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ABSTRACT
All-optical control of Surface Plasmon Polaritons (SPPs) can switch light with high speed and a large signal to noise ratio. We demonstrate 25 ps-time scale switching of continuous wave light by pump pulses copropagating in the same single mode fiber at different wavelengths near 1550 nm. The switching is due to hot carriers from the SPPs in a 45 nm-thin gold coating on the fiber cladding. The SPPs are generated by pump pulses coupled from the core to cladding modes by a tilted fiber Bragg grating (TFBG). Hot carriers modify the complex permittivity of the metal coating on a time scale of the order of picoseconds and hence the cladding mode resonance wavelengths of the TFBG. A probe light signal copropagating in the same fiber can therefore be modulated by the transmission resonance shifts. With 25 ps pulses at 1540.4 nm and 1 MHz and 50 mW average power, the modulation depth of a CW probe at 1543.4 nm copropagating in the core reached 4.5\% ± 1\% with a pulse width broadened to 56 ps. Under these conditions, the pump power density was 0.147 GW/cm\(^2\) in the metal layer, for a conversion efficiency as high as 30\% ± 7\% per GW/cm\(^2\). Since many other plasmonic and nonlinear active materials can be deposited on fiber claddings, we believe that this very simple all-fiber configuration to perform all-optical switching of core-guided light in single mode fibers by plasmon-modulated resonances has strong potential applications in studies of light-matter interactions over fast and ultrafast time scales.

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I. INTRODUCTION
Surface Plasmon Polaritons (SPPs) provide an effective way to confine electromagnetic fields to sub-micrometer thick layers near the surface of metals, and excitation of SPP at femtosecond time scales can initiate a cascade of linear and nonlinear processes with multiple outcomes such as nonradiative decay to carriers and phonons and radiative decay via emission of photons.\textsuperscript{1–4} Such processes have opened new avenues toward the realization of ultrafast nanophotonic devices for all-optical signal processing in high speed telecommunication networks, but one of the major challenges is how to achieve fast and efficient switching or modulation of light by SPPs without excessive insertion losses in fiber or optical waveguide systems. In earlier work, plasmonic devices combining phase control in external materials such as semiconductors\textsuperscript{5,6} or organic films\textsuperscript{7} achieved light switching or modulation but on relatively long time scales of microseconds because of dominant thermal processes. When generating SPPs by interband transition pulsed pumping on the other hand, hot carriers are generated on femtosecond time scales in the metal and they strongly modify the complex permittivity on the same time scale.\textsuperscript{8–12} Such changes can then be used to build modulators and switches from nonlinear effects in guided wave interferometers and resonators operating at sub-picosecond time scales.\textsuperscript{13–15} The issue with interband pumping is that it necessitates pump photons at wavelengths much shorter than the telecommunication bands in the near infrared.
To avoid this problem, intraband (using longer wavelength photons) transitions in so-called "epsilon near zero" materials have been identified as extraordinary new building blocks for high performance optical devices with extremely high nonlinearity and ultrafast switching speed covering the near- and mid-infrared wavelength ranges.5–19,23–25 Again, integration of epsilon near zero materials with optical fibers remains challenging for now and it is desirable to find devices and configurations where hot carriers from intraband pumping could be used. It has been shown that hot carriers generated by SPPs at near-infrared wavelengths in "conventional" metals can be used for switching but with modulation efficiencies much lower than those achieved by interband transitions.16,19,20 Therefore, designing practical switches with gigahertz-level bandwidths and high modulation efficiencies at telecommunication wavelengths remains an important goal for photonic and materials research.

One of the ways in which light guided in the core of standard single mode fibers can be made to interact with materials deposited on the surface of the cladding is through tilted fiber Bragg grating (TFBG) written in the fiber core by photosensitive processes.26 The transmission spectrum of TFBGs contains hundreds of "comb"-like discrete resonances corresponding to coupling from a forward propagating core mode light to backward propagating cladding modes. Under certain conditions, when the cladding is covered by a thin metal coating, some of the cladding modes have effective indices that are closely matched to the effective index of an SPP mode propagating at the outer surface of the metal coating. When this occurs, and when the metal layer is thin enough for the evanescent field of the cladding modes to tunnel through the metal layer, some of the cladding mode power is coupled to the SPP, resulting in increased attenuation of the cladding mode and some broadening of the corresponding resonances.27–29 It is important to note that the TFBG resonances have a Q-factor greater than $10^4$ and their signal to noise ratio can be greater than 40 dB, with near zero insertion loss out of the resonances. Because of the large spectral slopes of the cladding mode resonances, small perturbations of the cladding mode properties (such as those due to permittivity changes occurring near the outer metal surface) can lead to large changes in transmission at certain wavelengths, by tens of decibel in physical vapor deposition system (Balzers BA 510) at room temperature (20 $°$C). Two deposition steps were carried out, with a rotation of the fiber holder by 180 $°$ between the two runs. The depositions were carried out at a relatively rapid deposition rate of 6 nm/min for 7.5 min/side, resulting in a mass-equivalent thickness of 45 nm around the full circumference of the fiber.

C. Dynamic TFBG-SPR simulations

In the simulations of TFBG-SPR spectra modulated by hot carrier decay, three steps are considered for the ultrafast switching process. First, the temporal evolution of hot electron populations and lattice temperature due to plasmonic excitation of the gold film were simulated by using a delayed two-temperature model within the short pumping time (supplementary material, note 2). Second, the resulting change in the dielectric function (complex permittivity) of the gold layer was calculated by using a Drude model with two critical points and damping terms modulated by hot electron and phonon temperatures (supplementary material, note 3). Finally, the time-dependent TFBG-SPR spectra associated with the variations of the dielectric function of the gold layer were simulated by first calculating the changes in the radially polarized cladding mode families (i.e., the EH and TM mode families, which are the only ones that can hybridize with SPPs of a metal layer on the cladding) with wavelength associated with another cladding mode resonance (but still within the spectral bandwidth of the SPP), the latter signal is modulated by the pump-induced optical property changes in the metal coating, on the same time scale as that of the pump. We achieved the modulation of a CW probe signal with $\sim$56 ps response time and switching efficiency as high as 30% $\pm$ 7%/GW/cm$^2$.22

II. METHODS

A. Fabrication of the TFBG

The TFBGs (20 mm in length) were inscribed in hydrogen-loaded CORNING SMF-28 fibers by using the phase-mask method.23 The fibers were hydrogen-loaded at a pressure of 15.2 MPa, a temperature of 20 $°$C, and a duration of 14 days, conditions sufficient to saturate the fiber core and increase its photosensitivity to ultraviolet light. After stripping a 5 cm section of fiber jacket, gratings were inscribed in the stripped section with a pulsed high-energy excimer laser (model PM-848 from LightMachinery, Inc.) and the phase mask technique. The laser was operating at 248 nm with a fluence per pulse at the fiber of ~40 mJ/cm$^2$ over an area of $2 \times 50 \mu m$ determined by selecting a portion of the laser beam and focusing it along the fiber. The tilt of the grating fringes was obtained by rotating the fiber(phase mask assembly (as well as the cylindrical focusing lens) around an axis perpendicular to the fiber axis and the plane of incidence.

B. Fabrication of the gold coating

A 45 nm thick uniform gold film (supplementary material, note 1) was deposited on the TFBG by thermal evaporation. To achieve a high-quality coating, the stripped fiber section is cleaned by immersing it in a piranha solution (an 8:1:1 mixture of de-ionized water, ammonium hydroxide, and hydrogen peroxide) for 30 min. Then, the TFBG was fixed on a movable holder in a thermal-evaporation physical vapor deposition system (Balzers BA 510) at room temperature and under vacuum ($10^{-7}$ Torr). Two deposition steps were carried out, with a rotation of the fiber holder by 180 $°$ between the two runs. The depositions were carried out at a relatively rapid deposition rate of 6 nm/min for 7.5 min/side, resulting in a mass-equivalent thickness of 45 nm around the full circumference of the fiber.
a cylindrical finite-difference vector mode solver at each wavelength (and taking into account material dispersion) and then using complex coupled-mode theory and a Runge-Kutta algorithm to calculate the transmission at each wavelength.

D. Ultrafast switching measurement

As shown in Fig. S4 (supplementary material, note 4), the pump-probe interrogation setup consists of a tunable picosecond laser (Genia Photonics programmable picosecond laser, pulse width: 25 ps; repetition rate: 1 MHz; adjustable average power: 0–100 mW) and a CW tunable laser (HP 81640A tunable laser, power: 2 mW) coupled into a common fiber with a 3 dB coupler. A manually adjustable bandpass filter (Yenista XTM-50) was used downstream of the TFBG to prevent the pumping pulse from reaching the detector. The pump modulated probe light was detected by a 40 GHz photodetector (New Focus, Inc) and measured with a digital oscilloscope (86100D Infiniium DCA-X wide-bandwidth oscilloscope). Pump and probe wavelengths were adjusted precisely relative to the TFBG resonances by directing the output of the TFBG to an optical spectrum analyzer (ANDO AQ 6317B) instead of the photodiode, in between switching experiments.

III. RESULTS

A. The hybrid SPP-cladding modes of a TFBG

The effective index of the SPP propagating at the interface of two media with relative permittivity \( \varepsilon_1 \) and \( \varepsilon_2 \) is given by

\[
N_{\text{eff}}^{\text{SPP}} = \sqrt{\frac{\varepsilon_1 \varepsilon_2}{\varepsilon_1 + \varepsilon_2}}.
\]

At near infrared wavelengths in the vicinity of 1550 nm, the relative permittivity for gold and water is \(-115.13 \pm 11.259i\) and \(1.7371 - 0.000 098 6i\), respectively, yielding \(N_{\text{eff}}^{\text{SPP}}\) values near 1.328. Then, in order to excite the SPP at the surface of a gold coating deposited on the cladding of an optical fiber immersed in water and remembering that the cladding supports guided mode with effective indices between the index of silica glass (near 1.444 at these wavelengths) and that of the external medium (1.315), it is necessary that the cladding supports guided mode with effective indices near that of pure SPP waves of a gold-water interface. For an S-polarized input, no SPP can be excited even in water because the cladding mode polarization is then TE-like. When a SPP is excited, the mode power fraction in water increases from 0.68% because the cladding mode polarization is then TE-like. When a SPP is excited, the mode power fraction in water increases from 0.68% to 25.7% and in the metal layer from 0.035% to 0.736% (supplementary material, note 5, Fig. S7). This has two consequences: shifts in the resonance wavelengths and resonance amplitude decreases of those cladding modes with effective indices in the vicinity of the required value at wavelengths near 1541 nm where cladding modes have effective indices near that of pure SPP waves of a gold film. As shown in Fig. S4, the pump probed excited SPP on the gold surface and hot carriers. Those phonon-assisted transitions are important contributors to losses arising from various damping mechanisms such as a Landau damping and electron-phonon scattering which add up to contributions from electron-electron scattering and losses arising from defects of the metal such as nanoscale roughness (Fig. 1).22

Figure 2 shows the transmission spectra of a 12° tilted Au-coated 2-cm long TFBG measured in air and immersed in deionized water. The main feature of these spectra is the presence of a very dense comb of cladding mode resonances that are individually very narrow (sub-nanometer). In both cases, in order to fulfill the necessary condition for the excitation of SPP waves of the gold film, the input light injected in the core is polarized in the plane of the tilt (P-polarized) which results in the excitation of cladding modes that are radially polarized at the outside surface of the fiber (i.e., TM-like). When the TFBG is in air, the index contrast is highest, and the cladding modes are well confined in the fiber by the 45 nm thick gold coating and the spectrum is not much different from that of the same TFBG in a bare fiber. However, when the TFBG is immersed in water, the lower index contrast allows some tunneling of energy to the external surface of the gold and to the excitation of a SPP at wavelengths near 1541 nm where cladding modes have effective indices near that of pure SPP waves of a gold-water interface. The measured resonance modifications are reported in Fig. 2(b) and fitted by a Lorentz line shape function of the mode effective indices (supplementary material, note 6) where

![FIG. 1. Schematic diagram of the plasmonic gold-coated TFBG. The pulsed pump (I_p) coupled out of the core by the tilted grating excited SPP on the gold surface and hot carriers (e^- and h^+) that in turn modulate the metal permittivity. A copropagating CW probe at a different wavelength gets modulated as the grating resonances shift in response to the permittivity change.](image_url)

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the real and imaginary parts are associated, respectively, with wavelength shifts (directly caused by a change in the real part of the mode effective index, through the grating phase matching condition) and with transmission changes (since the increase in the imaginary part, i.e., loss, of the mode effective indices shortens the effective coupling length and hence the reflectivity of the resonance). A Lorentzian fit of the shifts gives a SPP-hybridization width of 0.32 nm for the metal coated fiber in Fig. 2(c). As in other surface plasmon resonance phenomena, the hybridization width is strongly dependent on the metal thickness (supplementary material, note 7, Fig. S8). Here, the 44 nm thickness and very low surface roughness (near 1 nm) result in a very narrow lossy bandwidth where collective collision damping is the dominant source of loss. In these conditions, only 1 or 2 cladding mode resonances are strongly attenuated. While these particular resonances are rendered "useless" by the excessive loss and consequential weakening of the coupling, adjacent resonances retain enough coupling to the SPP while maintaining significant coupling with the core mode as well as a narrow linewidth required for efficient switching.

In order to demonstrate the validity of the Lorentz model for absorption, the complex effective indices of the hybridized cladding-guided-SPP modes of the Au-coated fiber were simulated using the FIMMwave mode solver (from Photon Design, Inc.). By use of the grating phase matching relation, the simulation results can be plotted as the mode loss (imaginary part of the effective mode index) against wavelength, as shown in Fig. 2(c). The imaginary part of the cladding mode effective index peaks at a value of 0.0007 close to the SPP resonance and fits the Lorentzian line shape of the measured resonance loss increases. At the peak, the loss corresponds to an attenuation coefficient of 57 cm$^{-1}$ for the cladding mode centered on the SPR wavelength, much too large for efficient coupling by the 2 cm-long grating. Pumping on the nearest neighbor resonance however still couples energy to the SPP but with a cladding mode that has an attenuation coefficient 10 times smaller (of the order of 6 cm$^{-1}$) which allows for strong coupling from the input core mode.

While this excitation condition is relatively narrowband, its consequences following intense in-band pumping of one resonance
affect all the other cladding mode resonances within the SPP bandwidth due to the global permittivity change induced by the hot carriers. Therefore, probe light at neighboring resonances will experience the consequences of pumping (i.e., modulation) at some wavelength separation from the pump wavelength and peak SPP coupling without the excessive damping losses experienced by in-band light.

B. Ultrafast dynamic of surface plasmon decay

In noble metals, it was demonstrated that internal thermalization of the electrons (i.e., establishment of an electron temperature following some excitation mechanism) takes place on the time scale of a few picoseconds, thereby allowing ultrafast switching mechanisms to take place. The change in temperature of the conduction band electrons modifies the Fermi-Dirac distribution function, which decreases the population below the Fermi level and increases it above the Fermi level. As a result, the response time and the non-linear susceptibility of hot electrons in metals in terms of plasmonics have been widely investigated. In particular, hot carriers generated by surface plasmon decay in thin metal films make it possible to achieve ultrafast switching based on the modulation of high Q-factor transmission resonances in TFBGs. Figure 3 shows a simulation of the effect of the dynamic hot carriers generated by picosecond pulsed excitation of SPP on the transmission spectrum of a TFBG.

A delayed two-temperature model for the hot electrons (supplementary material, note 2, Fig. S2) is used to calculate the non-thermal energy dissipation by electron-electron scattering and by reaching thermal equilibrium via electron-phonon scattering. This results in a time-dependent change in the damping term of the imaginary part of the complex permittivity of the Au film (supplementary material, note 3, Fig. S3). The calculated permittivity as a function of time is then input into a cylindrical finite-difference complex vector mode solver to calculate perturbed mode fields and complex effective indices which are then used to calculate transmission spectra by coupled mode theory. The end result is a sequence of simulated transmission spectra during and after the arrival of a 25 ps duration pump pulse from time t = −50 to +125 ps [Fig. 3(a)] and more closely during the pulse (t = 3 ps) in Fig. 3(b). In this simulation, the fact that the physical length of the 25 ps pulse (FWHM 5 mm) is similar to that of the TFBG was not taken into account (i.e., it was assumed that the electron temperature was the same, as a function of time, over the whole length of the grating). It is still clear from Figs. 3(a) and 3(b) that during high intensity irradiation by the pumping pulse many resonances shift slightly, and some weaken considerably as well, over a certain bandwidth around the SPP peak attenuation and mostly on the long wavelength side. The broadening of the SPP-induced
attenuation agrees with previous observations in free-space plasmonics.\textsuperscript{46,47} The postpump recovery is somewhat slower than the rise time because the 25 ps input pulse is much longer than the intrinsic hot carrier relaxation time (less than 1 ps)\textsuperscript{22} and because thermalization of excited electrons also occurs through other, slower processes and continues to disrupt the permittivity of Au. Another factor that will slow down recovery in actual experiments is the temperature dependence of the overall refractive index of the structure and the TFBG itself while heat generated by the excitation process must be dissipated. The main prediction of this simulation remains however that the arrival of the pump pulse and its coupling to the hybridized-cladding mode identified in Fig. 3(c) will cause a shift in the resonance position of another cladding mode [at the “probe” wavelength in Fig. 3(c)]. Therefore, a copropagating continuous wave core mode at the probe wavelength will be modulated synchronously by the pump pulse but with a slightly longer “tail” (leading to a modulated pulse length of 56 ps) due to the slow relaxation processes predicted in the two-temperature model. Figure 3(d) shows a typical experimental result for the modulated CW probe (described in detail in Sec. III C) which agrees well with the predicted pulse shape.

C. Ultrafast switching experimental results

Figure 4 shows the change in the transmitted power of a CW laser probe copropagating in the fiber core with the pump, as a function of pump power and probe wavelength. The pump wavelength was fixed at 1540.4 nm, on the long wavelength side of the first resonance located on the short wavelength side of the SPP maximum. This pump wavelength was determined experimentally to yield the strongest probe modulation. When the probe wavelength is located on the transmission minimum of the resonance, the arrival of the pump pulse blue-shifts the resonance and the transmitted power increases. Figure 4(a) shows the result for this situation, i.e., a pulsed increase in the probe power with rise times ranging from 27 to 39 ps and slightly longer recovery times. The maximum modulation depth increases with pump power and reaches 4.5% at an average pump power of 50 mW (corresponding to 0.147 GW/cm\textsuperscript{2} in the metal layer, given an excitation efficiency of 0.7% between the input power and the guided power in the metal, as described in supplementary material, note 8). The modulation depth was determined by comparing the power level changes to those measured by blocking the CW beam. For higher pump powers, the modulation depth of the
CW beam decreases, likely due to saturation of SPR together with competing slow processes caused by the relatively high (1 MHz) pulse repetition rate of our tunable 25 ps laser. Self-detuning of the pump from its resonance may also contribute to lower efficiencies at higher pump levels. The self-detuning can be observed in simulations [Fig. 3(c)] and experiments: the transmitted pumping pulses are broadened (even broken up into a train of smaller pulse) and show an ultrafast amplitude modulation (greater than 35%) (supplementary material, note 4, Figs. S5 and S6). It was verified that the decrease at higher pump powers is not due to damage (such as delamination of the gold film) because it was verified that high modulation efficiencies return after testing at higher pumping levels. Figure 4(b) shows corresponding simulations for transmission at the probe wavelength where the range of power density in the metal layer is from 0.088 to 0.206 GW/cm².

In Fig. 4(c), the switching response at the optimum pump power is measured for different wavelength positions of the probe relative to the resonance. The response time remains stable, and the highest efficiency occurs when the probe is right on the resonance transmission minimum, in good agreement with simulations [Fig. 4(d)], except for the case where the CW probe is tuned to the shorter wavelength edge of the probe-mode resonance, because the “reverse” pulse generated by the switching is below the background noise.

Figure 5 summarizes the switching results in terms of modulation efficiency and pulse width of the modulated probe, up to the maximum power level available from our system. The maximum efficiency occurs at the same power level as the largest temporal broadening, and the effects appear to decrease and saturate beyond that point. Finally, it was verified that no modulation is observed when pumping and probing at resonances further away from the SPP spectral peak, but also at any wavelength (even close to the SPP maximum), for S-polarized input light in agreement with the well-established result that surface plasmon resonances are never observed for gold thicknesses of a few tens of nanometers due to the fact that this polarization couples only to TE-like cladding modes.

IV. DISCUSSION

The observations reported in this paper constitute both experimental and simulation evidence that hot carriers can be generated by SPP waves excited on the surface of metal coated optical fibers by in-core TFBGs and that these hot carriers modulate the complex permittivity of the metal layer on a picosecond time scale. The spectral width over which the SPP excitation is possible for an optimal gold thickness and low roughness is still quite larger than the wavelength spacing between the grating resonances of the modes of the fiber cladding which means that pumping and probing of the complex permittivity can be carried out simultaneously with different wavelengths copropagating simultaneously in the single mode core. While the results for high speed modulation of a CW probe by hot carriers from pump pulses show a reasonable overall agreement with simulations based on a very simple model of the light matter interaction, several factors may impact the maximum modulating efficiency observed here. First, the impact of the hot carriers on the complex permittivity shifts the transmission resonance not only at the probe wavelength but also at the pump wavelength which causes a kind of self-modulation of the pump, i.e., the pump pulse “detunes” itself from resonance during the pulse. The main effect of this is an unresolved ultrafast relative intensity noise as large as 18% superimposed on the measured transmission of the pump pulses which, most significantly, is maximum for the same input pulse intensity where the maximum probe modulation is observed (0.147 GW/cm²). Another issue is that the spatial extent of the 25 ps pump pulse in the fiber is around 5 mm, i.e., ¼ of the grating length. Therefore, contrary to what is assumed in the simulations, an SPP wave is only excited in a small part of the grating at any given instant. While the SPP waves propagate some distance backward from their excitation point and the cladding mode responsible for it has a power distribution that falls exponentially with distance from the beginning to the end of the grating (with an effective length of 6 mm, for the pumping resonance, based on coupled-mode theory for contradirectional coupling), it is fair to say that the complex permittivity change associated with the hot carriers is likely to be quite nonuniform along the grating, resulting in a fast chirp of the grating resonances. Compounding this issue is the fact that the spectral width of the 25 ps is near 0.13 nm, i.e., roughly the same width as the transmission resonances so that the total power transmission factor results from the convolution of the pulse and resonance spectra. Finally, there may be saturation of the surface plasmon in our layers beyond the maximum switching power, and the efficiency of probe modulation goes down. A significantly more complex theoretical modeling will be required to fully explain these results and lead to improved efficiency by optimizing all the adjustable parameters (such as grating strength and length, and coating material).

V. CONCLUSION

Despite these mitigating factors, an “all single-mode fiber” configuration was demonstrated where core-pumping a gold-coated TFBG in water with 25 ps pulses of near infrared light at 1 MHz and 50 mW of average input power resulted in a synchronous modulation of a CW signal propagating in the core of the same fiber.

FIG. 5. Summary of pump-induced modulation of a CW probe as a function of the power density in the metal layer. The input (pump) pulse width is 25 ps.
by 4.5%, with a modulating efficiency of 30% ± 7% per GW/cm² and a pulse duration widening of ~30 ps. This achievement stems from many factors, including the low insertion of loss of the TFBG inscribed in standard telecom fiber, the dense set of narrowband cladding mode resonances with a high Q factor, and the strong light-matter interaction efficiency of the plasmonic hybridized cladding modes. Even though the reported modulation efficiency obtained in these devices is currently too low for conventional switching applications (in data transmission for instance), the fact that watt level input powers can be guided in this kind of fiber without damage leads to further potential applications in fiber-based nonlinear light-matter interactions such as nonlinear sensing, plasmonic-assisted frequency conversion, and terahertz antenna. It is also believed that further experiments with parameter variations (of grating properties, material choices for the coating, and pump pulse widths, for instance) will lead to an improved understanding of the effects observed and optimization of their performance.

SUPPLEMENTARY MATERIAL

See supplementary material, Video 1 and Video 2.

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