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# Significant field enhancements in an individual silver nanoparticle near a substrate covered with a thin gain film

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**Abstract:** In this paper, we propose a method to significantly enhance the local-field of a gap plasmonic system by placing a metallic nanoparticle in close proximity to a substrate covered with a thin gain film (~100 nm thickness). Compared with a conventional dielectric substrate, the thin gain film can contribute to several, or dozens, of times more intense local electric fields in the gap between the particle and the substrate. We use the finite difference time domain method to numerically analyze the influences of the gain coefficient of the film and of the other parameters on the field enhancement. The numerical results show that there is an optimal refractive index of the gain film that enables to achieve a maximal field enhancement for a given NP radius. Moreover, the optimal refractive index of the gain film can be accommodated to any available materials by using metal nanoparticles with an appropriate radius.

#### Introduction

Metal nanoparticles (NPs) are very different from bulk materials and are considered to be very promising due to strong local surface plasmon resonance (LSPR), which results in producing intense optical scattering around the particles and significant optical near-field enhancement<sup>1-3</sup>. Among the different LSPR structures, gap plasmons obtained by bringing a metal NP close to a substrate surface can lead to record levels of field amplification in the gap between the NP and the substrate<sup>4</sup>, favoring wide applications in light trapping of solar cells<sup>5,6</sup>, surface-enhanced Raman scattering (SERS)<sup>7,8</sup>, and dark-field microspectroscopy<sup>9,10</sup>.

In fact, the ease with which the properties of localized nanoparticle plasmons are influenced by their local environment has made definitive measurements of their fundamental characteristics a significant ongoing challenge. As a result, the study of plasmonic NPs at the individual nanostructure level has become a major experimental and theoretical focus and has enabled numerous significant advances in our understanding of plasmons in nanoscale systems. It is very important to give a physical insight of the effect of the substrate on the plasmonic properties of the NP placed on its surface. The substrate provides a mechanism for symmetry breaking, which has been shown in other plasmonic systems to lift mode degeneracies and modify the coupling of the plasmon modes to the far field. Different technologies based on different substrate materials have been studied to support a gap plasmon system. Single metallic NPs on a smooth metallic surface can create high fields in the gap between the NP and the substrate<sup>11</sup>. The interaction between the NP and the metallic substrate results in strongly red-shifted plasmons due to hybridization between the localized plasmon modes of the NP and the propagating plasmon modes of the substrate<sup>11,12</sup>. The strength of these modes is critically dependent on the gap distance and consequently affects the field enhancement<sup>11</sup>. However, for a metallic NP near a dielectric surface, there are no substrate surface-plasmon modes; only the LSPR mode of the individual metallic nanoparticle interacts with the dielectric substrate<sup>11-15</sup>. In 2011, Spinelli et al. published a study regarding a silver nanoparticle placed on top of a highrefractive-index substrate. In their work, they studied the effect of the silver particle shape and of the substrate on the plasmon mediated light coupling to a high-index substrate. They showed

that, for spherical NPs, both the dipolar and the quadrupolar resonances are almost unaffected by the presence of the substrate<sup>16</sup>. Recently, Hutter *et al.* performed systematic simulations of the near-field and the enhancement in the gap, in order to provide a good understanding of the electric field enhancement in a system consisting of nanoparticle-on-dielectric-substrate separated by a nano-sized gap. They studied the effect of the optical constants of the substrate as a result of its interaction with the spherical nanoparticle, *e.g.*, the size of the NP and the gap value. In addition, they also showed how the optical properties of the dielectric substrate (n and k) affect the plasmonic field enhancement in the nano-gap<sup>11</sup>. However, only conventional absorption materials were considered in their analysis. Moreover, the near-field enhancement is not high enough for practical applications.

A problem for many applications of plasmonics and metamaterials is posed by losses inherent in the interaction of light with metals, even in gap plasmon systems. To overcome this limitation, a method by using gold nanobox particles with a gain medium embedded within their core region has been proposed to compensate the absorption in metals and maintain the superior plasmonic properties of the metal nanostructures<sup>17</sup>. In the present paper we will propose a simpler gap-plasmon system by placing a metallic NP very close to a substrate covered with a thin gain-film. An effective loss compensation is obtained when the metal NP is close to the surface of the thin gain film, so that several, or dozens, of times more intense local electric fields in the gap between the particle and the substrate can be achieved. Based on the present system, many important characteristics (such as the maximal field enhancement, the resonant wavelength, and the environmental sensitivity) for variable system parameters will also be accurately analyzed using a finite difference time domain (FDTD) method. Compared with conventional dielectric substrates, the new method has no high requirement on substrate refractive index, *i.e.*, a substrate with a low refractive index can also be used as long as we can synthesize the NP with an appropriate radius.

#### SIMULATION METHOD

We start our analysis by calculating the electric field of a silver NP on a substrate covered with a thin gain film by means of FDTD calculations. The simulation setup is shown in Fig. 1. Perfectly matched layers (PMLs) are used at the boundaries of all directions to absorb the scattered radiation in all directions (in order to eliminate reflection back to the model). The simulation box size is  $0.65 \times 0.65 \times 0.65 \ \mu\text{m}^3$ . A mesh size of 1 nm is used in the entire region containing the particle and at the substrate-environment interface, while a smaller (0.2 nm) mesh grid is used around the sharp features at the particle-substrate contact point. Before the analysis based on the present model in Fig. 1, we firstly compute the electric field for the substrate only (i.e., no NP is put above the gain film) to ensure the validity of the PML layer. Results show that a 100 nm PML is enough thick to remove the effect from any non-physical

scattering. Optical constants for silver were taken from Ref. 18 and fitted using a Drude-Lorentz model. In practice, the thin gain film in the system can be a semiconductor film fabricated by using a typical film deposition technology, e.g., magnetron sputtering, or also a conventional dielectric material doped with rare earth ions. For the present theoretical model, the refractive index of the thin gain film is represented by the letter n, while its gain coefficient (k) is assumed to be independent of frequency and, as in Ref. 19, is introduced through the dielectric imaginary part of the permittivity  $k = \text{Im}(\sqrt{\varepsilon' + i\varepsilon''})$ . Therefore, the refractive index of the thin gain film is n-ik, where a negative imaginary part means an active gain, and the thickness of the film is fixed to 100 nm in the following simulations. To verify our FDTD code, we have computed the plasmonic behavior of gold dimers and the results are consistent with those reported in Ref. 20.



Fig. 1. Schematic configuration (a) and calculation model (b) of a silver NP above a substrate covered with a gain film.

#### **RESULTS AND DISCUSSION**

#### 1. Effect of the gain coefficient of the thin gain film

Local electric fields are of great importance to many applications, e.g., solar cells, SERS and near-field microscopy. In the gap between the NP and the substrate, a region of high electromagnetic field, a so-called 'hot-spot' is generated. The gap can be created by a molecular spacer layer or by a transparent dielectric layer, and thus can be manipulated<sup>11</sup>.Based on the system shown in Fig. 1, we will firstly study the effect of the gain coefficient of the thin gain film placed above the substrate, on the near-field in the gap between the NP and the film surface. A numerical example of a typical gap plasmon system is used to illustrate the effect of the gain film. We choose the following parameters for the system: n=2 and  $n_s=1$  for the refractive indices of the thin gain film and the medium around the NP, respectively; the gap between the NP and the thin film is equal to 2 nm; the thickness of the gain film is equal to 100 nm; a silver NP (with radius R=80 nm) illumined with a *p*-polarized plane wave excitation at the upper boundary,  $E_x = E_0 \cdot \exp(ik_0z)$ , where  $E_0 = 1$  V/m, propagating in the -z direction.

The position of maximum electric field is not constant in space and it will change with the strength of the interaction between the NP and the dielectric surface. In addition, we have shown that the following variation trends of the enhanced electronic filed at different system parameters are similar no matter the maximal electronic field around the Ag NP, in the whole calculation region, or in the middle of the gap is considered in the numerical research, only with a slight difference of the absolute value of the field enhancement. Hence, in this study, we use the highest electric field (expressed as E) along a horizontal plane in the middle of the gap, as it will be a more appropriate indicator of the strength of the interaction between the NP and the dielectric substrate, rather than using the field at a fixed point. Figure 2(a) shows the plot of the normalized electric field  $|E|/|E_0|$  spectra for different gain coefficients of the film. Obviously, when the NP is placed above a thin gain film, the larger the gain coefficient, the stronger the interaction between the NP and the thin film. Figure 2(b) gives the electric field distributions at four peak wavelengths, marked with A, B, C, and D in Fig. 2(a). From this figure, one can see that a 0.4 gain coefficient of the substrate film can contribute to a field enhancement four times larger than that reached without gain film. The electromagnetic fields across a NP near a dielectric surface are different than for a NP in an isotropic environment. Substrate-induced changes have been understood qualitatively using a simple image theory<sup>21</sup>. The screening introduced by the dielectric surface is equivalent to the potential generated by a NP image with its image charges reduced by a factor of  $\eta = (\varepsilon - 1)/(\varepsilon + 1)$ . The gap system can simply be viewed as an interaction of the NP with its image with a strength determined by the NP-substrate separation, substrate permittivity, and NP structure. For a conventional gap system, the dielectric substrate with a larger permittivity will give rise to a stronger "image" and larger interactions. However, in the present work, the gain film with a negative imaginary part of the permittivity. Therefore, the complex factor not only can lead to a strong image, but also will contribute to a conjugated enhancement of the object itself.

In the numerical work, the used gain coefficients are dispersionless. However, any practical gain material has its specific gain spectrum. Typically, some rare earth doped materials can provide a gain spectrum with tens of nanometers bandwidth, and some semiconductor materials can provide a wider bandwidth of the gain spectrum, even up to 100nm. Moreover, these gain materials appear various distributions of the gain spectrum. In order to evaluate the effect of the dispersive behavior of the gain film, we calculate the normalized electric field  $|E|/|E_0|$  spectra for different bandwidths of the gain material with a 0.4 gain coefficiency, where the gain spectrum is sublimated as a Gaussian distribution, and all other parameters are the same as Fig. 2(a). One can see that the results obtained from the Gaussian gain spectrum agree very well with those obtained from the dispersionless gain model for the present gap system. Only when the wavelength is far away from the resonant peak, a slight difference occurs. Furthermore, the difference tends to

reduce as the bandwidth increases. Therefore, in the following numerical calculations, we will ignore the effect of the dispersion of the gain coefficiency to focus on the analysis of the field enhancement from the gain film.

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Fig. 2. (a) Plot of the normalized electric field  $|E|/|E_0|$  spectra for different gain coefficients of the film with a 2 nm gap distance between the gain film and the NP and an 80 nm particle radius. (b) The electric field distributions at four peak wavelengths, marked with A, B, C, and D in Fig. 2(a), respectively. (c) The normalized electric field  $|E|/|E_0|$  spectra for different bandwidths of the gain material with a 0.4 gain coefficiency, where the gain spectrum is sublimated as a Gaussian distribution, and other parameters are the same as Fig. 2(a).

Figure 3(a) shows the variation of the normalized electric field  $|E|/|E_0|$  at the resonant wavelengths (see Fig. 2(a)) as the gain coefficient of the film increases, for five given refractive indices of the film. The figure shows that the maximal local electric field increases almost exponentially as the gain coefficient increases. However, the rate of field enhancement versus the change of gain coefficient is different for different refractive indices of the gain film. From this figure, one can see that when  $n \sim 1.5$ , an optimal effect of field enhancement can be achieved for the present parameters. Figure 3(b) shows how the corresponding position of the resonant wavelength reported in Fig. 3(a) varies as the gain coefficient increases. The resonant wavelength hardly changes when the optimal n (*i.e.*, 1.5) is chosen. The resonant peak will shift towards a longer wavelength for a film with higher refractive index (e.g., n = 2) whereas it will shift towards a shorter wavelength for a lower refractive index (e.g., n = 1.3). Figure 3(c) shows the comparison of the electric field distribution of a system with film gain coefficient equal to 0.4 and to 0 (no gain coefficient of the film), at five different refractive indices of the gain film. Figure 3(d) shows how the maximum electric field around the NP varies as the refractive index of the substrate increases. For a non-gain dielectric substrate, the higher the refractive index of the substrate, the stronger the effect of field enhancement (see the circle line in Fig. 3(d)). Therefore, one has to use a dielectric substrate with high enough refractive index to obtain a strong local field enhancement for a conventional gap plasmon system<sup>16</sup>. Conversely, for the present gain-film assisted substrate, there is an optimal refractive index of the gain film where the local field in the gap can be improved dozens of times.

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Fig. 3. The normalized electric field  $|\mathbf{E}| / |\mathbf{E}_0|$  (a) and the corresponding peak wavelength (see Fig. 2 (a)) (b) vary as the gain coefficient of the film increases for five given refractive indices of the film. (c) The comparison of the electric field distribution for films with gain coefficient equal to 0.4 and without gain coefficient, at five different refractive indices of the gain film. (d) The maximum electric field around the NP varies as the refractive index of the substrate increases. Other parameters are as follows: the radius of the NP is equal to 80 nm; the gap distance between the NP and the gain film is 2 nm; the thickness of the gain film is 100 nm; the medium around the NP is air.

Furthermore, the radius of the NP was reduced to 30 nm while keeping the other parameters unchanged; the normalized electric field  $|E|/|E_0|$  spectra was calculated and is shown in Fig. 4. For the present NP, the contribution of the local field from the gain film is similar to that shown in Fig. 2, merely with lower values of field enhancement.

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Fig. 4. (a) Plot of the normalized electric field  $|E|/|E_0|$  spectra for different gain coefficients of the gain film with a 2 nm gap distance between the film and an NP with a 30 nm radius. (b) The electric field distributions at four peak wavelengths, marked with E, F, G, and H in Fig. 4(a), respectively.

Comparatively, we have numerically calculated similar curves to those shown in Fig. 3, but setting the radius of the metallic NP equal to 30 nm. The results are given in Fig. 5. The variation of both normalized fields and resonant wavelengths versus the gain coefficient (see Fig. 5 (a-c)) follows the same trend as shown for previous results in Fig. 3. However, the optimal refractive index of the gain film is close to 2, which is different from the value observed for NPs with 80 nm radius (*i.e.*,  $\sim$ 1.5). The result suggests that we can select an appropriate radius of the metallic NPs to match the practical gain materials in order to achieve a maximal near-field enhancement for the present gap plasmon system. In other words, a relatively low refractive index of the gain substrate can also be used with a strong near-field enhancement, as long as we control the radius of metallic NPs to match the gain medium. In contrast, for any conventional non-gain systems, only a dielectric substrate with a very high refractive index can be used for gap plasmon applications<sup>16</sup>.



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Fig. 5. The normalized electric field  $|E|/|E_0|$  (a) and the corresponding peak wavelength (see Fig. 4 (a)) (b) vary as the gain coefficient of the gain film increases for five given refractive indices of the film. (c) The comparison of the electric field distributions for systems with film gain coefficient equal to 0.6 or to 0 (no gain coefficient), at five different refractive indices of the gain film. (d) The maximum electric field around the NP varies as the refractive index of the film increases. Other parameters are as follows: the radius of the NP is equal to 30 nm; the gap distance between the NP and the gain film is 2 nm; the thickness of the gain film is 100 nm; the medium around the NP is air.

Figure 6 shows the calculated optical spectra of extinction  $(C_{ext})$ , scattering  $(C_{sca})$ , and absorption  $(C_{abs})$  cross sections for three different gain coefficients of the film (n=2) with a 2 nm gap distance between the gain film and the NP. Both the NPs with an 80 nm and a 30 nm particle radius are analyzed using the FDTD. For the gap system with an adjacent dielectric on an individual plasmonic NP, two significant resonant peaks can be observed in all calculated spectrums. Physically, the first peak results from the plasmonic resonance of the metallic NP. However, the gap system could play an important role in the second resonant peak. For both peaks, an obvious feature is that they remain at the same wavelength for different gain coefficiencies (similar conclusion has been given in Ref. 17).

From the absorption spectrum, one can see that a larger gain coefficiency of the gain film can give a higher peak intensity and a narrower line width for both resonant peaks. However, the gain coefficiency has two widely divergent influences on the two scattering peaks, i.e., the first peak reduces, but the second increases as the gain coefficiency increases. This is because the gain film compensates the ohmic loss of metallic NP so that more energy can be localized in the gap. Therefore, the scattering from plasmonic NP itself can be extremely restrained as the gain coefficiency increases. Based on previous image theory<sup>21</sup>, the different permittivity of the dielectric substrate will give rise to a different strength of the interaction between the NP with its image, which might lead to the different variation tendency between the peak wavelength and gain coefficiency shown in Fig.3 (b) and Fig. 5(b).



Fig.6. Calculated optical cross section spectra of absorption (a and d), scattering (b and e), and extinction (c and f) for different gain coefficients of the film with a 2 nm gap distance between the gain film (n=2). Among them, figures (a)-(c) are based on the NP with an 80 nm particle radius, and figures (d)-(f) are for the NP with a 30 nm radius.

# 2. Effect of the gap distance between a silver NP and a thin gain film

The gap distance between the silver NP and the gain film is crucial for determining the interaction between them. We varied the gap from 1 to 4 nm to investigate its effect on the near-field for the case of a silver NP on a dielectric substrate covered with a thin gain film. Figure 7(a) shows the normalized electric field  $|E|/|E_0|$  spectra for different gap distances between the NP with an 80 nm radius and the gain substrate with a 0.15 gain coefficient. The corresponding electric field distributions at four peak wavelengths, marked with I, J, K, and L are shown in Fig. 7(b), respectively. Obviously, the electric field increases as the gap decreases due to stronger interaction of the NP with the film. Moreover, a slight red-shift of the resonant peak can be observed as the gap decreases. The variation is consistent with that based on a conventional non-gain substrate<sup>11</sup>.

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Fig. 7. (a) Plot of the normalized electric field  $|E|/|E_0|$  spectra for different gap distances between the NP with an 80 nm radius and the gain substrate with a 0.15 gain coefficient. (b) The electric field distributions at four peak wavelengths, marked with I, J, K, and L in Fig. 7(a), respectively.

Figure 8 shows how the normalized electric field  $|E|/|E_0|$  varies as the gain coefficient of the gain film increases, for a few different gap values. The electric field distributions with different parameters are also given and are marked with M, N, O, and P in Fig. 8(a), respectively. In the calculation, the radius of the NP is 80 nm. From this figure, one can see that a small gap can lead to an ultrafast enhancement of the local field as the gain coefficient increases. When the gap equals 1 nm, a gain coefficient as little as 0.2 can contribute to a four times field enhancement (see Fig. 8(b)).



(b)

Fig. 8. (a) The normalized electric field  $|E|/|E_0|$  varies as the gain coefficient of the gain film increases for a few different gap values. (b) The electric field distributions with different parameters, marked with M, N, O, and P in Fig. 8(a), respectively. Other parameters are as follows: the radius of the NP is equal to 80 nm; the thickness of the gain film is 100 nm; the medium around the NP is air.

# 3. Effect of the environment refractive index around the silver NP

In the previous discussion, we only considered silver NP surrounded by air. In the section, we will analyze the influence of different surrounding media on the gain assisted gap plasmon system. Figure 9(a) shows the normalized electric field  $|E|/|E_0|$  spectra for different refractive indices of the medium around the NP with a 30 nm radius placed above a gain film with a 0.15 gain coefficient and a 2 nm gap. The corresponding electric field distributions at six peak wavelengths are given in Fig. 9(b), and are marked with Q, R, S, T, U and V in Fig. 9(a), respectively. One can see that both the peak intensity and the peak position vary as the refractive index around the NP changes. When  $n_s = 1.3$  (which varies at different radii of the silver NPs), the system can provide a maximal local electric field. Figure 9(c) shows how the peak wavelength varies as the refractive index of the medium around the NP changes, for a gain coefficient of the substrate equal to 0.3 and 0. From the figure, we can see that the thin gain film does not have any influence on the position of the resonant wavelength at different refractive indices of the surrounding environment.

Figure 10(a) shows how the normalized electric field  $|E|/|E_0|$  varies as the gain coefficient of the gain film increases for a few different refractive indices of the medium around the NP. The comparison of the electric field distribution for gain coefficient equal to 0.3 and 0 for three different refractive indices around the NP is given in Fig. 10(b). When the gain coefficient is low, *e.g.*, 0.25, the maximal field enhancement occurs when  $n_s = 1.3$ , as shown in Fig. 9(a). However, for a larger coefficient of the thin film, the local field will be quickly enhanced as the coefficient increases. Therefore, it is helpful using surrounding media with high refractive index for a high gain system in order to gain super strong field enhancement.



Fig. 9. (a) Plot of the normalized electric field  $|E| / |E_0|$  spectra for different refractive indices of the medium around the NP with 30 nm radius placed above a gain film with 0.15 gain coefficient and 2 nm gap. (b) The electric field distributions at six peak wavelengths, marked with Q, R, S, T, U and V in Fig. 9(a). (c) The peak wavelength varies as the refractive index of the medium around the NP changes, with gain coefficient of the substrate equal to 0.3 and 0.

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Fig. 10. (a) The normalized electric field  $|E|/|E_0|$  varies as the gain coefficient of the gain film increases for a few different refractive indices of the medium around the NP. (b) The comparison of the electric field distribution with 0.3 gain coefficient and without gain coefficient for three different refractive indices around the NP. Other parameters are as follows: the radius of the NP equals 30 nm; the gap distance between the NP and the gain film is 2 nm; the thickness of the gain film is 100 nm.

#### Conclusion

A novel gap plasmon system made by placing a thin gain film on a dielectric substrate has been presented to achieve significant near-field enhancement. Based on an FDTD, we have effectively characterized the effect of the structure and environment parameters on the enhancement intensity. The numerical calculation has shown that a substrate coated with a 100 nm thin gain film can lead to a near-field enhancement of several, or dozens, of times. The local field in the gap based on the present gain substrate can exponentially be amplified as the gain coefficient increases. However, in terms of the difference of the refractive index of the thin gain film, the resonant wavelengths will have a red or blue shift as the gain coefficient increases. For a given radius of the metallic NP, there is an optimal refractive index of the gain substrate, where a maximal field enhancement can be obtained. Moreover, the optimal refractive index of the gain substrate will increase as the radius of the metallic NP decreases. This feature can be exploited to match any practical substrate materials so to achieve an optimal effect of the near-field enhancement simply by using NPs with

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an appropriate size. We also analyzed the influence of the gap value and of the environment refractive index around the NP on the novel gap plasmon system. Numerical results show that the gain substrate can contribute to a more significant field enhancement for a smaller gap value or a larger environment refractive index around the NP.

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