

Surface plasmon-polariton amplifiers

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ABSTRACT

Propagation of surface plasmons at metal surfaces is receiving much interest nowadays because of its broad range of potential applications, like subwavelength photonics or biosensing. Although plasmonic devices achieve unique properties, surface plasmons suffer from high attenuation because of the absorption losses in the metal. This limitation can be overcome by providing the material adjacent to the metal with optical gain. Under these conditions, absorption losses are compensated and the propagation length of the plasmon is significantly increased. In this work, a review of plasmonic amplifiers is presented. To this end, the state of the art of such devices and the propagation characteristics of surface plasmons under amplification are described. Finally, a novel material based on the incorporation of colloidal quantum dots in a polymer matrix is proposed as a gain medium. This kind of nanocomposite (polymer+quantum dots) is important because it combines the novel properties of colloidal quantum dots (temperature independent emission and color tuning with the base material) with the technological feasibility of polymers. Wavelength tunability of the device (from 400 nm to more than 2 μm) is achieved by changing the material of the dots and their size. Furthermore, electrical injection is possible in the future.

Keywords: plasmonic amplifier, colloidal quantum dots, nanocomposite, PMMA

1. INTRODUCTION

Surface plasmons (SP) are coherent oscillations of free electrons present at the boundary between a metal and a dielectric. Under appropriate conditions when SPs are exposed to a flow of visible or infrared light, they can be coupled to the incident photons leading to a hybrid electromagnetic-wave and charge-surface state known as surface plasmon polariton (SPP) [1]. In these conditions SPP can propagate along the interface showing unique properties as subwavelength confinement, strong near electromagnetic field enhancement or high sensitivity to the environment [2]. Then, SPP have led the development of a wide range of applications [3] like subwavelength photonics [4], biosensing [5] or metamaterials [6]. However, the ohmic losses present in the metal causes a strong attenuation of the electromagnetic wave limiting the propagation length of SPP and in consequence the potential applications. Indeed, good mode confinement, necessary to exploit the possibility to guide light beyond the diffraction limit, is achieved using thick metal layers. But propagation losses of SPP increase with the thickness of the metal as well, so there is a trade-off between good mode confinement and low propagation losses [2-3]. Nevertheless this problem can be overcome by means of providing gain in the dielectric adjacent to the metal. In this way, absorption losses can be compensated and the SPP propagation length significantly increased.

The idea of obtaining a lossless propagation SPP by a gain assisted dielectric were studied theoretically in planar geometries in [7], being the use of III-V multiple quantum wells proposed as a gain medium adjacent to silver layers. Then, similar structures using MQW were simulated in [8-9]. In [10] organic dyes are suggested as a medium with strong gain able to increase propagation length in silver films. In [11] stimulated emission at 633 nm of surface plasmons in silver films is experimentally demonstrated optically pumping a dye solution and using a prism-coupling method. In recent works, the dielectric gain material consisted of organic dyes embedded into a polymer matrix. Such a multicomponent material (polymer+active medium) is called nanocomposite and has the advantages of joining the active properties of the dyes with the technological feasibility of polymers (coating, lithography...). Therefore, in [12] a silver planar film surrounded by CYTOP and Rhodamine has been studied, demonstrating theoretically that for high enough powers and dye concentration amplification in the visible range is possible. In [13] the same dye dispersed in PMMA was investigated as a medium able to provide gain equal to 420 cm^{-1} . In these conditions the nanocomposite can compensate the losses of a silver film at 594 nm investigated using the attenuated total reflection setup. Then, in [14] a similar system was used the same authors to demonstrate stimulated emission of surface plasmons with the aid of unintentional scatters in the polymer film. Recently, a fluorescent polymer made by the dispersion of PFS in MDMO-PPV was chosen in

[15] to obtain a net optical gain of 8 cm^{-1} at 600 nm in the SPP. Finally, a mode power gain of 8.55 dB/mm at 882 nm was obtained [16] in a $1 \text{ }\mu\text{m}$ gold stripe using IR140 dye molecules adjacent to the metal. Not only dyes, but also other kind of active centres can be selected to provide gain in a SPP. For example in [17] an erbium doped glass gave stimulated emission in a gold stripe at 1532 nm. In the same way in [18], all plasmonic modulation of SPP signal is obtained as a consequence of the suppressed absorption of the SPP signal with Er ions. Other sorts of nanoparticles that can be a good choice to provide gain in a SPP are colloidal quantum dots (QD). These kinds of nanostructures are semiconductor nanocrystal synthesised by colloidal chemistry, and they join the three dimensional confinement of QDs with the feasibility of the chemical methods, able to provide radius of nanoparticles in the range from 1 to 10 nm [19]. As a consequence the emission is temperature independent (due to the small size), and the emission wavelength can be easily tuned by controlling the size or the base material without modifying substantially the surface chemistry. In this way optical amplification has already been demonstrated in dielectric waveguides dispersing the QDs in solgel matrixes [19]. Moreover, in [20] CdSe and CdTe QDs have been mixed in PMMA, and the optimum conditions for waveguiding were obtained. Indeed, amplification of SPP using colloidal QDs-PMMA nanocomposites has already been studied [21,22]. In [21] PbS QDs are proposed as a method to provide gain in a dielectric load surface plasmon polariton waveguide. This structure consists of a dielectric strip deposited on a metal film, being it possible to improve waveguiding characteristics choosing adequately its parameters [23]. Then, in [21] a 27% increase of propagation length at 1500 nm was obtained depositing a PbS-PMMA stripe on a gold layer evaporated on a glass substrate. Within a similar structure working at 876 nm [22] a 32 % compensation of SPP was demonstrated.

In this work preliminary results of the combination of dielectric and plasmonic waveguides integrated on a silicon substrate are presented as a hybrid photonic/plasmonic CMOS compatible device. If the dielectric medium can provide optical gain it is expected that SPP propagation lengths can increase significantly or even net amplification spectral regions can be formed. For this purpose an active medium consisted of QD-polymer nanocomposites is proposed with the intention of suggesting a technologically feasible material able to tune the amplified wavelength just by changing the nanostructure. To make a suitable design of the amplifier the propagation of SPP under amplification is studied, and an analysis of CdSe-PMMA nanocomposites as a gain medium is presented.

2. PROPAGATION OF SURFACE PLASMON POLARITONS UNDER AMPLIFICATION

The simplest method to simulate and predict the propagation characteristics of a signal in plasmonic waveguides is by solving for the propagation constant of supported modes. Maxwell's equations and boundary conditions lead to the dispersion relation in a given structure. In turn, the dispersion relation can be solved analytically or numerically to yield the dispersion curves (and, thus, the propagation constants) of the supported modes [1]. Loss can be modelled by considering complex permittivities for the lossy layers. For gain materials, the imaginary part of the refractive index has an opposite sign in comparison to the sign in lossy materials [7].

For example, let us consider a symmetric PMMA-Au-PMMA plasmonic waveguide of the insulator-metal-insulator (IMI) type. We utilize the Drude model for the metal layer, i.e. its permittivity is given by $\epsilon_1 = \epsilon_h - \omega_p^2 / (\omega^2 + i\omega\gamma)$, where $\epsilon_h = 9.84$ is the high frequency permittivity, ω_p is the plasma frequency (with $\lambda_p = 2\pi c / \omega_p = 137.78 \text{ nm}$), and $\gamma = 1.018 \times 10^{14} \text{ rad/s}$ is the collision frequency. We assume a complex PMMA refractive index of $n_2 = 1.522 - i0.000247$. The imaginary part models gain and its value corresponds to a PMMA-layer amplification of 20 cm^{-1} at $1.55 \text{ }\mu\text{m}$. The dispersion relation for the long-range mode is $\tanh(k_1 a) = -k_2 \epsilon_1 / (k_1 \epsilon_2)$, where ϵ_i are the material permittivities and $k_i = (\beta^2 - \epsilon_i \omega^2 / c^2)^{1/2}$, where β is the propagation constant. The dispersion relation can be solved by complex root-finding algorithms such as Müller's method [24].

Figure 1a presents the dispersion curve of the long-range mode for a metal thickness of $2a = 30 \text{ nm}$, while figure 1b shows the loss coefficient. The curve is approximately linear for low frequencies, similarly to conventional dielectric waveguides, and dispersion is normal, i.e. $d^2\beta/d\omega^2 > 0$. As frequency increases, group velocity, $d\omega/d\beta$, decreases, and the waveguide can be a candidate for slow light applications [25]. Then, dispersion turns anomalous at 469 nm, leading to an infinite group velocity at 465 nm. It is well-known that group velocity does not account for signal velocity in this regime [26,27]. For even higher frequencies, group velocity turns negative and the pulsed signal enters the "fast-light" regime, where the peak of the pulse can be observed exiting the waveguide before entering it [28].

Regarding losses, the propagation length (the distance where the signal peak power falls to 1/e of initial peak power) of the SPP is limited to $1/(2\kappa k_0)$, where k_0 is the wavenumber in free space and $n_{\text{eff}} = n + ik$ is the effective index for the mode. As shown in figure 1c, the loss coefficient, κ , is positive for $\lambda < 1072 \text{ nm}$, indicating that metal losses dominate PMMA gain for such frequencies. The loss coefficient vanishes at 1072 nm, and it is negative for higher frequencies, indicating that the waveguide presents net gain and is effectively an amplifier. The propagation lengths of the long-range mode improve with decreasing metal film thickness.

In comparison to the case without amplification, the major difference is that in the latter case the loss coefficient never vanishes or becomes negative; it converges asymptotically to zero for very low frequencies.

There is no spectral region with amplification and the resulting propagation lengths are always shorter. For example, at 1550 nm the propagation length in the context of the lossy case is <1 mm, while there is a net gain of 9.7 cm^{-1} when amplification is present. At 1000 nm, the propagation length is $280 \text{ }\mu\text{m}$ without gain and 2.5 mm with gain present; at 700 nm the respective propagation lengths are $62.2 \text{ }\mu\text{m}$ and $88.5 \text{ }\mu\text{m}$.

We note that the results for the case with losses and amplification are fundamentally different to the results of the ideal lossless case (i.e., $\gamma=0$ for the metal, no imaginary part for the PMMA index) [29,25]. In the lossless case, the dispersion curve of the fundamental long-range mode consists of 2 branches, one for positive and one for negative propagation constants. There is a highest allowed frequency and dispersion is always normal.

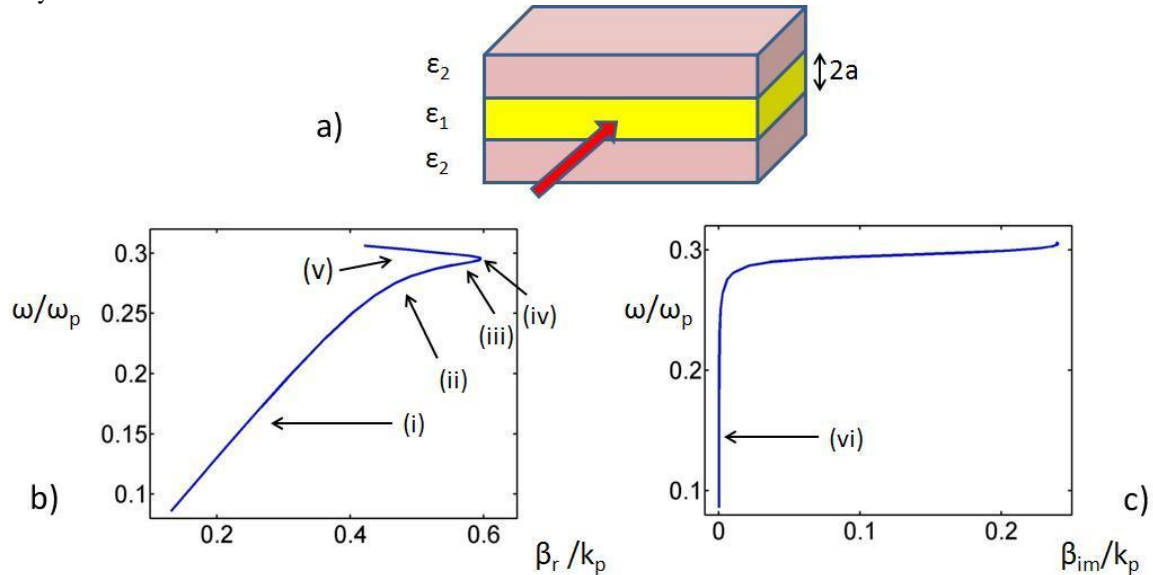


Figure 1 a) IMI waveguide. b) Real part of the propagation constant. (i) normal dispersion region (ii) slow light (iii) anomalous dispersion (iv) infinite group velocity (v) fast light. c) Imaginary part of the propagation constant. (vi) the frequency where the curve crosses the $\beta_{im}=0$ axis. PMMA gain and metal loss cancel out at that point. For lower frequencies, the waveguide presents net gain. Normalization constants are the plasma frequency and $kp=\omega_p/c$.

3. COLLOIDAL QUANTUM DOTS/PMMA WAVEGUIDES

Colloidal QDs are suitable nanostructures able to be integrated in photonic devices and provide gain in planar waveguides [19]. They present the properties of temperature independent emission able to be tuned by the base material or the size, and they are easily embedded in polymer matrices using a common solvent [20]. In this way, CdSe-PMMA nanocomposite has already been chosen as an active material of planar waveguides depositing the film on a SiO_2/Si substrate (see figure 2a) [20]. When appropriate concentrations of QDs into the polymer are chosen (filling factors between 10^{-3} and 10^{-4}) PL is coupled to the waveguide modes when the sample is excited by a pumping laser as it is shown in figure 2b. A GaN laser is focused on the surface of the sample with the aid of a cylindrical lens. Since CdSe quantum dots have strong absorption at this wavelength (see figure 2c), they are excited and its photoluminescence (PL) shown in figure 2c is waveguided by the structure. As it is depicted in figure 2b such PL can be collected by means of a microscope objective and can be focused on a spectrograph to analyze its spectra (figure 2d) or to a CCD camera to see its modal distribution (figure 2e). It is interesting to note that the different spectrum between the colloidal CdSe and the waveguided PL is due to the reabsorption in the structure. Finally it is worth mentioning that the conclusions obtained for CdSe QDs can be extrapolated to other sort of nanostructures just by changing the base material [20].

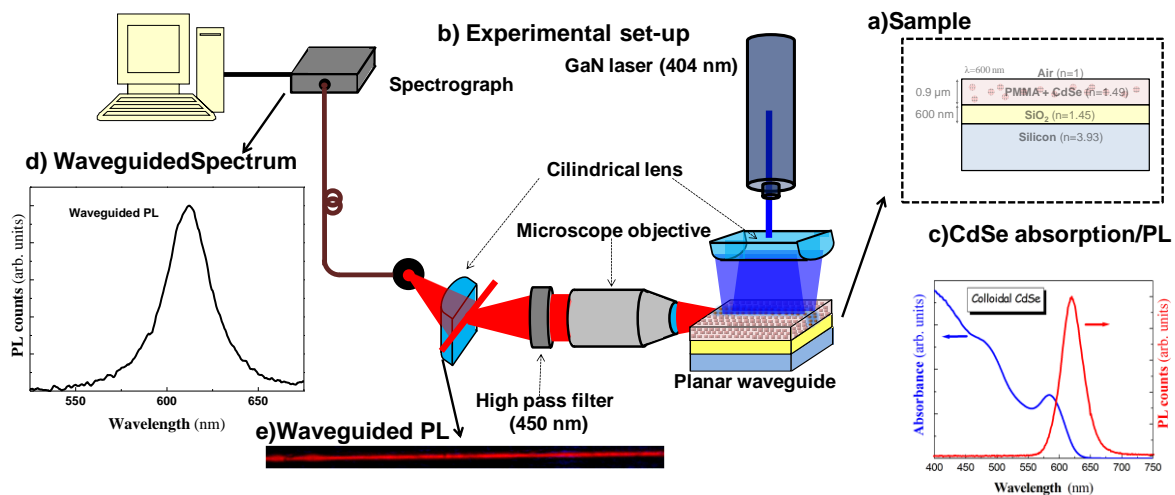


Figure 2 a) Experimental set-up for dielectric waveguides characterization. b) Structure of the sample. c) Photograph of the waveguided PL d) Waveguided (continuous line) and colloidal (dash line) CdSe spectrum.

4. PROPOSED AMPLIFIER IN NAVOLCHI

In this manuscript QD-PMMA nanocomposites are proposed as a dielectric medium adjacent to gold layers able to amplify the SPP propagating in the structure. The configuration shown in figure 2 is suggested as a new SPP amplifier design. It consists of a gold stripe surrounded by an active dielectric material (QD-PMMA nanocomposite) deposited on a SiO₂/Si substrate [16]. The samples can be analyzed by pumping the QDs with the aid of a cylindrical lens (as it was shown in figure 1b) and studding the output light of a probe beam coupled at the input edge of the structure. Since the substrate (SiO₂/Si) has a lower refractive index than PMMA the regions of the sample where there is not gold act as dielectric waveguide, making the coupling of the probe and pump beams.

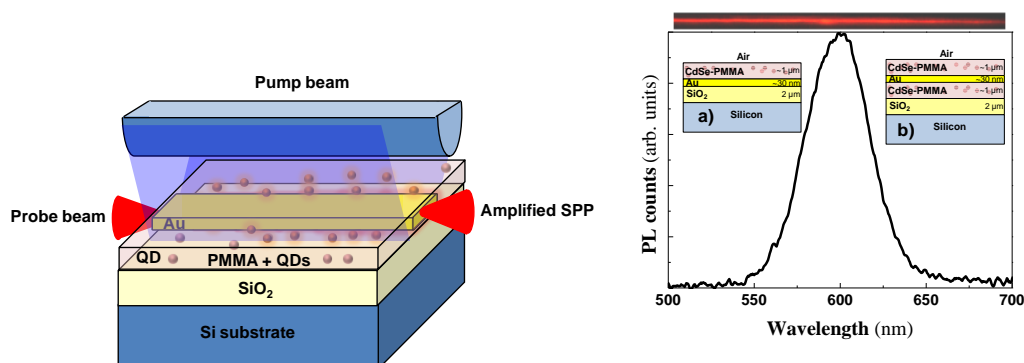


Figure 3. Left. SPP amplifier design. Right. CdSe PL coupled to a TM mode of the structure shown in the inset.

The insets of figure 3 right show a first structure analyzed. The first one (a) consists of a gold layer deposited on SiO₂/Si substrate and covered by CdSe-PMMA nanocomposite. In the second one (b) the gold layer is embedded into the nanocomposite. As a result, when the samples are pumped by a GaN the PL of the QDs is coupled to modes of the waveguide, making it possible to excite the plasmonic ones (TM). Figure 3 right shows the PL of the TM modes, and the top of the figure a photograph with a CCD camera. It is interesting to note that the a higher amount of signal can propagate in the symmetric structure (b), using the same excitation conditions; this is probably due to the fact that an index matching allows lower propagation losses [15]. Using the experimental set up proposed in [15] intrinsic gains between 0.5 and 20 cm⁻¹ have been measured depending on the concentration of quantum dots in the polymer and the pumping power. Although these figures look promising they are all cases in the order of the losses, and thus it is questionable whether the system can present net gain. Nevertheless, such CdSe PL coupling into plasmonic modes can be extrapolated to the design proposed in figure 3 a; and taking into account that it has already been demonstrated that use of metal stripes instead of layers increases the mode confinement and allows lower propagation losses [16], it is expected that such structure could be able to amplify the SPP modes. In addition, since propagation losses decreases with operation wavelength it is expected that the same structure can provide plasmonic amplification using QDs with PL at longer wavelengths (PbS and PbSe for emission in the infrared). Indeed, in section 2 it is demonstrated that a material gain of 20 cm⁻¹

¹ is enough for obtaining plasmonic amplification for wavelengths longer than 1072 nm. , new steps implementing such devices and using PbS and PbSe QDs will be carried out.

5. CONCLUSIONS

In this work an analysis of plasmonic amplifiers is presented, proposing the use of colloidal QD-PMMA nanocomposites as a suitable material able to provide gain to SPP modes propagating in a metal stripe or layer. To show the potentiality of this material, dielectric and plasmonic waveguides based on CdSe-PMMA nanocomposites are demonstrated by depositing the layers on a SiO₂/Si substrate and gold respectively. In both cases, when the structures are pumped a great amount of PL is coupled to the dielectric and SPP modes. Then, taking into account that this nanocomposite can show gains up to 20 cm⁻¹, it is expected that such a medium is able to provide gain in new plasmonic structures.

ACKNOWLEDGEMENTS

This work was supported through the Spanish MCINN and EU-NAVOLCHI Grants Nos. TEC2011-29120-C05-01 and 288869 respectively.

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