Strong optical modulation of surface plasmon polaritons in metal/semiconductor nanostructures

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Abstract: We demonstrate strong modulation of the transmission around the surface plasmon polariton (SPP) resonance in metal/semiconductor hybrid nanostructures based on Ag film on top of InGaAs. The change in the real and imaginary parts of the refractive index due to photoexcited carriers in InGaAs generates a shift in the SPP resonance and enhanced transmission near the SPP resonance. Temporal evolution of the complex refractive index was traced by comparing the transient transmission with finite-difference time-domain (FDTD) simulations.

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References and links


1. Introduction

Since the pioneering work on extraordinary light transmission through a periodic hole array perforated in a thin metal film by Ebbesen et al. [1], surface plasmon polaritons (SPPs) have attracted much interest in fundamental understanding of optical device applications [2–5]. The research area of active plasmonics, in which the main concern is to control propagation of the SPP electrically or optically, is also rapidly progressing nowadays [6–13]. Modulation of SPP propagation along the metal/dielectric interface can be achieved by manipulating the material properties of either the metallic or the dielectric layer.

Although the conventional method used to control carrier density in the metallic layer has enabled high-speed functionality in the terahertz bandwidth [14], its efficiency is relatively low because of the large amount of background carriers in metal. Consequently, to improve efficiency, optical switching of SPPs was pursued by incorporating phase transition materials such as Ga [6] and VO₂ [15, 16]. Bulk semiconductors have not been a popular choice for the dielectric material constituting the metal/dielectric interface because of their large absorption coefficient, which reduces the propagation length of the SPP and deteriorates its resonance behavior. However, when the fact that the controllability of semiconductors is well established and their efficiency can be quite good is considered, the metal/semiconductor hybrid system can be seen as a good candidate for switching SPPs. In addition, the switching efficiency can be further enhanced by designing the SPP resonance to occur near the semiconductor band gap [17, 18].

In this paper, we report on active control of SPPs at the Ag/InGaAs interface with a one-dimensional grating structure, which was studied by performing femtosecond transient transmission measurements. The shift in the SPP resonance and the pronounced resonance behavior due to carrier excitation in the InGaAs layer are attributable to the decrease in both the refractive index and the absorption coefficient of the InGaAs. By comparing experimental results with finite-difference time-domain (FDTD) simulations the dynamic changes in the real and imaginary parts of the complex refractive index were traced simultaneously.

2. Experimental results and discussion

A 400-nm-thick In₀.₅₃Ga₀.₄₇As layer, lattice-matched to an InP substrate, was grown via metalorganic chemical vapor deposition (MOCVD). Then, a 120-nm-thick layer of Ag was deposited on the InGaAs and one-dimensional gratings fabricated using the e-beam lithography technique within the metallic region. The grating period in this study was 420 nm and the filling factor, defined as the ratio of the region covered by the metal in the period, was 70%. From the photoluminescence spectrum at room temperature, we discovered that the
band gap of the InGaAs layer was located around a wavelength of 1.65 μm. The resonance wavelength of the SPP follows the relation,

\[ \lambda_{SPP} = \frac{2\pi}{k} \sqrt{\varepsilon_m \varepsilon_d} \]  

where \( k \) is the wave vector and \( \varepsilon_m \) (\( \varepsilon_d \)) is the permittivity of the metallic (dielectric) layer. For normal incidence light, the wavevector is determined only by the grating period (\( k = \frac{2\pi}{\Lambda} \)), and the SPP resonance wavelength is located marginally above the InGaAs bandgap for a Ag slit array with \( \Lambda = 420 \) nm.

Modulation of the transmission spectrum by photoexcited carriers was studied by performing the pump-probe experiment, which is schematically depicted in Fig. 1(a). Pump pulses at 800 nm from a Ti:sapphire laser with a pulse duration of 150 fs and a repetition rate of 91 MHz generated carriers in the InGaAs layer. An optical parametric oscillator synchronized with the Ti:sapphire laser provided probe pulses with a duration of approximately 300 fs, and which could be tuned over the range 1200 to 1600 nm. While changing the time delay between the pump and probe pulse, the transient transmission spectrum was measured with a tunable probe wavelength. To realize the time delay, the optical path length of the pump beam was controlled by passing it to a retro-reflecting mirror mounted on a linear stage. The zero time delay was confirmed by measuring the sum-frequency generation at a BBO crystal occurring between the pump and probe pulse. In the pump-probe experiments the pump pulse passes through a mechanical chopper and the...
modulated probe pulse intensity is measured by using a lock-in amplifier equipped with a Ge photodiode.

The transmission spectrum in Fig. 1(b), which was measured at normal incidence with the polarization perpendicular to the slit, reveals transmission dips at wavelengths of 1377 nm and 1510 nm. From our previous studies on the transmission spectrum depending on InGaAs thickness, we know that the 400-nm-thick InGaAs core, surrounded by the lower refractive index of the InP substrate at one side and the metal grating at the other side, supports the fundamental transverse magnetic (TM) waveguide mode together with the SPP mode (SP) [19]. The two transmission dips correspond to the resonant energy transfer from the incident light to the waveguide (WG) and SP.

The FDTD simulations of the field profiles at three wavelengths are displayed in Figs. 1(c)-(e). In the simulations, we used the reference values by Adachi as the dielectric constants of the InGaAs layer [20]. In waveguide mode ($\lambda_{WG} = 1377$ nm), a large portion of the field propagated along the InGaAs/InP interface. In SP mode ($\lambda_{SP} = 1510$ nm), the field was strongly confined at the metal/InGaAs interface and decayed exponentially in the InGaAs region. However, the field profile at the in-between wavelength of 1443 nm in Fig. 1(d) is distinctive from the resonant cases, showing that the field is not evanescent in the InP region but much of the energy is transferred through the InP substrate. Because both the waveguide mode and the SPP contribute at this wavelength, which is the lower frequency side of the waveguide mode but blue-shifted from the SP, the phase difference expected between the two guided modes will deteriorate the guiding effect and can lead to energy dissipation into the InP substrate. Figure 1(d) further shows that the field just below the metallic region is diminished in stark contrast to the other cases. We note that a similar field absence beneath a metallic wire was observed for the condition when the localized SP of the nanowire destructively interfered with the waveguide mode in a metallic wire array hybridized with an ITO waveguide [21]. But, the propagating SP mode in this study is always located at a longer wavelength than the waveguide mode, which is different from ref [21], where the anticrossing behavior of the localized SP with the waveguide mode was discussed.

Fig. 2. (a) Transmission spectra obtained at several time delays after pump pulse excitation with the shaded curve being the transmission spectrum at a negative time delay ($\tau = -30$ ps). The arrows at each curve points to the transmission peak. (b) Wavelength and (c) transmission intensity at the transmission peak as a function of time delay.
The change in the permittivity of the InGaAs layer due to the carrier generation induces modulation of the transmission spectrum and SPP resonance. The transmission spectra for several time delays (τ = 1–300 ps) after the pump pulse are displayed in Fig. 2(a), where the shaded gray curve is the transmission spectrum before the pump pulse (τ = −30 ps). For carrier excitation, a pump fluence of approximately 1 nJ was used with a spot size of 20 μm at the sample surface. The arrow at each curve indicates the position of the transmission peak.

At initial time delays τ = 1 ps and 15 ps, it can clearly be observed that the structure of the transient transmission is shifted to shorter wavelengths compared to that of the negative time delay. The blue-shift in the transmission spectrum after photoexcitation is also accompanied by pronounced resonant behaviors. For instance, at the 15-ps time delay the transmission dip in SP mode is stronger and the transmission peak is increased to three times the value it was before photoexcitation. For the transmission peaks, the peak wavelength and intensity is plotted as a function of the time delay in Figs. 2(b) and 2(c), respectively. While the wavelength shift is almost instantaneous after pump excitation reaching the maximum shift of Δλ ≈ 33 nm at τ = 2 ps, the largest transmission occurs only at τ = 20 ps with ~300% enhancement. The transmission enhancement is dominant between the waveguide and SP, which may be utilized for wavelength-selective optical switches.

Fig. 3. FDTD simulation of the transmission spectrum at normal incidence obtained (a) while varying the real part and (b) while varying the imaginary part of the refractive index. The wavelength at the peak transmission shifts with the real part, and the transmission becomes pronounced when the imaginary value is reduced.

To determine the underlying mechanism that modulates the transmission spectrum, we carried out FDTD simulations in which we assumed constant dielectric constants for simplicity. Only the variation in the dielectric constant of the InGaAs was considered because most carriers are generated in there. In the complex value of the refractive index \( n = n + iκ \), the real part \( n \) is connected with the phase velocity \( v_p = c / n \), while the imaginary part \( κ \) determines the absorption coefficient \( α \) according to the equation \( α = 4πκ / λ \). First, the transmission was simulated with the real part \( n \) adjusted while the imaginary part was neglected. The results in Fig. 3(a) indicate that the transmission peak shifts proportionally with the value of \( n \). This is as expected because, according to Eq. (1), the SPP wavelength is...
proportional to \( n \) in the approximation where the absolute dielectric constant of metal is much larger than that of the dielectric material. On the other hand, the simulations in Fig. 3(b), conducted while adjusting the imaginary part \( \kappa \), indicate that the transmission peak is stronger for smaller \( \kappa \) values. This is also plausible because the SPP cannot propagate over any great distance and the resonant features can be smeared out when the absorption coefficient is large. Thus, the wavelength shift and the pronounced transmission peak in the experiment can be explained if both the real part \( n \) and the imaginary part \( \kappa \) in the complex refractive index of InGaAs decrease because of the photoexcited carriers.

![Fig. 4. (a) Experimentally obtained transmission spectra at time delays \( \tau = 7 \) ps and \( \tau = -30 \) ps. (b) Reconstructed transmission spectra using FDTD simulation to match the transmission peak of the experiments. The complex refractive indices used for the simulation are denoted for each curve. (c) Real and imaginary refractive indices as functions of time delay—extracted by ascertaining the parameters that best simulate the transient transmission spectrum at each time delay.](image)

After performing the FDTD simulations in which the values of \( n \) and \( \kappa \) were independently controlled, the transmission spectrum at each delay was reconstructed to match the experimental results, especially in terms of position and strength of the transmission peak. Figure 4(a) demonstrates that \( \bar{n} = 3.622 + 0.146i \) and \( \bar{n} = 3.513 + 0.068i \) for the InGaAs well simulates the transmission spectrum at \( \tau = -30 \) ps and \( \tau = +7 \) ps, respectively. We note that the value of the refractive index used to simulate the transmission at a negative time delay is very close to that at 1400 nm obtained by Dinges et al. from the ellipsometry measurement [22]. From these procedures, the complex refractive index was extracted at each delay, which is summarized in Fig. 4(b). The fast recovery of the refractive index with the imaginary part having a decay constant of about 110 ps, might originate from the accelerated carrier recombination due to the Purcell effect in our sample with metal gratings. For the real part, the maximum change of \( \Delta n = -0.11 \) corresponding to a 3% decrease, occurs at \( \tau = 20 \) ps from the value \( n = 3.62 \) at a negative delay. On the other hand, the smallest value of \( \kappa \) for the imaginary part was obtained at \( \tau = 20 \) ps. Figure 4(b) also reveals that recovery of the nonequilibrium refractive index is slower for the imaginary part. At time delays greater than 100 ps, the refractive index \( n \) becomes even larger than the pre-excitation value. It is interesting that the change in the refractive index \( n \) is not monotonic but the sign reversal is possible with time delay.
The occupation of the conduction band and the vacancy of the valence band modify the absorption coefficient. As the carrier density decreases with the recombination process, the distribution of excited carriers and the energy of large absorption change moves toward the band edge. According to the Kramers-Kronig relation, the change in the absorption coefficient induces modulation in the real part refractive index, such that

$$\Delta n(\omega) = \frac{c}{\pi} \text{Pf} \int \frac{\Delta \alpha(\omega')}{\omega'^2 - \omega^2} d\omega'.$$

Equation (2) suggests that $\Delta n$ is negative (positive) when it is caused by negative $\Delta \alpha$ at higher (lower) frequencies. Thus, with the carrier recombination, as the energy of the absorption change sweeps to a lower energy across the SPP resonance, the value of $\Delta n$ will change from negative to positive values. In this case, the time delay at the sign reversal will correspond to the situation in which the dominant absorption change occurs near the SPP resonance. The faster recovery of the real part witnessed in Fig. 4(c) can also be explained by the Kramers-Kronig relation, in that the transient recovery of the real part is reached at the point of sign reversal.

Figure 5 shows how the complex refractive index evolves with time delay after photoexcitation. The complex refractive index at each time delay is marked as a point in the X-Y plane, where the X- and Y-axis represents the real and imaginary part, respectively. The refractive indices plotted in Fig. 5 clearly demonstrate that the evolution of $\Delta n$ does not follow $\Delta \alpha$ linearly, although the two values behave dependently. We note that the analysis based on the SPP resonance has enabled simultaneous determination of the real and imaginary parts of the complex refractive index with time delay, which is not possible from pump-probe measurements on bare semiconductor layers.

Fig. 5. The complex refractive index at each time delay after the pump pulse excitation is pointed in the X-Y plane with the horizontal (vertical) axis representing the real (imaginary) part of the refractive index.
3. Conclusion

Efficient optical modulation up to 300% of transmission was demonstrated near the SPP resonance on metal/semiconductor nanostructures of a Ag grating fabricated on 400-nm-thick InGaAs. The transient transmission spectrum also enabled time-resolved determination of the complex refractive index. Whereas the reduced absorption coefficient ($\Delta\alpha$) recovered monotonically with time delay, a reversed change of the real part ($\Delta n$) was observed at time delays greater than 100 ps, which can be explained by the energy shift of the $\Delta\alpha$ across the SPP resonance. We believe that efficient modulation of the SPP mode can be developed for wavelength-sensitive optical switches.

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