from a positive thermo-optic coefficient to a negative one, and an over 48-fold increase in the thermal conductivity. In addition, the 2D GO films exhibit significant anisotropic response for light in TE and TM polarizations, including a difference of ~0.1400 for the refractive index, a ratio of ~4 for the extinction ratio, a ratio of ~6 for the thermo-optic coefficient, and a ratio of ~18 for the thermal conductivity. Finally, we demonstrate reversible GO reduction and enhanced optical bistability in the hybrid MRRs induced by photo-thermal effects. These results reveal the versatile thermo-optic properties of 2D GO, which greatly enrich potential functionalities and devices that can be developed for a range of thermo-optic applications.

**Conflict of Interest:** The author declares no competing financial interest.

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