Nanosecond thermo-optical dynamics of polymer loaded plasmonic waveguides

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Abstract: The thermo-optical dynamics of polymer loaded surface plasmon waveguide (PLSPPW) based devices photo-thermally excited in the nanosecond regime is investigated. We demonstrate thermo-absorption of PLSPPW modes mediated by the temperature-dependent ohmic losses of the metal and the thermally controlled field distribution of the plasmon mode within the metal. For a PLSPPW excited by sub-nanosecond long pulses, we find that the thermo-absorption process leads to modulation depths up to 50% and features an activation time around 2ns whereas the relaxation time is around 800ns, four-fold smaller than the cooling time of the metal film itself. Next, we observe the photo-thermal activation of PLSPPW racetrack shaped resonators at a time scale of 300ns followed however by a long cooling time (18μs) attributed to the poor heat diffusivity of the polymer. We conclude that nanosecond excitation combined to high thermal diffusivity materials opens the way to high speed thermo-optical plasmonic devices.

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1. Introduction

Thermo-optical (TO) effects are to date the most common way to control dynamically integrated plasmonic components such as variable attenuators [1, 2] and low bandwidth modulators [3, 4]. Initially implemented by using waveguided long-range surface plasmon modes [5], more compact thermo-optical plasmon based devices relying on dielectric loaded surface plasmon waveguides [6–8] have been proposed [9–11] and demonstrated [12–15]. So far, and most often, the dielectric loading material is a polymer deposited onto a metal film or strip and in this respect the waveguides will be denoted hereafter as polymer loaded surface plasmon waveguides (PLSPPWs).

Single-mode PLSPPWs feature cross-sections with typical sub-micron dimensions enabling low-loss coupling with passive silicon-on-insulator circuitry [16–18]. The ability of integrating PLSPPW devices into a photonic passive circuitry has triggered the development and the demonstration of hybrid Si-PLSPPW thermally-activated routers for controlling high-bit rate traffic at the system level [19–21]. The key interest of the plasmon based devices in the context of thermo-optical control resides in the fact that the metal sustaining the plasmon mode can also be used as an heating electrode. However, photo-thermal activation of thermo-plasmonic devices is also an appealing approach. Indeed, this “all-optical” control offers the possibility to tune parameters such as the incident pump wavelength and/or polarization to optimize the TO devices activation [22].

Except when implemented with ultra-high quality factor resonators with corresponding narrow spectral range of operation [23], the main limitation of TO devices, whatever the material platform, is their moderate response times in the range of a few microseconds. For TO-PLSPPW routers integrated into a silicon on insulator circuitry, response times around 3-4 μs have been obtained [19, 20]. Although many dc or low-frequency characterizations of TO-PLSPPW devices have been reported, the dynamics of the thermo-optical response of these components has not been investigated in details. It is therefore the main objective of this work to analyze experimentally and numerically the PLSPPW TO dynamics down to the nanosecond time scale. For this purpose, we operate a photo-thermal excitation of the PLSPPW devices by using a Q-switched nanosecond pulsed visible (532nm) pump laser. The nanosecond pulse can be viewed, in the first approximation, as a delta-function excitation from which the overall TO dynamics of the PLSPPWs can be extracted. However, beyond PLSPPW TO dynamics, we also consider the use of nanosecond pulses for fast photo-thermal activation of our devices. In this context, the pulse is used as a pre-conditioning of the TO component by abruptly setting it into its hot state. Our approach can be viewed as the optical equivalent of the so-called “pre-emphasis driving” recently operated for fast reconfiguration of electrically driven silicon-based TO devices [24].

The study is organized as follows. The experimental setup based on a combined leakage radiation microscope and a fiber-to-fiber characterization scheme is briefly described in section two along with the fabrication process of our samples. We detail in the third section our model of the PLSPPW TO response relying on a numerical computation of temperature distributions for a nanosecond photo-thermal excitation coupled to an effective index method. In section four, we investigate the thermo-absorption of a PLSPPW mode traveling along straight waveguides irradiated by nanosecond pulses. We demonstrate depth of modulations as large as 50% and we find that the presence of the polymer impacts the thermo-absorption dynamics at the
sub-nanosecond scale. In this way, we show that the heat diffusion into the polymer over characteristic lengths around 20nm is detectable from the PLSPPW properties. Next, we consider in section five, the TO dynamics of photo-thermally excited PLSPPW racetracks resonators. We demonstrate an activation of the resonators at a time scale of 300ns followed however by a cool down characteristic time of about 18μs. By analyzing the origin of these performances from the thermal properties of our polymer-based configuration, we conclude that nanosecond photo-thermal excitation is a convenient approach for the development of high-speed TO plasmonic devices.

2. Experimental

Figure 1(a) shows a schematic view of the experimental setup we use for the characterization of the PLSPPW nanosecond photo-thermal excitation. The setup is built up around a leakage radiation configuration [25] relying on an inverted microscope equipped with a high numerical aperture objective (1.49NA) and a sensitive InGaAs camera. The samples mounted onto the microscope stage are excited by an infrared laser tunable over the C+L telecom bands. The infrared incident light is injected into a lensed-fiber producing a spot with a typical radius of 20μm focused onto the input grating coupler of the PLSPPW device under test (DUT). The signal at the output of the DUT is scattered by a grating coupler and collected by the output focuser. With this configuration, insertion losses for PLSPPW samples in the range of 25-30dB are typically achieved. A visible pump spot (532nm) produced by a Q-switched laser is directed at normal incidence onto the samples by means of a low magnification, long working distance objective. The laser has a pulse duration of 0.59ns (full width at half maximum), a repetition

Fig. 1. (a) Schematic view of the experimental setup combining a leakage radiation microscope and a fiber-to-fiber detection scheme. (b) and (c) Typical leakage radiation images recorded during the alignment procedure of the two lensed-fibers. (b) The infrared signal coupled by means of the left grating creates the output scattering spot shown by the arrow. For alignment purposes the output fiber is also coupled to an infrared source to generate the spot surrounded by the dashed perimeter. (c) At the end of the coarse alignment procedure, the spots generated by the input and output fibers are respectively located on the input and output grating couplers.
rate of 8.3kHz and a maximum energy per pulse of 6.5μJ. The thermo-optical activation of the PLSPPW devices is monitored by the infrared signal transmitted through the DUT. The output signal can be directed on two different photo-detectors. For short time scale measurements, the output signal is amplified with an erbium-doped fiber amplifier (EDFA), filtered to remove the spontaneous emission contribution and directed onto a 50GHz photo-diode. The voltage of the 50GHz photo-diode is next monitored by a real time 33GHz bandwidth oscilloscope triggered by a visible photo-diode receiving a tiny fraction of the pump signal. For long time-scale observations (in the range of several μs), the output signal is directed onto a 250MHz (2ns rise-time) sensitive photo-diode without optical amplification connected to a 1GHz bandwidth oscilloscope. Figures 1(b) and 1(c) show typical leakage radiation microscope images of a straight 100μm-long PLSPPW. The main interest of leakage radiation microscopy in this study is to provide a very convenient way for the optimization of the input and output coupling at the cost however of the use of transparent substrates. The samples we use in all the following were fabricated by electron beam lithography using a negative tone resin (All-Resists AR-N7500) spin-coated at a speed of 1800 rotations per minute onto a gold film with a thickness of 70nm deposited onto a 170μm thick glass substrate. Once deposited, the resin layer with a typical thickness of 600nm was exposed to the electron beam at a dose of 170μC/cm² under an acceleration voltage of 20kV and eventually dissolved in a mixture of developer (all-resists AR 300-47: 4 vol) and DI water (1 vol) for 5 minutes.

3. Thermo-optical modeling

When a nanosecond pulse irradiates a metal film, the free-electron gas and the lattice are at thermal equilibrium after a characteristic time (typ. 10ps) much shorter than the pulse duration. The use of a two-temperature model considering the lattice and the electron gas temperature separately is then usually not necessary in the nanosecond regime. In addition, if the characteristic heat diffusion times into the system are larger than phonons relaxation times (1ps), the classical Fourier’s law holds and the thermal problem can be solved in the framework of classical heat diffusion theory. For simple configurations such as a semi-infinite absorbing medium illuminated by a pulsed gaussian beam, analytical solutions exist. However, in our situation, these approximate solutions are not sufficient and a numerical approach is necessary. Numerically, we consider the two-dimensional (2D) situation schematically shown in Fig. 2(a). The PLSPPW we consider is excited by a two-dimensional pulsed beam with a gaussian intensity distribution in space (along x-axis) centered onto the z-axis. The pulse has also a gaussian temporal profile. The temperature distribution of the system under photo-excitation is obtained by solving the time-dependent heat diffusion equation:

$$\rho(\vec{r})C_p(\vec{r})\frac{\partial T(\vec{r},t)}{\partial t} = \tilde{Q}_E(\vec{r},t) + \vec{V} \cdot (k(\vec{r})\nabla T(\vec{r},t))$$ (1)

where $\rho$, $C_p$ and $k$ are the position dependent density, specific heat and thermal conductivity of each materials of the system. The source term $\tilde{Q}_E$ is the volume power density created by the absorption of the incident pulse and is given by [26]:

$$\tilde{Q}_E(\vec{r},t) = \frac{1}{2}\omega \varepsilon_0 \varepsilon''_m \bar{E}^2(\vec{r},t)$$ (2)

where $\varepsilon''_m$ denotes the imaginary part of the relative dielectric function of gold and where $\bar{E}(\vec{r},t)$ is the electric field for observation points located within the gold film. Figure 2(b) shows a typical example electric field intensity distribution computed with the differential method [27] in the case of 2D gaussian beam with a waist $w_0 = 10\mu m$ illuminating a PLSPPW. The source term $\tilde{Q}_E(\vec{r},t)$ can be obtained readily from this intensity distribution and used to solve Eq. (1). We
compute the temperature distribution by using a non-regular rectangular mesh finite-difference implicit scheme with non-uniform time steps ranging from 50ps to 100ns depending on the delay with respect to the excitation pulse. Figures 2(c) and 2(d) show typical temperature distributions computed at different times in the case of a 2D nanosecond pulse with temporal characteristics similar to the experimental case (pulse duration FWHM duration=0.59ns, repetition rate 8.3kHz) and a central fluence (at $x=0$) of 4.8 mJ/cm². Once the temperature is known, the refractive index at each point is computed from the thermo-optical coefficients (TOC) of each material listed in table 1 along with their thermal parameters. Next the PLSPPW is divided into ten slices and the average refractive index of these slices is used to compute the complex propagation constant of the plasmonic waveguide mode from the effective index method [6] implemented on the basis of the reflection pole method [28]. In this way, we obtain the complex propagation constant of the PLSPPW mode as a function of temperature from which the TO dynamics can be extracted.

4. Thermo-absorption dynamics of PLSPP waveguides

The thermo-modulation of the signal intensity transmitted through a straight PLSPPW is mediated by the temperature dependence of the PLSPPW mode damping constant. Many works have been devoted to the thermally induced change of the real effective index of PLSPPWs (thermo-optical effect) but so far and to the best of our knowledge, none of them have addressed the
temperature dependence of the PLSPPW mode imaginary effective index (thermo-absorption effect). This dependence deserves a careful examination as it impacts the performances of PLSPPW devices in practical applications. Waveguiding in a PLSPPW results from the combined actions of metal and polymer from which the vertical and lateral field confinement of the guided mode originates respectively. These two materials are then expected to contribute to the thermo-optical properties of a PLSPPW. At this stage, it is instructive to refer to the situation of a SPP mode traveling at an interface between a metal and a dielectric. At telecom wavelengths, metal of interest for plasmonic applications are such that $|\varepsilon'_m| >> \varepsilon''_m$ where $\varepsilon'_m$ and $\varepsilon''_m$ are the real and imaginary part of the relative dielectric function of the metal. The relative dielectric function $\varepsilon_d$ of typical dielectric media such as standard polymers is also small compare to $|\varepsilon'_m|$ in such a way that the temperature derivative of the SPP mode damping constant $k''_{spp}$ is given by [29]:

$$\partial_T k''_{spp} = k''_{spp} \left( \frac{3}{2} \frac{\partial_T \varepsilon_d}{\varepsilon_d} + \frac{\partial_T \varepsilon''_m}{\varepsilon'_m} - 2 \frac{\partial_T \varepsilon'_m}{\varepsilon'_m} \right)$$

where the notation $\partial_T$ is used in place of $\partial / \partial T$. The metal contribution to the SPP thermo-absorption is not only mediated by the ohmic losses (through $\partial_T \varepsilon''_m$) but also by the temperature-dependent $\varepsilon'_m$. At telecom wavelengths, where the dielectric function of noble metals are dominated by the free-electron contribution, the ohmic losses depend on temperature mostly through the electron-phonon scattering rate whereas a change of the free-electron density resulting from a thermally induced lattice volume expansion is at the origin of the $\partial_T \varepsilon'_m$ coefficient. The dielectric medium contributes also to the SPP thermo-absorption through the change of the SPP field confinement within the metal which depends on $\partial_T \varepsilon_d$. For a PLSPPW with a complex geometry, the respective contribution of the metal and the dielectric to the thermo-absorption effect is difficult to estimate. It is then useful to investigate first a situation where the dielectric medium contribution is expected to be negligible such as in the case of a SPP mode traveling at a gold/air interface.

The Au/air SPP is coupled in and out using dielectric gratings separated by a distance of 100μm [Fig. 3(a)] and designed according to the approach described in ref. [30]. A very similar configuration has been previously used to extract the thermo-optical coefficients of gold given in table 1 [31]. Typical leakage radiation images of the SPP jet launched by the input grating coupler are shown in Figs. 3(b) and 3(c) with the pump spot respectively off and on. Figures 3(d)-3(f) show the oscilloscope traces at different time scales of the Au/air SPP signal thermo-modulated by a gaussian pump beam with a waist of 50μm. For this experiment, the average pump power was 5.6mW ($E_p = 0.68 \mu J / pulse$) corresponding to a fluence at the center of the gaussian beam of $F = E_p / (\pi w_0^2) = 8.5 mJ / cm^2$ far below the damage threshold (140mJ/cm² [32]). The normalized signal plotted in Figs. 3(d)-3(f) is defined as $S_N = \frac{1 - V(t)}{V_M}$ where $V(t)$ is the ac-coupled voltage measured at the output of the photodiode and where $V_M$ denotes the maximum modulation amplitude. For the 5.6mW pump power,
Fig. 3. (a) Scanning electron microscope image of the in and out grating couplers (scale bar=50μm). (b) (resp. (c)) Typical leakage radiation microscope images of the plasmon jet (1540nm) propagating at the Au/air interface with the pump beam off (resp. on, cut-on filter off). The pump spot features a gaussian intensity distribution in $I(r) = I(0) \exp\left(-\frac{r^2}{w_r^2}\right)$ with a waist of $w_r = 50\mu$m. (d)-(e)-(f) Observation of the thermo-absorption of the SPP signal under the nanosecond excitation at different time scales. In (d) and (e), the dashed lines are the experimental signal whereas the solid lines are the computed thermo-absorption profiles for an interface SPP. The dashed-dotted line in (d) shows the temporal profile of the incident pulse used in the calculations.

$V_M$ is only 3.7% of the cold state transmitted SPP signal for a maximum thin film temperature rise at the center of the pumped beam evaluated at 87K by solving numerically the heat diffusion equation in cylindrical coordinates [31]. For the Au/air SPP mode, the modulation amplitude is given by $V(t) \propto I(0)(1 - \exp(-2\pi k'_{sp} \int_0^{\Delta T_f(y',t)} dy'))$ [31] where $I(y_{out})$ is the electric field intensity of the SPP jet in the cold state at the location of the output grating $y_{out}$ and $\Delta T_f(y',t)$ is the temperature rise in the gold film along the SPP propagation pathway. The argument of the exponential in the definition of $V(t)$ being small, the normalized signal $S_N$ can be approximated by:

$$S_N = 1 - \frac{\int_0^{y_{out}} \Delta T_f(y',t) dy'}{\max(\int_0^{y_{out}} \Delta T_f(y',t) dy')}$$

Thus, for a SPP mode traveling at the interface between the metal and a dielectric with negligibly small TO coefficient, the thermo-absorption of the transmitted SPP signal is characteristic of the thin film temperature change accumulated over the SPP mode propagation distance. By using the profile displayed in Fig. 3(d), we measure a 90%-10% fall-time of $\tau_F = 1.0$ns indicating that the thin film temperature changes at the time scale of the pulse duration. On the other hand, the 10%-90% rise-time evaluated from the stretched exponential fit [33] of the experimental profile displayed in Fig. 3(f) is measured at $\tau_R = 3.1$μs. As discussed in the following, the value of $\tau_R$ depends on the ability of the system to dissipate heat and can be decreased down to the sub-μs regime by choosing a proper design. The thermo-absorption of the Au/air SPP is compared in Figs. 3(d) and 3(e) to the computed TO response following the approach described in the previous section. Our model underestimate the fall-time by about 0.5ns compared to the experimental value. On the other hand, we note a very good agreement between experimental and computed profiles for the cooling process. The discrepancy of the computed and experimental results at very short time scale (a few nanoseconds) is not fully understood but could be related to a specific heat diffusion regime at the interface between the metal film and the glass substrate. Despite these differences, we note that our model can predict quantitatively the dy-
namics of the thermo-absorption process (at least for times larger than a few nanoseconds after the pulse) and then can be safely used to anticipate the TO response of other plasmonic devices at the sub-μs scale. In Fig. 3(e), we observe a small increase of the transmitted signal right before the thermally induced abrupt signal drop. This increase, which is not visible onto the computed profile, could be due to a slight improvement of the in and out coupling conditions of the polymer gratings for a moderate temperature rise.

We consider now the situation of a straight PLSPPW photo-thermally excited by the nanosecond pulsed beam. The excitation conditions are the same as for the interface SPP mode. Figures 4(a)-4(c) show respectively a scanning electron microscope image of the straight PLSPPW as well as a typical radiation leakage image of this waveguide with the pump spot off and on. Although we focus in this work onto the dynamics of the TO response of PLSPPWs, it is worth to consider first the curve displayed in Fig. 4(d) showing the depth of modulation of the signal collected at the output of the PLSPPW as a function of the average pump power. For an average pump power of 5.6mW, the depth of modulation is about 38% of the cold state signal amplitude, up to ten times larger than in the case of the interface SPP. This difference can be understood from the larger field confinement within the metal (and accordingly the shorter propagation length) for the PSLPPW mode compared to the SPP mode [31]. Figures 4(e)-4(g) show the normalized signal $S_N$ recorded at different time scales. The solid lines are computed profiles whereas the dashed lines are experimental profiles. The dash-dotted line in Fig. 4(e) is the profile of the excitation pulse used in the calculations.

Fig. 4. (a) Scanning electron microscope image of a typical PLSPPW equipped with grating couplers (scale bar=40μm). (b) (resp. (c)) Leakage radiation microscope images of the PLSPPW mode (1530nm) propagating at the Au/air interface with the pump beam off (resp. on, cut-on filter off). The excitation conditions are the same as in Fig. 3. (d) Depth of thermo-absorption of the PLSPPW mode as a function of the incident average pump power. The depth of modulation is defined with respect to the signal level in the cold state. (e)-(f)-(g) Experimental and computed thermo-absorption of the PLSPPW signal under nanosecond excitation at different time scales. The solid lines are computed profiles whereas the dashed lines are experimental profiles. The dash-dotted line in Fig. 4(e) is the profile of the excitation pulse used in the calculations.
Figs. 4(e) and 4(g), we find that the fall-time for the thermo-absorption of the PLSPPW mode is $\tau_F = 1.7\text{ns}$ and the sub-$\mu$s rise-time drops at $\tau_R = 800\text{ns}$ respectively two times longer and four-fold shorter than in the case of the interface SPP mode. For a PLSPPW, the normalized signal $S_N$ does not follow anymore the dynamics of the metal film temperature but results from the combined contributions of the metal and polymer as illustrated by Eq. (3). The activation time for the thermo-absorption of PLSPPWs can be explained from a slower thermalization of the gold film in the presence of the polymer but also from its negative TOC. The typical heat diffusion length for the pulse duration $\tau_p$ is $L = 2\sqrt{\alpha \tau_p}$ [34] where $\alpha$ is the thermal diffusivity ($\alpha = k/(\rho C_p)$) of the material of interest. In our situation ($\alpha_{poly} = 8.3 \times 10^{-8}\text{m}^2\text{s}^{-1}$, $\tau_p \simeq 1\text{ns}$), the heat diffusion length into the polymer is around 20nm. At the nanosecond scale, the temperature rise of the polymer generates a thin low index layer in contact with the metal film leading to a decrease of the PLSPPW mode field confinement into the metal and a corresponding increase of its propagation length. Hence, during the heating cycle, the increase of the metal losses is partly compensated by the PLSPPW mode field delocalization resulting in a longer activation time of the PLSPPW thermo-absorption process than for the Au/air SPP mode. At longer times, after the pulse, the temperature of the metal film drops and the propagation length of the PLSPPW mode increases accordingly. However the rise-time $\tau_R$ for PLSPPWs being about four times shorter than for the Au/air SPP, we conclude that the polymer still contributes to the increase of the PLSPPW damping distance during the cooling cycle. Once again, we note that our model fails to capture the dynamics of the thermo-absorption during the first nanoseconds whereas computed and experimental profiles are in good agreement for times larger than 5ns after the pulse. From these results, we conclude that our model is reliable for time scales larger than 10ns which is still of key interest to predict the performances of high bandwidth thermo-plasmonic devices. The assessment of the characteristic time for the polymer contribution to the TO PLSPPW dynamics is difficult to complete from thermo-absorption experiments owing to the metal contribution and requires the use of frequency resonant devices.

5. Thermo-optical dynamics of a PLSPPW ring resonator

We have identified in the previous section the role of the polymer in the dynamics of the thermo-absorption of a PLSPPW mode by comparison with a reference Au/air SPP mode. To push further this qualitative analysis, we consider now the thermo-optical response of a PLSPPW based resonator. Prior to the discussion of the experimental results, we evaluate the respective contribution of the metal and polymer in this context by considering once again the situation of the interface SPP mode. At telecom wavelengths, the temperature derivative the SPP phase constant can be approximated by:

$$\partial_T k'_{spp} \simeq \frac{k'_{spp}}{2(\varepsilon'_m + \varepsilon_d)} \left( \frac{\partial_T \varepsilon'_m}{\varepsilon'_m} + \frac{\partial_T \varepsilon_d}{\varepsilon_d} \right). \quad (5)$$

The metal contribution to the PLSPPW TO coefficient is roughly two orders of magnitude smaller than the dielectric contribution in such a way that the thermally-controlled properties of a PLSPPW based resonator can be interpreted in terms of polymer properties only. The resonators we consider are racetrack resonators excited by a straight bus waveguide [Fig. 5(a)]. Plasmonic switches relying on optimized racetrack resonators have been already demonstrated in ref. [14] and the opto-geometrical parameters we use are directly inspired from this previous study. The resonator we consider features well pronounced resonances as illustrated by the leakage radiation images recorded at 1560nm [Fig. 5(b)] and 1565nm [Fig. 5(c)]. The spectral properties of the resonator have been characterized by exciting the bus waveguide with a broadband spontaneous emission source and by collecting the output signal with an optical spectrum analyzer. The normalization of this first spectrum by the spectrum of a reference waveguide
Fig. 5. (a) Scanning electron microscope image of the racetrack shaped resonator coupled to a straight bus waveguide (scale bar=40μm). The radius of the resonator is R=5.5μm, straight interaction length with the bus waveguide is 6μm long and the nominal gap between the resonator and the bus waveguide is 250nm. (b) (resp. (c)) Leakage radiation microscope image of the resonator at 1560nm (resp. 1565nm). (d) Cold state spectrum of the resonator. (e) (resp. (f)) Thermo-optical response of the resonator under ns excitation for blue-detuned (resp. red-detuned) wavelengths compared to the cold state resonance of 1538nm. The photo-excitation is achieved with a large pump spot exciting simultaneously the resonator and the bus waveguide.

(without resonator) leads to the spectrum displayed in Fig. 5(d). We focus onto the resonance centered at 1538nm located within the optimum amplification band of the EDFA. The resonator is photo-thermally excited using a pump spot with a waist of 50μm (average power 5mW) illuminating the bus waveguide and the resonator simultaneously. Figures 5(e) and 5(f) show the time resolved signal modulation for wavelengths located respectively on the blue and red side of the central resonance wavelength (in the cold state). For wavelengths corresponding to a blue detuning, the photo-excitation leads to an abrupt drop of the signal followed by a slower signal rise. For those wavelengths the modulation is negative indicating that the signal in the hot state is lower than in the cold state. For a red detuning, we observe first a very sharp signal drop followed by a positive modulation of the signal at longer time scales. These results clearly indicate that the resonator has been photo-activated by the nanosecond pulse. Indeed, the polymer we use having a negative TOC, the spectrum of the resonator blue-shifts in the hot state causing the positive (resp. negative) modulation for red detuned (resp. blue detuned) wavelengths. For red-detuned wavelengths, the sharp drop of the signal is due to the thermo-absorption of the PLSPPW mode investigated in the previous section. For blue detuned wavelengths, the negative modulation of the signal can be unambiguously attributed to the heating of the polymer and not to the ohmic losses of the heated metal film. Indeed, the time scale for the signal rise after

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the excitation in Fig. 5(e) is much slower for 1532nm and 1534nm than for 1550nm for which the spectral response of the resonator is flat. With this experiment, we show that nanosecond photo-thermal activation of plasmonic devices is possible and could be of practical interest for high-bandwidth thermo-optical devices. However in our situation neither the amplitude nor the dynamics of the optical response is sufficient to target such an application. Indeed, by increasing the average pump power up to 10mW slightly below the damage threshold of the resonator, we could achieve a positive depth of modulation of $\Delta T = 25\%$ at 1541nm, corresponding to a resonance detuning of only 0.6nm, insufficient for switching with a low quality factor plasmonic resonator. Nevertheless, future designs of thermo-plasmonic devices activated in the nanosecond regime can greatly benefit from an analysis of the limited performances of the polymer based devices.

Figures 6(a) and 6(b) show the TO response of the photo-activated resonator at different time scales. The thermo-absorption of the PLSPPW mode occurring at a scale of 2ns, the abrupt drop of the signal in Fig. 6(a) is a reliable indication of the pump pulse arrival time from which the dynamics of the resonator can be accurately measured. The profile displayed in Fig. 6(a) indicates that the characteristic activation time for the PLSPPW resonator is $\tau_A = 280ns$ followed by a much longer relaxation time. We measure a 90%-10% fall time of $\tau_f = 18\mu s$ on the profile shown in Fig. 6(b). The average temperature in the polymer and the metal film are plotted in Fig. 6(c) for the PLSPPW excited in the same conditions as in Fig. 2. The polymer slice in contact with the metal film, which is expected to contribute dominantly to the TO properties of the PLSPPW, reaches a maximum temperature (averaged over a thickness of 125nm) of only one fourth of the maximum metal film temperature. For slices at larger distances from the metal film, the temperature is maximum after a time characterizing the diffusion speed of the thermal pulse. The time dependent PLSPPW effective index $n_{eff} = k'/k_0$ ($k_0 = 2\pi/\lambda_0$, $\lambda_0 = 1541nm$) plotted in Fig. 6(d) has been obtained by means of the effective index method with the refractive index of each polymer slice evaluated from the temperature profiles of Fig. 6(c). The minimum effective index is reached 200ns after the excitation pulse, in reasonable agreement with the experiment given that no adjustable parameter is used for this calculation. The thermo-optical activation of the PLSPPW occurs at sub-$\mu$s time scale whereas the cooling time is clearly the main limitation to high-speed operation. Indeed, the cooling time for the thermo-optical response of the ring resonator is about 20 times larger than the thermo-absorption relaxation time along a straight PLSPPW. This difference results from the fact that the thermo-optical response depends upon the polymer temperature only whereas for thermo-absorption, the temperature of the metal film plays the dominant role. Beyond numerical modeling, the physical quantities of interest for improved TO performances are more conveniently identified from an approximate analytical model. When a short pulse with a fluence $F$ is fully absorbed at $t = 0$ within a very thin layer at the surface of a semi-infinite medium occupying the $z > 0$ half-space, the temperature rise in the material at a distance $z$ from the surface is approximated by [35]:

$$\Delta T(z,t) = \frac{F}{\epsilon \sqrt{\pi t}} \exp\left(-\frac{z^2}{4\alpha t}\right)$$

where $\epsilon = \sqrt{k\rho C_p}$ is the thermal effusivity of the semi-infinite medium. We assume that the TO properties of the PLSPPWs can be analyzed from the temperature of a characteristic polymer slice located at a distance $z_{eff}$ from the surface of the metal film. For typical PLSPPW heights of 500nm, a reasonable choice for this effective distance is around $z_{eff} = 250nm$. According to Eq. (6), the temperature at $z_{eff}$ reaches its maximum value $\Delta T_{max}(z_{eff}) = F/($$\rho C_p z_{eff}^2) at t = \tau_H = z_{eff}^2/(2\alpha) = 280ns$ in perfect but fortuitous agreement with the experiment. From this approach, we conclude however that a high thermal diffusivity of the TO active medium is the key parameter for the fast activation time of PLSPPW devices. The average temperature of the
Fig. 6. (a) Photo-thermal excitation of the resonator for a red-detuned signal wavelength (1541nm). The photo-excitation is achieved by using a large spot ($w_r = 50 \mu m$) exciting the resonator and the bus waveguide (average power 7mW). The abrupt drop of the signal at $t = 0$ results from the thermo-absorption of the bus-waveguide mode and indicates the arrival time of the incident pulse. The activation (or heating) time of the resonator is 280ns. Long-time scale TO response of the resonator pump by a focused beam ($w_r \approx 5 \mu m$) (see the inset) with an average power of 150μW. With the local excitation of the resonator, the thermo-absorption is not observed anymore. The characteristic cooling time is 18μs. (c) Comparison of the average temperature into the gold film and polymer slices of the PLSPPW with a thickness of 125nm for the photo-excitation by a nanosecond pulse (0.6ns FWHM) arriving onto the PLSPPW at $t = 5ns$. (d) Computation of the effective index of the PLSPPW mode from the temperature profiles displayed in (c). (e) Comparison of the average temperature profile of the first polymer slice (125nm) in contact with the metal film and different fit models inspired by Eq. (6).
polymer slice in contact with the gold layer computed numerically is compared to fit models inspired by Eq. (6) in Fig. 6(d). We note that during the cooling cycle, the contribution of the exponential term in Eq 6 can be neglected in the first approximation. From this further approximation, one can show that the typical cooling time $\tau_C$ needed for the temperature at $z_{eff}$ to decrease from 90% to 10% of $\Delta T_{max}(z_{eff})$ is given by $\tau_C \sim \frac{2}{\pi \rho C_P} z_{eff}^2 \alpha$. Although this equation leads to a cooling time about five-fold overestimated compared to the experiment, the diffusivity $\alpha$ appears to be one again the main parameter for controlling the cooling dynamics of the system. However, the response time is not the only parameter that should be considered in a practical device. As pointed out recently in ref. [36], the dynamics of electrically driven TO PLSPPW devices is improved at the expense of larger activation powers. For photo-thermal excitation, such an increase can be minimized by a stronger focusing of the pump beam at least in the case of small footprint devices. In addition, Eq. (6) indicates that the heating efficiency scales as the inverse of the thermal effusivity of the material. Combining this requirement to the large thermal diffusivity needed for high-speed operation, we conclude that the optimum material in this context should have a large thermal conductivity $k$ and a small $\rho C_P$ product. The above analysis has been conducted considering heat diffusion in the TO active medium whereas in the experimental situation the role of the substrate is also critical. The conclusions given for the TO material also applies for the substrate which should feature a high thermal diffusivity as well. In addition, the thermal diffusivities and effusivities of the TO medium and the substrate should also be close in order to prevent a poor heating of the TO medium as in our configuration [Fig.6(c)] where heat diffusion is four times faster in the glass substrate than in the polymer load. On the basis of this analysis, we conclude that PLSPPW based TO devices operating in the sub-μs regime will be difficult to implement with standard polymers mostly due to their poor thermal diffusivity. In spite of their thermal properties, it is worth to note that sub-μs switching operations with PLSPPW based TO devices are achievable at the cost however of the implementation of sophisticated configurations such as a push-pull Mach-Zehnder interferometer. Indeed, for the latter, the switching speed is given by the typical activation time of each arm in the interferometer (in the range of 300ns in our case) whereas the relaxation time (several μs with PLSPPWs) imposes the switch latency [37]. Nevertheless, provided that high-speed TO devices are targeted, the polymer and the glass substrate used in this study must be replaced by highly thermally diffusive materials such as semi-conductors. For example, heat diffusivity of silicon ($\alpha \approx 8 \times 10^{-5} \text{m}^2\text{s}^{-1}$) is about three orders of magnitude larger than the polymer used in this study suggesting that the bandwidth of the TO semi-conductor loaded plasmonic devices could be extended up to several MHz.

6. Conclusion

In summary, we have experimentally and numerically investigated the dynamics of the thermo-modulation of PLSPPW devices photo-excited by nanosecond pulses. By operating a fiber-to-fiber detection scheme, we have demonstrated a response time for the thermo-absorption of the PLSPPW mode in the nanosecond regime at the scale of the pulse duration. Whatever the time scale, we have shown that the thermo-absorption of the PLSPPW mode is mediated by the temperature-dependent metal ohmic losses but also by the field distribution of the PLSPPW mode into the metal controlled by the polymer TOC. For a negative TOC, we observe a sub-μs thermo-modulation characteristic time (fall-time+rise-time) about four-fold shorter than the cooling time of the metal film itself. In addition, we find that the thermo-absorption amplitude for a PLSPPW mode is about 10 times larger than for a Au/air interface SPP photo-excited in the same conditions. On the basis of these results, we conclude that the thermo-absorption effect significantly impacts the performances of the PLSPPW based TO devices. Next, we have considered the thermo-optical response of PLSPPW racetrack resonators featuring well pro-
nounced resonances. By choosing a signal wavelength either blue or red detuned compared to the cold state resonance, we have shown that the nanosecond pulse can activate the resonator at a time scale of 300ns however followed by a characteristic cooling time of about 18μs in our configuration. The slow TO dynamics of these resonators is attributed to the poor thermal diffusivity of both the polymer and the glass substrate used in this study. In spite of these poor thermal performances, we note that the nanosecond photo-thermal excitation is convenient for sub-μs activation which is the key feature for the fast pre-conditioning of the TO devices. Finally, it should be also pointed out that, as long as the metal of the plasmonic devices is used as the heat transducer for electrical or optical driving, the TO plasmonic devices will suffer from the detrimental effect of thermo-absorption of the plasmon mode and from the time delay imposed by heat diffusion from the metal to the thermo-optically active material. Interestingly, we note that photo-thermal excitation offers the unique possibility to circumvent all these limitations by localizing the absorption of the incident light within the TO active medium rather than in the metal. By combining the nanosecond pulsed photo-thermal approach to the use of inorganic materials featuring all high thermal diffusivity, TO plasmonic devices with several MHz bandwidth turn to be achievable. Actions for the fabrication of such devices are currently in progress and will be reported elsewhere.

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