

Loss compensated extraordinary transmission in hybridized plasmonic nanocavities

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Abstract. Extraordinary optical transmission has been utilized in many optical applications, but the plasmonic losses hinder their full potential. To obtain enhanced transmission, one of the loss compensation methods is to introduce gain. However, an enhanced transmission or even eliminated absorption does not guarantee plasmonic loss compensation. Here, we reveal the distinction between the transmission enhancement mechanisms in gain-assisted plasmonic arrays. To uncover the underlying mechanisms of the modified transmissions, we calculate the effective electric permittivity by employing a self-consistent gain model. We demonstrate that a large transmission enhancement in a plasmonic system composed of periodic nanocavities and coaxially placed nanoislands, is led by the loss compensation, which manifests itself as narrowing in effective permittivity. In contrast, a slight transmission enhancement in a plasmonic array without the nanoislands arises from the background amplification.

Keywords: extraordinary optical transmission, plasmon hybridization, gain, loss compensation

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1. Introduction

One of the most attractive plasmonic phenomena is Extraordinary Optical Transmission (EOT), where “extraordinarily” large fraction of incident light is transmitted through an array of periodic subwavelength apertures in a metal film [1, 2]. This invalidates Bethe’s classical aperture theory, which predicts extremely low transmission [3] through

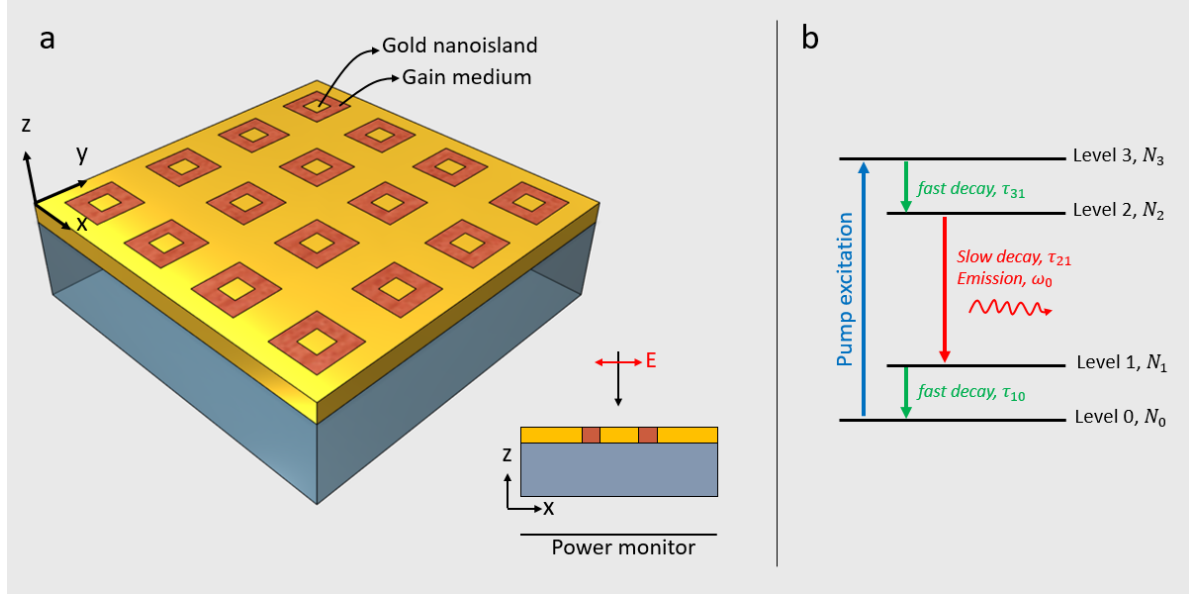


Figure 1. (a) Schematics of the EOT nanostructure. The periodicity (unit cell size) is 300 nm, cavity size is 170 nm, core size and thickness of the perforated gold film is 70 nm. (b) Energy state diagram of the 4-level atom, which is used for modelling the gain medium.

a subwavelength aperture on a perfect metal screen. In a periodic metal nanoaperture array, surface plasmon polaritons (SPPs) can lead to an enhanced electromagnetic field at the metal-dielectric interface, as well as in the apertures. The energy is transferred to the far field on the backside and observed as an enhanced transmission for certain wavelengths. The spectral positions of the EOT resonances are determined by the periodicity, and the optical properties of the metal and the dielectric, as the SPP resonances are governed by these properties [4]. The factors affecting the EOT response, such as the hole shape [5–9], film thickness [10, 11], and type of metal [12] have already been analyzed.

One of the drawbacks of light confinement is the ohmic loss. Strategies such as synthesizing alternative low-loss plasmonic materials [13] or using gain [14, 15] to compensate the loss have been previously developed. These methods have also been implemented in EOT systems to enhance transmission further via plasmonic loss mitigation [16–20]. In these works, the properties of a gain-assisted system have been investigated by the transmission spectrum and the dielectric function of the gain material, at different pump rates. However, examining the modifications in the dielectric properties of the gain material, as previously suggested, is not completely sufficient to explain the modifications in the transmission. As the gain and the plasmonic components are not independent from each other, the combination of these two components lead to an overall effect [21, 22]. This effect can be governed either by loss compensation as a result of near-field coupling, or background amplification, due to indirect coupling between gain and plasmonic materials, via background electromagnetic

waves [23, 24]. While transmission can be enhanced and absorption can be reduced or eliminated in both of the cases; the loss compensation manifests itself as an increase in the quality factor of the effective response. Hence, the underlying mechanism of the enhanced transmission in a gain incorporated plasmonic array can be exclusively revealed by a self-consistent treatment, as we present here, which calculates the effective parameters of the overall system.

In this paper, we investigate the optical response of an EOT system consisting of periodic apertures (nanocavities) and nanoislands, placed in the middle, where the gaps are filled with gain material. We demonstrate that the incorporation of gain medium provides a significant transmission enhancement at moderate pump rates. The effective parameters are retrieved using S-parameter inversion to analyse the system. We show that due to strong local field enhancement of nanoislands, gain medium couples to the plasmonic local field, modifying the effective permittivity, whereas in the case without the nanoislands, amplified gain simply adds on the plasmonic loss without coupling.

2. Plasmonic response

First, the EOT system (Figure 1(a)) without the gain material is investigated by finite difference time domain (FDTD) simulations [25]. We perform the three-dimensional simulations in commercially available software, Lumerical FDTD. We use tabulated experimental data provided by Johnson and Christy [26] for Au dielectric function and set the refractive index to 1.45 for glass substrate. The surrounding medium in the upper side of the nanostructure, as well as the gaps are air ($n=1$). A plane wave source, linearly polarized in the x direction and propagating in the z direction illuminates the unit cell (see the cross section view in Figure 1) of the nanostructure from the air side, and a monitor on the back side collects the transmitted fields. The boundary conditions are set to periodic in the directions parallel to the source propagation (x, y), and perfectly matched layers in the direction perpendicular to the source propagation (z). Conformal meshing within the FDTD domain is used in simulations, while a finer mesh constraint of 5 nm is employed in the region enclosing the plasmonic unit cell, to get a better resolution. Figure 2(a) shows that the transmittance spectra of the nanocavities with and without the nanoislands present resonance in both cases. We observe that the transmittance is 67%, at 656 nm for the cavity array without the cores. The cavities cover about 32% of the total area of the metal film. Therefore, the effective transmittance, which is defined by the transmittance normalized to the gap fraction, is 210%. This EOT resonance is associated with the short-range surface plasmon polaritons (SR-SPPs) excited by grating coupling [27]. Since the metal film thickness is small compared to the wavelengths of the incident electromagnetic field, the propagating plasmons on each metal-dielectric interface are coupled. SR-SPPs exhibit a large density of electromagnetic states boosting the EOT peak, while highly delocalized long-range surface plasmon polaritons (LR-SPPs) does not contribute to the transmission. In this regime, the spectral position of the EOT resonance is red-shifted

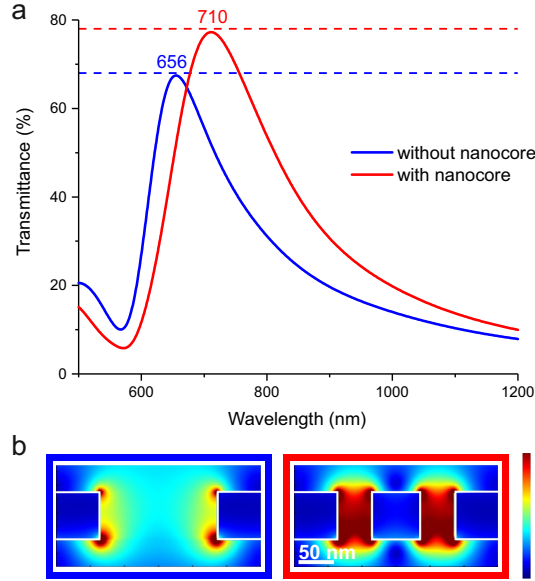


Figure 2. (a) Transmittance spectra of the EOT systems without (blue) and with (red) the nanoislands. (b) Electric field profiles on the xz plane for the unit cell, for the cases without (left) and with (right) the nanoislands, at the EOT resonances, 656 nm and 710 nm, respectively. White borders indicate the metal (Au) boundaries. In the color scale, maximum electric field magnitude is indicated by red, and minimum by blue.

a few hundreds of nanometers from the Rayleigh resonance [27] ($\lambda_R = n_s p = 435$ nm, where $n_s = 1.45$, the index of the substrate, and $p = 300$ nm is the array periodicity), as observed.

On the other hand, the transmittance of the EOT system with nanoislands is 77%, at 710 nm. Despite the fact that the gap fraction is decreased by the presence of the nanoislands, the transmittance is enhanced further. In this case, the effective transmittance is 290%. The nanoislands effectively localize the incident light, creating hotspots in spaces between the cavity and core walls, resulting in higher amount of incident light to be transmitted through. The electric field profiles calculated for each case; without and with the nanoislands, at the EOT resonances, are displayed with the same color scale in Figure 2(b). The fields are obtained from the three-dimensional simulations by a two-dimensional monitor lying on the xz plane, enclosing the unit cell of the periodic array. The presence of the nanoislands leads to a red-shift in the transmittance due to the hybridization of plasmon modes [28, 29] supported by the nanostructure. The nanoislands support localized surface plasmons, and the metal film itself supports propagating surface plasmons, where the phase matching condition is satisfied by the structure periodicity. The optical response of the overall system is determined by the hybridization of these two plasmon modes. Hence, the observed EOT resonance is shifted from the EOT resonance obtained in the absence of the nanoislands.

3. Gain incorporation

Next, we investigate the EOT system by embedding emitters into the gaps. We model the gain medium as a generic four-level atomic system [30], which dynamically and spatially associates the fields and the occupation numbers, and accounts for the energy exchange among the atom and the plasmonic field, pump field, as well as non-radiative decays. There are four energy levels ($i = 0, 1, 2, 3$) (Figure 1(b)), which can be occupied by the electrons in the atom (fluorescent dye molecule for this study). The occupation density of each level is denoted by $N_i(\mathbf{r}, t)$, where \mathbf{r} is the spatial position and t is the time. The total dye concentration is defined by \bar{N}_0 , where $\bar{N}_0 = N_0(\mathbf{r}) + N_1(\mathbf{r}) + N_2(\mathbf{r}) + N_3(\mathbf{r})$ at any time t . In the absence of the external pump, electrons are occupied only in the ground state, with all other excited states are empty. The response of the gain medium to the external pump source is determined by the polarization density, $\mathbf{P}(\mathbf{r}, t)$, which obeys the following semi-classical equation of motion,

$$\frac{\partial^2 \mathbf{P}(\mathbf{r}, t)}{\partial t^2} + \Delta\omega_0 \frac{\partial \mathbf{P}(\mathbf{r}, t)}{\partial t} + \omega_0^2 \mathbf{P}(\mathbf{r}, t) = -\sigma_0 \Delta N(\mathbf{r}, t) \mathbf{E}(\mathbf{r}, t), \quad (1)$$

where $\Delta\omega_0$ is the emission linewidth, ω_0 is the emission frequency of the dye molecules, $\sigma_0 = 6\pi c^3 \gamma_{rad} / (\sqrt{\varepsilon_h} \omega_0^2)$ is the coupling efficiency of the dye molecules to the external field $\mathbf{E}(\mathbf{r}, t)$, with c , γ_{rad} , and ε_h being the speed of light in vacuum, radiative decay rate of the molecules, and permittivity of the host medium, respectively, and $\Delta N(\mathbf{r}, t) = N_2(\mathbf{r}, t) - N_1(\mathbf{r}, t)$ is the population inversion that drives the polarization. Assuming the time harmonic polarization density, \mathbf{P} , and electric field, \mathbf{E} , and steady state population inversion, ΔN , (1) can be expressed as,

$$\mathbf{P}(\mathbf{r}) = \frac{\sigma_0 \Delta N}{\omega^2 + i\Delta\omega_0 \omega - \omega_0^2} \mathbf{E}(\mathbf{r}), \quad (2)$$

where the population inversion is expressed as [30],

$$\Delta N = \frac{(\tau_{21} - \tau_{10}) \Gamma_{pump}}{1 + (\tau_{32} + \tau_{21} + \tau_{10}) \Gamma_{pump}} \bar{N}_0, \quad (3)$$

with the assumption that the electric field is small enough to neglect nonlinear effects which may deplete it. Following from the definition of total displacement, $\mathbf{D}(\mathbf{r}) = \varepsilon_h \mathbf{E}(\mathbf{r}) + \mathbf{P}(\mathbf{r})$, the effective absolute permittivity of the gain medium is

$$\varepsilon_g = \varepsilon_h + \frac{6\pi c^3 \gamma_{rad} / (\sqrt{\varepsilon_h} \omega_0^2)}{\omega^2 + i\Delta\omega_0 \omega - \omega_0^2} \times \frac{(\tau_{21} - \tau_{10}) \Gamma_{pump} \bar{N}_0}{1 + (\tau_{32} + \tau_{21} + \tau_{10}) \Gamma_{pump}}. \quad (4)$$

Here, the radiative decay rate, γ_{rad} , can be obtained from quantum yield (QY), i.e., $\text{QY} = \gamma_{rad} \tau_{21}$. Based on (4), the constraints on the parameters of the gain medium are,

- (i) The emission wavelength should overlap with the plasmonic response of the passive (without gain) EOT system, and

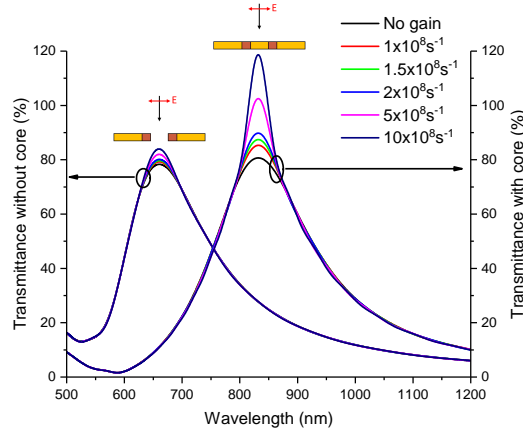


Figure 3. (a) Transmittance spectra of the EOT systems without (left axis) and with (right axis) the nanoislands for the passive case and active cases with different pump rates.

- (ii) The radiative transition lifetime should be short (≤ 1 ns), while the emission linewidth should be narrow (≤ 50 nm) to generate large enough permittivity for the active system to compensate the loss for moderate pump rates.

IR140 dye emits in the spectral range of 830 – 880 nm with narrow linewidths (~ 50 nm), short lifetimes (~ 1 ns or shorter), and quantum yield of $\sim 20\%$ [31,32]. Therefore, it meets the requirements stated above, to be used in the nanocavity-nanoisland system. For the system without the nanoislands, most of the dye molecules used in fluorescence applications, with emission resonance at ~ 660 nm would meet the requirements, i.e., short lifetime and narrow linewidth, for example Alexa Fluor dyes [33]. The gain parameters that we use in the S-parameter calculations are given in Table 1.

Considering the fact that these dye molecules will be embedded in a host medium; filling the cavities with a host material with $n = 1.45$ (i.e., PMMA resist) shifts the EOT resonance, from 656 nm to 660 nm for the plasmonic array without the nanoislands, and from 710 nm to 832 nm, for the case with the nanoislands. We also note that, for the rest of the paper, the results are obtained from simulations of free-standing EOT

Table 1. Gain medium parameters

Parameter name	Parameter value
Permittivity of the host medium, ϵ_h	1.45 ²
Radiative transition lifetime, τ_{21} (ns)	1
Non-radiative transition lifetime, $\tau_{32,10}$ (ps)	1
Quantum yield	0.2
Emission frequency, ω_0 (Prad/s)	2.264, 2.854
Emission linewidth, $\Delta\omega_0$ (Prad/s)	$\omega_0/16$
Gain concentration, \bar{N}_0 (m ⁻³)	6×10^{24}
Pump rate, Γ_{pump} ($10^8 \times \text{s}^{-1}$)	1 – 10

structures (instead of structures on glass substrates), in order to simplify the calculations of effective parameters. If the systems were simulated with a glass substrate, the EOT resonances would shift ~ 30 nm to longer wavelengths.

Figure 3 shows the calculated transmittance spectra of the EOT systems without the nanoislands (left vertical axis), and with the nanoislands (right vertical axis) for the passive case (no gain) and active case with different pump rates of $1 \times 10^8 s^{-1}$, $1.5 \times 10^8 s^{-1}$, $2 \times 10^8 s^{-1}$, $5 \times 10^8 s^{-1}$, and $10 \times 10^8 s^{-1}$. To make a fair comparison between the cases with and without the nanoislands, we replace the dielectric function of the metal nanoisland with that of air ($n = 1$) to simulate the gain incorporated plasmonic array without the nanoislands. Hence, the volume of the gain material (number of emitting dyes) used in each case is the same. It is clearly seen that increasing the pump rate results in a slightly enhanced transmittance for the plasmonic array without the nanoislands, whereas the same amount of increase in the pump rate significantly enhances the transmittance of the plasmonic array with the nanoislands. For the case of nanoislands, transmittance peaks exceed 100% when the pump rate is large enough. We note that even the largest pump rate used in the simulations is a moderate pump rate, which refers to an amount that is well below the damaging threshold. The minima of the calculated reflectance spectra (not shown) at the resonances are at zero, and the reflectance spectra is not altered with different pump rates for any of the plasmonic arrays. Consequently, the absorbance of the plasmonic arrays is suppressed as the pump rate increases. Furthermore, at a sufficiently large pump rate, which is approximately $5 \times 10^8 s^{-1}$ for the plasmonic array with the nanoislands, the absorbance is completely eliminated; and at larger pump rates, it becomes negative.

4. Loss compensation

In order to reveal the full physical mechanism, we retrieve the effective optical parameters (i.e., effective permittivity, effective refractive index) using effective medium theory which incorporates S-parameter inversion [34]. This allows us to observe how the optical properties of the overall system around the plasmonic resonance is modified with different pump rates. Effective medium approach treats the EOT system, which is an inhomogeneous composition of Au and gain media, as if it is a homogeneous structure possessing effective dielectric properties. The S-parameters, which are the complex reflection and transmission coefficients (in contrast to the real power reflection and transmission coefficients), are extracted by the amplitude and phase of the reflected and transmitted fields, obtained in FDTD simulations. For the given system, the S-parameters are defined as, $S_{11} = E_r/E_i$, and $S_{21} = E_t/E_i$, where E_i , E_r , and E_t are the incident, reflected and transmitted electric fields respectively. Once the S-parameters are obtained, the effective parameters are calculated using the definitions of the effective refractive index and effective impedance, as the following [34],

$$n_{eff} = \frac{1}{kd} \cos^{-1} \left(\frac{1 - S_{11}^2 + S_{21}^2}{2S_{21}} \right), \quad (5)$$

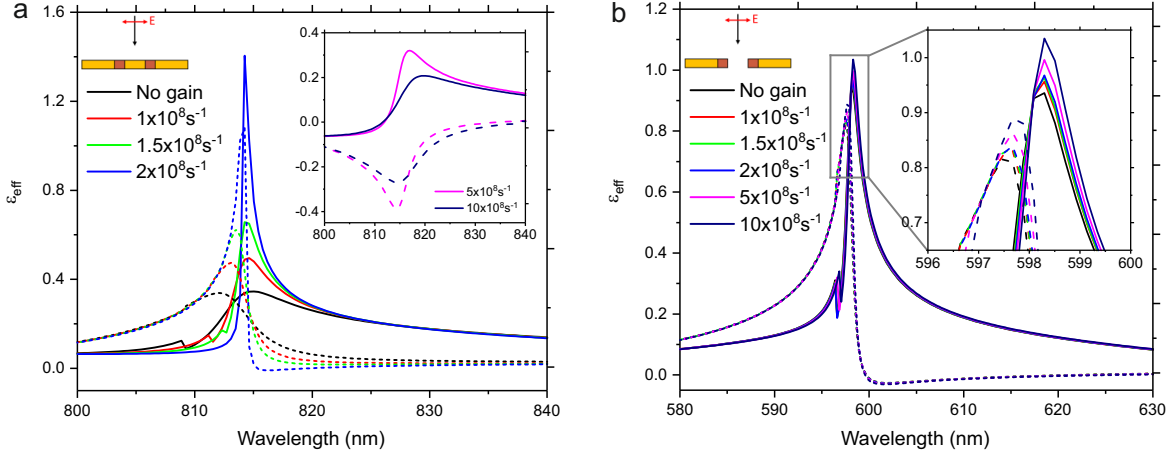


Figure 4. Real (solid) and imaginary (dashed) parts of the effective electric permittivity retrieved by effective medium theory using S-parameters inversion, for the plasmonic array with (a) and without (b) the nanoislands.

$$z_{eff} = \sqrt{\frac{(1 + S_{11})^2 - S_{21}^2}{(1 - S_{11})^2 - S_{21}^2}}, \quad (6)$$

where, k and d are the propagation constant and the film material thickness, respectively. Then, effective permittivity is calculated using,

$$\epsilon_{eff} = \frac{n_{eff}}{z_{eff}}. \quad (7)$$

Figure 4(a) shows how the presence of the gain medium modifies the effective electric permittivity at different pump rates, for the plasmonic array with the nanoislands. Comparing the no-gain case with gain pumped at rates, $1 \times 10^8 s^{-1}$ to $2 \times 10^8 s^{-1}$, we observe that the resonance becomes narrower with increasing pump. This modification is an indicator of coupling between gain and plasmon field. More specifically, plasmonic field interacts with gain to form a new, less damped field, with sharpened resonant response, which subsequently homogenize into a new, loss compensated, bulk system. Further increase of the pump rate (inset of Figure 4(a)) changes the imaginary part of the effective permittivity to negative values, fully compensating the absorptive loss for pump rate of $5 \times 10^8 s^{-1}$, and then broadens the spectral feature, which corresponds to the over-compensation for $10 \times 10^8 s^{-1}$ pump rate [23]. On the other hand, as presented in Figure 4(b), the presence of the gain medium does not lead a narrowing in the imaginary part of the effective permittivity when there are no nanoislands. In fact it makes the resonance become slightly broader (see the inset of Figure 4(b)). In this case, the gain is not affected from the near-field, but the incident field, and just is added on transmitted light; resulting in slightly higher transmission.

Figure of merit ($FOM = |\text{Re}(n)/\text{Im}(n)|$) of the EOT systems (Figure 5) quantifies the resonance quality. In both cases, FOM in the absence of the gain is almost the same. However, increasing the pump rates boosts the FOM for the plasmonic array

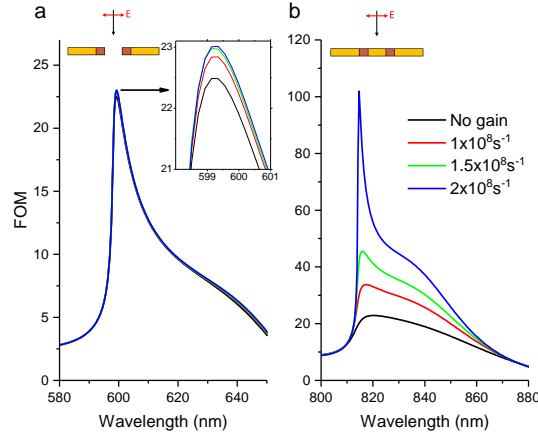


Figure 5. Figure of merit ($\text{FOM} = |\text{Re}(n)/\text{Im}(n)|$) for the plasmonic array without (a) and with (b) the nanoislands.

with the nanoislands, whereas it does not alter much for the plasmonic array without the nanoislands. We observe that for the pump rates, $5 \times 10^8 \text{ s}^{-1}$ and higher, the FOM becomes very large (~ 800).

5. Discussion and conclusions

In summary, we investigate the transmission enhancement mechanisms in a plasmonic array having EOT response, by gain incorporation. We examine the homogenized bulk response of the gain incorporated plasmonic array systems by calculating the effective optical parameters, in the absence and presence of the nanoislands. We conclude that the enhanced transmission is led by loss compensation in the plasmonic array with the nanoislands, whereas it is the background amplification of the gain which slightly enhances the transmission in the plasmonic array without the nanoislands. The difference between the two enhancement mechanisms, i.e., loss compensation and background amplification, lies on whether the gain is coupled to the plasmonic near field or not. In the plasmonic array with the nanoislands, the hotspots created by the plasmon excitation is much stronger than the one without the nanoislands. This is due to plasmon hybridization of the localized surface plasmons supported by the nanoislands, and the propagating surface plasmon polaritons excited at the metal-dielectric interface. In the case of very strong local field enhancement, the gain medium located at the hotspots couples with the plasmon mode and reshapes the optical effective permittivity of the overall system. However, when the field enhancement is not as strong as the case of the plasmonic array with the nanoislands, the gain does not directly couple to the plasmon field, but amplified by the plasmon mode. Without the nanoislands, the background propagation (due to the incident field) excites the passive system and the active gain medium individually, without the two systems being coupled to each other directly, which is the opposite extreme of loss compensation [24, 35]. We observe

that in the studied nanostructure systems, direct coupling between the gain medium and the plasmonic array results in a significantly enhanced transmission, whereas the background amplification mechanism only slightly enhances the resonance.

The numerically solved, self-consistent model, incorporating gain in EOT nanostructure systems could be a proper guide in designing experiments where plasmonic loss compensation is targeted. The design of the proposed structure can be further improved by employing different substrates and different quantum emitters.

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