Optical gain, spontaneous and stimulated emission of surface plasmon polaritons in confined plasmonic waveguide

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Abstract: We develop a theoretical model to compute the local density of states in a confined plasmonic waveguide. Based on this model, we derive a simple formula with a clear physical interpretation for the lifetime modification of emitters embedded in the waveguide. The gain distribution within the active medium is then computed following the formalism developed in a recent work [Phys. Rev. B **78**, 161401 (2008)], by taking rigorously into account the pump irradiance and emitters lifetime modifications in the system. We finally apply this formalism to describe gain–assisted propagation in a dielectric–loaded surface plasmon polariton waveguide.

OCIS codes: (240.6680) Surface plasmons; (230.4480) Optical amplifiers; (020.5580) Quantum electrodynamics.

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1. Introduction

Surface plasmon polariton (SPP) results from coupling a surface charge density to an electromagnetic wave. Metallic strips or nanowires are supporting propagating SPP with mode area that could be below the diffraction limit. As such, they are subject of intense research in view of integrated opto-electronics applications [1]. However, due to the presence of the metal, Joule losses are always present. These losses, which are of interest for specific applications such as thermo-optical devices [2, 3], are nevertheless most of the time unwanted since they limit the waveguide performances. Therefore, strong efforts have been put forwards recently to realize a *plasmonic amplifier*. Gain–assisted propagation can be implemented if a gain region is placed next to the metal interface [4] and even SPP amplification can be achieved above a certain threshold [5–7]. Because the effective volume of the mode is reduced, plasmon gain can be achieved at low threshold compared to photonic equivalent system [5].

For the design and optimization of such plasmonic amplifier, direct transposition of standard photonic model to plasmonic system can be extremely useful. Importantly, the excited emitters that constitute the gain medium can either incoherently or coherently couple to SPP [8, 9] so that these competitive effects have to be carefully taken into account to analyse SPP propagation in presence of a gain medium. Amplified spontaneous emission of SPP also limits the gain efficiency [10]. Recently, de Leon and Berini adapted dye laser modelling to long-range surface plasmon polariton waveguide [11,12]. Particularly, they treat carefully the competition between coherent stimulated emission of SPP, responsible for gain, and incoherent surface plasmon coupled (spontaneous) emission, extremely efficient for small emitter-metal separations [13–15]. However, their model considers infinitely lateraly extended waveguide system. In this article, we adapt this model for finite arbitrary 2D-cross section plasmonic waveguide.

Our model relies on the description of the (local) density of guided modes for both passive and active configurations. Passive system, already presented in a previous paper [16] is briefly

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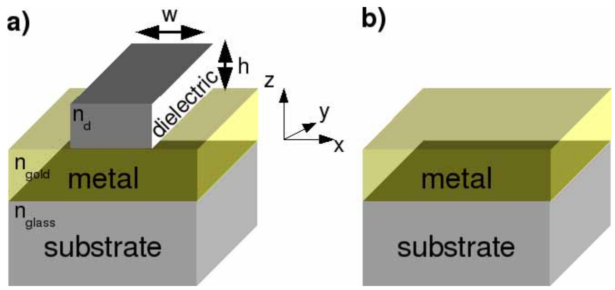

Fig. 1. a) DLSPPW configuration. A $w \times h$ dielectric strip (optical index $n_d = \varepsilon_d^{1/2}$) is deposited on a gold film, supported by a substrate. b) Corresponding so-called reference geometry consisting of the same system without the dielectric strip.

presented in section 2 as a starting point of our discussion. In the next section, we investigate spontaneous coupling of dipolar quantum emitters to guided modes. The contribution of spontaneous and stimulated emission to the optical gain is finally discussed in section 4. As an example, we also apply the developed model to the experimental situation described in Ref. [4].

In this article, we focus on the dielectric–loaded surface plasmon polariton waveguide (DLSPPW) (Fig. 1), a configuration for which experimental data about amplification were reported recently [4]. A dielectric strip, possibly containing a gain medium, is deposited on a metal film to laterally confine the SPP. The bare multilayer system (substrate/metal/air) will be referred as the reference system (Fig. 1b).

2. Density and local density of guided modes

We have previously established the behavior of the density of modes (DOS, unit length) near a waveguiding resonance [16]. The DOS is computed from the 2D-Green's dyad $\mathbf{G}^{2D}$ associated with the system according to

$$\rho^{2D}(k_y) = -\frac{2k_y}{\pi} Im \int d\mathbf{r}_{//} \varepsilon(\mathbf{r}_{//}) \mathbf{G}^{2D}(\mathbf{r}_{//}, \mathbf{r}_{//}, k_y) \,, [\mathbf{r}_{//} = (x,z)] \,. \tag{1}$$

where k_y is the wavevector component along the (invariant) waveguiding axis. This leads to define a *local* density of guided modes (LDOS, unit lenght^{-1}) as

$$\rho^{2D}(\mathbf{r}_{//}, k_y) = -\frac{2k_y}{\pi} Im \mathbf{G}^{2D}(\mathbf{r}_{//}, \mathbf{r}_{//}, k_y) \,, \tag{2}$$

so that the density of guided modes can be written as $\rho^{2D}(k_y) = \int d\mathbf{r}_{//} \varepsilon(\mathbf{r}_{//}) \rho(\mathbf{r}_{//}, k_y)$. Importantly, in Ref. [16], we demonstrated that near a resonance $k_y = k_{y0}$, the difference between the waveguide DOS and the DOS of a reference system (substrate/metal/air in the present case) follows a Lorentzian profile given by

$$\Delta\rho^{2D}(k_y) = \rho^{2D}(k_y) - \rho^{2D}_{ref}(k_y) \approx \frac{g_s}{\pi} \frac{1/2L_{SPP}}{(k_y - k_{y0})^2 + (1/2L_{SPP})^2} \,. \tag{3}$$

where g_s indicates the mode degeneracy, and L_{SPP} the mode propagation length. The density of guided modes is centered at the mode propagation constant k_{y0}, with a full width at half maximum (FWHM) $1/L_{SPP}$. One defines equivalently the complex effective index of the mode $\tilde{n}_{eff} = n'_{eff} + i n''_{eff}$ with $n'_{eff} = k_{y0}/k_0$ and $L_{SPP} = 1/(2k_0 n''_{eff})$.

#129802 - $15.00 USD Received 8 Jun 2010; revised 6 Jul 2010; accepted 7 Jul 2010; published 19 Jul 2010

As a consequence, it is reasonable to assume that a modification of the local density of guided modes follows a similar profile

$$\Delta\rho^{2D}(\mathbf{r}_{//},k_y) \approx \frac{\Delta\rho^{2D}(\mathbf{r}_{//},k_y^0)}{(2L_{SPP})^2}\,\frac{1}{(k_y-k_{y0})^2+(1/2L_{SPP})^2}\,. \quad (4)$$

This assumption has been checked by computing the LDOS profiles at different locations within the waveguide for k_y values spanned accross a resonance. As shown on Fig. 2, the expected Lorentzian shape is obtained at each observation point.

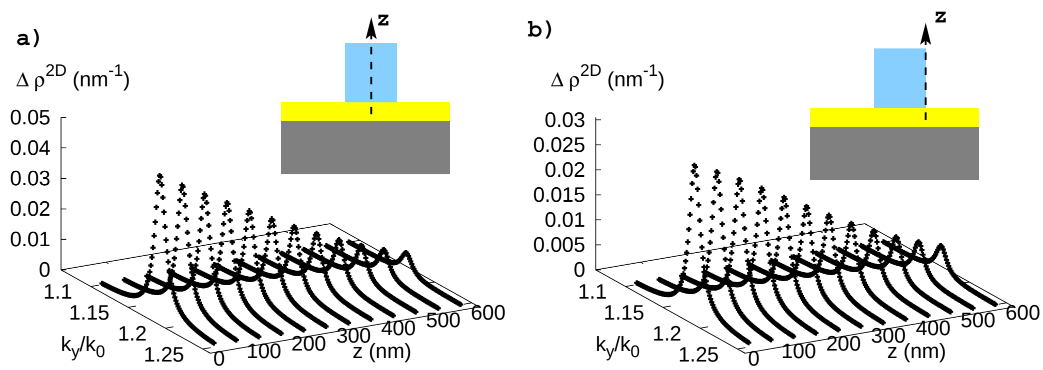

Fig. 2. LDOS calculated at the waveguide center (a) and edge (b) in function of both normalized propagation constant k_y/k_0 and height z in the dielectric strip. DLSPPW is a $w = 400$ $nm \times h = 600$ nm polymer strip ($n_d = 1.5$) on a 40 nm thick gold film. The wavelength is $\lambda = 1.55$ μm.

3. Dipolar emitters coupled to guided modes

The key step in the description of the coupling of (dipolar) quantum emitter with a DLSPPW mode is to evaluate the 3D-LDOS $\rho(x,z)$. In the present case, $\rho(x,z)$ gives the emission of point–like dipole source embedded in a 2D-geometry and is usually referred as 2.5D LDOS. Specific numerical technics have been developed to evaluate the LDOS in e.g. photonic crystals [17] or cylindrical plasmonic waveguides of arbitrary cross–section [18]. However, the presence of the substrate and the gold film in the present geometry renders difficult the application of such methods. Therefore, we propose here an original approach that allows us to calculate the 3D-LDOS for an arbitrary 2D-geometry. Moreover, and more importantly, this will lead us to a compact and general expression for the emitter coupling rate to a guided mode with a straightforward physical meaning. This method relies on the Fourier transform of the 3D-Green's tensor $\mathbf{G}$. Indeed, the LDOS is expressed as

$$\rho(\mathbf{r}) = -\frac{k_0^2}{\pi\omega} Im Tr \mathbf{G}(\mathbf{r},\mathbf{r})\,. \quad (5)$$

Additionally, the 3D-Green's tensor can be linked to the 2D-Green's tensor $\mathbf{G}^{2D}$ by a Fourier transform [17]

$$\mathbf{G}(\mathbf{r},\mathbf{r}') = \int_{-\infty}^{+\infty} dk_y \mathbf{G}^{2D}(\mathbf{r}_{//},\mathbf{r}'_{//},k_y)e^{-ik_y(y-y')} \quad (6)$$

#129802 - $15.00 USD Received 8 Jun 2010; revised 6 Jul 2010; accepted 7 Jul 2010; published 19 Jul 2010

so that

$$\rho(\mathbf{r}) = -\frac{k_0^2}{\pi\omega} ImTr\mathbf{G}(\mathbf{r},\mathbf{r}) \tag{7}$$

$$= -\frac{k_0^2}{\pi\omega} ImTr \int_{-\infty}^{+\infty} dk_y \mathbf{G}^{2D}(\mathbf{r}_{//},\mathbf{r}_{//},k_y) \tag{8}$$

$$= -\frac{k_0^2}{\pi\omega} \int_{-\infty}^{+\infty} dk_y ImTr\mathbf{G}^{2D}(\mathbf{r}_{//},\mathbf{r}_{//},k_y) \tag{9}$$

$$= \rho_{ref}(\mathbf{r}) - \frac{k_0^2}{\pi\omega} \int_{-\infty}^{+\infty} dk_y ImTr\Delta\mathbf{G}^{2D}(\mathbf{r}_{//},\mathbf{r}_{//},k_y)\,, \tag{10}$$

$$\text{with} \quad \Delta\mathbf{G}^{2D} = \mathbf{G}^{2D} - \mathbf{G}^{2D}_{ref}\,. \tag{11}$$

$\mathbf{G}^{2D}_{ref}$ represents the 2D-Green's dyad associated to the reference system, namely, the substrate/metal/air structure. Writing the last equation, we explicitely separate the multilayer contribution (ρ_{ref}) since it can be numerically evaluated by the well-known Sommerfeld expansion of dipole emission [19]. Moreover, profiting of the 2D-density of states profile near a resonance [16], the last integration is easily achieved. Indeed, the modification $\Delta\rho^{2D}$ of the 2D-density of modes explicitely appears in Eq. 10. Specifically, introducing Eq. 4, describing the Lorentzian behaviour near the resonance, leads to

$$\rho(\mathbf{r}) = \rho_{ref}(\mathbf{r}) - \tfrac{k_0^2}{2\omega L_{SPP}} ImTr\Delta\mathbf{G}^{2D}(\mathbf{r}_{//},\mathbf{r}_{//},k_y^0) \tag{12}$$

$$= \rho_{ref}(\mathbf{r}) + \tfrac{\pi k_0^2}{4\omega k_y^0}\tfrac{1}{L_{SPP}}\Delta\rho^{2D}(\mathbf{r}_{//})\,, \tag{13}$$

$$\text{with} \quad \Delta\rho^{2D}(\mathbf{r}) = -\tfrac{2k_y}{\pi} ImTr\Delta\mathbf{G}^{2D}(\mathbf{r}_{//},\mathbf{r}_{//},k_y^0)\,. \tag{14}$$

Equation 13 is an important result of this paper since it writes the 3D-LDOS modification in the simple form $\Delta\rho(\mathbf{r}) \propto L_{SPP}^{-1}\Delta\rho^{2D}(\mathbf{r}_{//})$ and have a straightforward interpretation. The dipole emission rate is proportional to the overlap efficiency between the dipole emission and the guided mode ($\Delta\rho^{2D}(\mathbf{r}_{//})$) and is proportional to the loss rate ($\Gamma_{loss} = 1/(2L_{SPP})^{-1}$). Since $\Gamma_{loss} = \Gamma_i + \Gamma_{rad}$ includes both the intrinsic (non radiative, Γ_i) and radiative losses (Γ_{rad}) rates [20], Eq. 13 properly takes into account both the non–radiative and radiative decay rates modifications by coupling to the waveguide modes. Note however, that it only describes the dipolar emision coupling to the available guided modes. Emission scattering on the dielectric strip could also contribute to modify the radiative decay rate of the emitter. This process is however negligible for quantum emitters located inside the waveguide.

4. Surface plasmon coupled emission: spontaneous and stimulated emission of SPPs

We have obtained in the preceding paragraph an explicit expression of the 3D-LDOS describing the emission of a dipolar quantum emitter in a 2D geometry. This quantity is one of the main ingredient for modelling spontaneous and stimulated emission of SPPs. Indeed, according to the de Leon & Berini model, the small gain coefficient $g(x,z)$, deduced from rate equations, is *locally* defined as [11,12]:

$$g(x,z) = N\frac{I_p(x,z)\tau(x,z)\sigma_p\sigma_e - \sigma_a\hbar\omega_p}{I_p(x,z)\tau(x,z)\sigma_p + \hbar\omega_p} \tag{15}$$

where N is the emitter density, I_p is the pump irradiance, σ_p and σ_a are the absorption cross-section at the pump and emission wavelengths, respectively and $\hbar\omega_p$ is the pump photon energy.

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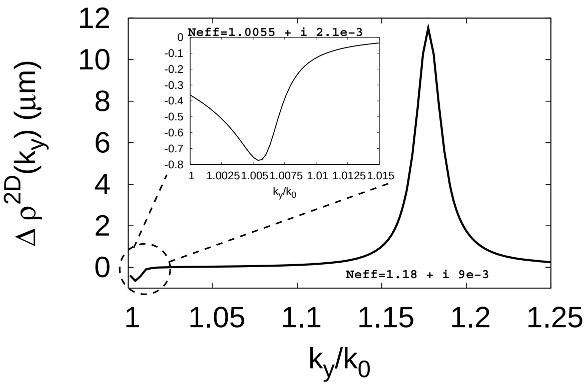

Fig. 3. 2D-DOS variation in function of wavector component k_y along the DSLPPW axis. The inset shows a zoom near the Au/air SPP mode effective index.

σ_e is the stimulated–cross-section of the emitters in their excited states. Finally, τ is the lifetime of the excited level of the emitter which obeys the Fermi's golden rule

$$\frac{\tau(x,z)}{\tau^0} = \frac{\rho^0}{\rho(x,z)}, \tag{16}$$

where τ_0 and ρ_0 are respectively the lifetime and the density of states in vacuum.

In presence of the DLSPPW, both pump irradiance and emission lifetime are modified, justifying the position dependence (x,z) clearly indicated in Eq. 15. The pump irradiance is given by [21]

$$I_p(x,z) = \frac{\varepsilon_0 c}{2} n_d |\mathbf{E}_p(x,z)|^2 \tag{17}$$

in the dielectric strip. $\mathbf{E}_p$ represents the pump excitation field that can be also calculated using the Green's dyad formalism

$$\mathbf{E}_p(\mathbf{r}_{//}) = \mathbf{E}_{inc}(\mathbf{r}_{//}) + k_0^2 \int d\mathbf{r}'_{//} \mathbf{G^{2D}}(\mathbf{r}_{//}, \mathbf{r}'_{//}, k_y^{inc})(\varepsilon_{ref} - \varepsilon_d)\mathbf{E}_{inc}(\mathbf{r}'_{//}) \tag{18}$$

where $\mathbf{E}_{inc}$ is the incident field and k_y^{inc} the incident wavector component along the invariant y-axis.

We apply the formalism described above to a recent experiment we realized, demonstrating gain-assisted propagation in a DLSPPW. In Ref. [4], we used a simplified model, assuming homogeneous gain in the doped strip. Except for the QDs absorption cross-section at telecom wavelength ($\sigma_a = 2.510^{-16}\ cm^2$), all the parameters are identical to those already used in Ref. [4]. Stimulated emission cross–section is the only unknown parameter, that we consider as a free parameter to adjust in order to reproduce experimental data.

Figure 3 represents the 2D-DOS variation. Clear antiresonance and resonance appear at the gold/air SPP effective index ($n_{eff}^{Au/air} = 1.0055$) and DLSPPW guide mode effective index ($n_{eff}^{DSLPPW} = 1.18$), respectively. These are the only two modes contributing to the 3D-LDOS modification in Eq. 13. Specifically, the negative 2D-DOS contribution near $k_y = 1.0055k_0$ indicates that the QDs couple less to the Au/Air SPP mode compared to the bare metal film wheareas the DOS enhancement near $k_y = 1.18k_0$ reveals a strong coupling to the guided DLSSPW mode, as expected.

The small gain coefficient is calculated using Eq. 15 (see Fig. 4a) where the QDs excited state lifetime τ modification is obtained from Fermi's Golden rule (Eq. 16) taking into account both the Au/air and DSLPPW guided mode contributions in Eq. 13. The incident angle is fixed to 45° to reproduce the experimental setup (see supplementary information in Ref. [4]). Figure 4a) shows strong inhomogeneities of the small gain coefficient within the doped polymer strip. Non-radiative coupling to the metal leads to absorption instead of gain for the emitters located within the first nanometers near the gold film ($g < 0$). Almost everywhere else, the small gain coefficient is in the range $150 - 210\ cm^{-1}$ so that efficient stimulated emission of SPPs could occur. The two green regions correspond to small gain coefficient about $100\ cm^{-1}$, below the average value. It originates from the small pump depletion at theses locations. However, it also participates in reducing the losses of the guided SPP. Finally, the gain of the guided mode depends on the overlap between the SPP and the gain coefficient profiles. SPP profile is presented in Fig. 4b). A better overlap with the small gain coefficient would be expected for QDs pumped with a propagating SPP at $\lambda = 532\ nm$. However, the low propagation length of the SPP at this wavelength would lead to a restricted active region along the waveguide compared to the large pumping area used in our experiment.

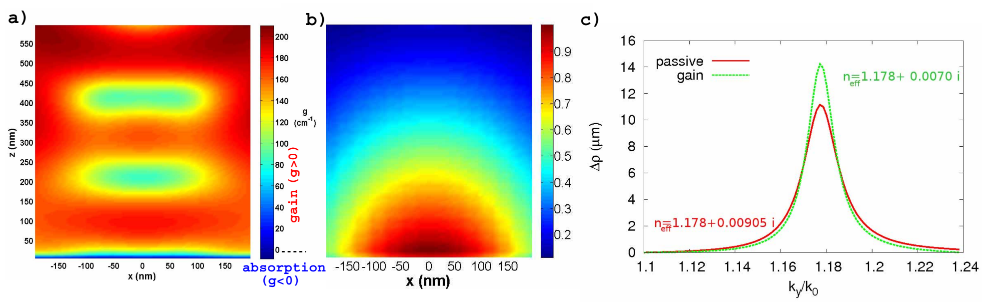

Fig. 4. a) Small gain coefficient within the doped wavequide calculated using Eq. 15 at pump irradiance $1000 W.cm^{-2}$ incident from the top at incident angle 45° and wavelength $\lambda = 532\ nm$. b) Guided SPP mode profile. c) Guided modes DOS calculated for null pump irradiance (passive) and $Ip = 1000\ W.cm^{-2}$ pump irradiance (gain). An offset has been added so that the two curves coincide far from the resonance. The complex mode effective index, deduced from a Lorentzian fit, is indicated on the figure.

Last, the active medium is represented by an imaginary dielectric constant $Im[\varepsilon(x,z)] = -\lambda n_d g(x,z)/2\pi$. The density of guided modes is represented on figure 4c) for passive and gain medium. The propagation length in the passive configuration is $L_{SPP} = 1/(2k_0 n''_{eff}) = 13.6\ \mu m$. At pump irradiance $Ip = 1000\ W.cm^{-2}$, we observe a resonance narrowing and the propagation length is increased to $L_{SPP} = 17.6\ \mu m$, thanks to optical gain. The stimulated emission cross-section was fixed to $\sigma_e = 4.10^{-19}\ cm^2$ to reproduce the propagation length measured for an incidence pump irradiance of $Ip = 1000\ W.cm^{-2}$. Previously, assuming a homogeneous gain within the pump doped polymer strip, we adjusted this value to $\sigma_e = 2.4\ 10^{-19}\ cm^2$ (in Ref. [4], we erroneously wrote $\sigma_e = 3.\ 10^{-19}\ cm^2$). The incoherent spontaneous emission coupling of quantum emitters to the metal has to be counterbalanced by an increase of the medium gain [6, 11]. We note however that mode confinement within the waveguide limits the effect since the estimated stimulated cross-section is of the same order even if quenching is neglected.

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5. Conclusion

On the basis of numerical evaluation of the density of guided modes and the local density of guided modes, we were able to fully investigate spontaneous and stimulated emission of SPPs in a dielectric-loaded surface plasmon-polariton waveguide. To this aim, we demonstrated an original relation that expresses the spontaneous decay rate as a ratio between quantum emitter emission overlap with the guided mode and its propagation length. This simple relation, as well as the full numerical model presented here, could be of interest for the design of an integrated plasmonic amplifier. Additionally, we would like to mention that this model could also be adapted to the absorption based plasmonic modulator described in Ref. [22]. It would be also important to discuss the linear gain saturation by non linear losses [23]. Last, Eq. 13 that expresses the coupling rate of a point–like dipolar quantum emitter with guided modes can be generalized to various configurations. Beyond its interest for numerical modelling we would like to emphasize that Eq. 13 brings important physical informations concerning the coupling of an emitter to waveguided modes.

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