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Simulations of the Spontaneous emission of a Quantum Dot near a Gap Plasmon Waveguide

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In this paper we modeled a quantum dot at near proximity to a gap plasmon waveguide to study the quantum dot-plasmon interactions. Assuming that the waveguide is single mode, this paper is concerned about the dependence of spontaneous emission rate of the quantum dot on waveguide dimensions such as width and height. We compare coupling efficiency of a gap waveguide with symmetric configuration and asymmetric configuration illustrating that symmetric waveguide has a better coupling efficiency to the quantum dot. We also demonstrate that optimally placed quantum dot near a symmetric waveguide with 50 nm x 50 nm cross section can capture 80% of the spontaneous emission into a guided plasmon mode.

I. INTRODUCTION

Spontaneous emission rate of an excited quantum dot (QD) depends on transition strength between two states and Local Density of States (LDOS)[1]. Interaction between emitter and the field can be enhanced by enhancing the number of available density of states into which photons can be emitted. This can be done by altering the density of the electromagnetic modes of the environment. Surface plasmon is an excitation of charge density waves with a tight electromagnetic field confinement[2]. By placing a QD in near vicinity of a subwavelength structure that supports SPs it is possible to manipulate the photonic environment of the QD resulting in increasing the spontaneous emission.

Surface plasmons have potential applications in near field imaging, sensing, solar cells and waveguiding[3, 4]. Plasmonic waveguides are used in plasmonics circuitry to achieve miniaturization[2]. A subwavelength waveguide with metal-dielectric-metal configuration supports highly confined gap modes[5].

There are three possible decay channels for an excited QD when it is placed at proximity to a gap waveguide. First, radiative decay of spontaneous emission into free space[6]. Second, non-radiative decay due to lossy metal[6]. Third is the most important decay channel which is decay into a guided plasmonic mode of the waveguide[7]. Experimentally, Jun *et al.* has shown QD-plasmon coupling for QD films on gap plasmon

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waveguides [8]. In their experiment, they used a pump laser polarized parallel to the gap, and the resultant collected QD emission (centered at 610 nm) was polarized normal to the slit. When they decreased the slit width, the life time of the QD decreased and the QD emission became more polarized normal to the gap. Gap modes are polarized normal to the gap so the fact that they got strongly normally polarized light from the gap is a clear example that QD emission is coupled to the gap mode[8]. Similar result have been shown by Gruber et al. for nanowires[9].

Jun *et al* have theoretically studied the spontaneous emission of a QD near a metal gap plasmon waveguide using Fermi's golden rule and FDTD simulations [10].Their theoretical analysis is based on simplifying assumptions for the LDOS and quantum efficiency of the quantum emitter. [11]. In this paper we extend this model further by using the Green's dyadic function and finite element modeling simulations [12] . To the author's knowledge this work presents the first model of the QD-GPW coupling that takes in to account all decay channels. In this paper we investigate how to increase the coupling efficiency between the QD and GPW by altering gap dimensions, symmetry of the waveguide and distance between emitter and waveguide surface.

This paper focused on studying the effect of waveguide dimensions on the efficiency of coupling QD decay into plasmon mode. The numerical model we used was based on the finite elemental method (FEM) described in Chen *et al*[12]. Gap Plasmon waveguide (GPW) considered has dimensions less than 100 nm in width and height ensuring it supports single mode[13]. Like Jun et al. [8, 10], we found that the polarization of the QD emission significantly affected the QD-plasmon coupling.

II. THEORY

Electric field dyadic Green's function for one guided plasmon was constructed using electric field[12]. In calculating projecting LDOS for the plasmonics mode, it is assumed that the dipole emitter is oriented along *y* axis [refer Figure (i)]. Probability of QD exciting a single plasmon mode is given by spontaneous emission β factor[12].

$$\beta = \frac{\hat{\gamma}_{pl}}{\hat{\gamma}_{tot}} \tag{1}$$

where $\hat{\gamma}_{pl}$ represents the QD emitter decay into plasmon channel and $\hat{\gamma}_{tot}$ is the sum of all decay channels. $\hat{\gamma}_{pl}$ and $\hat{\gamma}_{tot}$ are normalized with respect to the decay rate of a QD in vacuum (γ_0).

Numerical analysis for determining $\hat{\gamma}_{pl}$ is shown explicitly elsewhere[12]. Normalized by the spontaneous emission decay rate in the vacuum, the emission enhancement due to plasmonics excitation is given in Equation (2),

$$\hat{\gamma}_{pl}\left(x_{QD}, y_{QD}\right) = \frac{3\pi c\varepsilon_0 \,|\, E_y\left(x_{QD}, y_{QD}\right)|^2}{k_0^2 \int_A \left(\vec{E}\times\vec{H}^*\right)\hat{z}dA} \qquad (2)$$

where ε_0 is the permittivity of vacuum, k_0 is the wavenumber in vacuum, $\hat{z}$ is the unit vector in z direction and *A* is the transverse plane of the waveguide with the well-defined field components. In Jun et al. they show the importance of positioning the Quantum dots in regions of highly localized field [8]. In particular, they show that by changing the polarization of the input beam they can excite different GPW modes. Depending on the polarization and the GPW mode excited, different QDs will couple to the GPW. In our work we look at QD coupling to the fundamental mode of the GPW. The fundamental mode of the GPW [Figure (ii) and (iii)] has a strong E_y component with maximal field along the waveguide edges. By placing QDs along the waveguide edges we can obtain strong QD-plasmon coupling. We choose the QD's emission axis along the y-direction. For other emission axes it was found that the QD-plasmon coupling was significantly less.

When the QD is placed on top of the waveguide edge, components of the QD emission parallel to y direction can strongly couple to the plasmon modes of the GPW. QD is placed such that QD-plasmon coupling is maximum. According to the field plots [refer Figure (ii) and (iii)] maximum intensity of electric field was at the edge of the waveguide. So the QD was placed 5 nm (unless stated otherwise) above the waveguide edge [refer Figure (i)]. Integration in Eq. (2) takes over the entire *x-y* plane. A 2D finite elemental modeling is used to determine the fundamental plasmon mode fields. Waveguide was considered to be infinitely long so that plasmon reflected from the end of the waveguide will not couple back to the QD.

Numerical method was constructed using dyadic Green's function for guided plasmon mode since field components tend to concentrate more on the metallic edges and decay at the borders for larger domains[12]. Radiation mode field components don't vanish even in larger modeling domain[12]. Therefore it is necessary to build a 3D model to accommodate radiation mode decay in calculating β factor. According to Eq. (1), total decay rate of the QD near metallic waveguide should be calculated in order to calculate β factor. Assuming quantum emitter as a current source at near proximity to the waveguide, total decay rate can be found from the total power dissipation of the current source coupled to the metallic waveguide[12]. It should be normalized with the total power dissipation of the same current source when it is in vacuum. It is important to construct the computational domain properly. We have used scattering boundaries ensuring the absorption of plane waves.

$$\hat{\gamma}_{tot} = \frac{0.5\int_v \mathrm{Re}\left(\vec{J}^* \cdot \vec{E}\right) dV}{0.5\int_v \mathrm{Re}\left(\vec{J}^* \cdot \vec{E}_0\right) dV} \tag{3}$$

where $\vec{J}$ is the current source, $\vec{E}_0$ is the main electric field of the QD in vacuum. V is defined by the volume enclosed by the scattering boundaries. In our model, QD is considered as a 1A line current source. The dipole moment (μ) of a line current source with finite subwavelength size (l) and current I_0 is given by[14],

$$\mu = \frac{jI_0 l}{\omega} \tag{4}$$

Size of the current source should be restricted in order to avoid higher order multipole moments[12]. It is found that variation of total power dissipation depending on the size of the emitter is negligible when the emitter is less than 2 nm[12]. In our model, QD is modeled as a 1 nm line of current source carrying 1 A. Length of the plasmonic waveguide should be long enough to assume that reflected plasmons at the end of the waveguide don't couple back to QD. It is found that a waveguide with a length four times higher than the propagation length is sufficient[15]. Therefore, waveguide was constructed with length 10 µm.

III. RESULTS

Using the 2D finite element modeling simulations of the gap waveguide, normalized spontaneous emission of a QD into a plasmon mode ($\hat{\gamma}_{pl}$) was determined. QD was chosen with an emission wavelength of 633 nm. There were two waveguide configurations considered. Symmetric waveguide consisted of gold with refractive index n_1= 0.197+3.09i surrounding dielectric medium of air with n_2= 1[16]. Asymmetric waveguide structure consisted of gold (n_1) on a glass substrate (n_3=1.5) with upper dielectric medium as air (n_2). QD is positioned 5 nm on top of the gap edge. Gap height (h) or width (w) was varied from 25 nm – 100 nm by keeping one parameter fixed at 50 nm. Schematic of the two waveguide configurations are shown in Figure (i)_

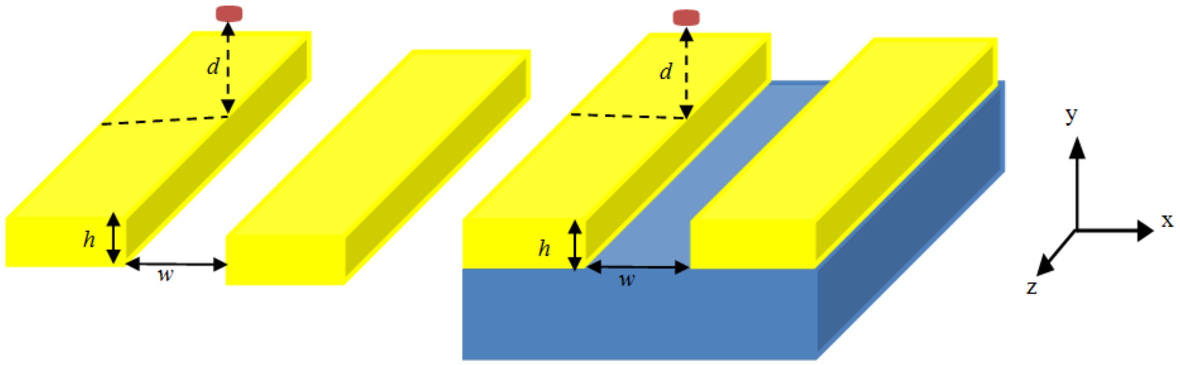

Figure (i).Schematic diagram of a QD sitting on top of the edge of the symmetric GPW and asymmetric waveguide.

According to Eq. (2), $\hat{\gamma}_{pl}$ depends on the strength of the field at the position of the QD. Fundamental mode fields are more concentrated on the edges for larger width and larger heights. So it was chosen to place QD on top of the edge of the waveguide. Field plots of the norm |E| are shown in Figure (iii) and (iv).

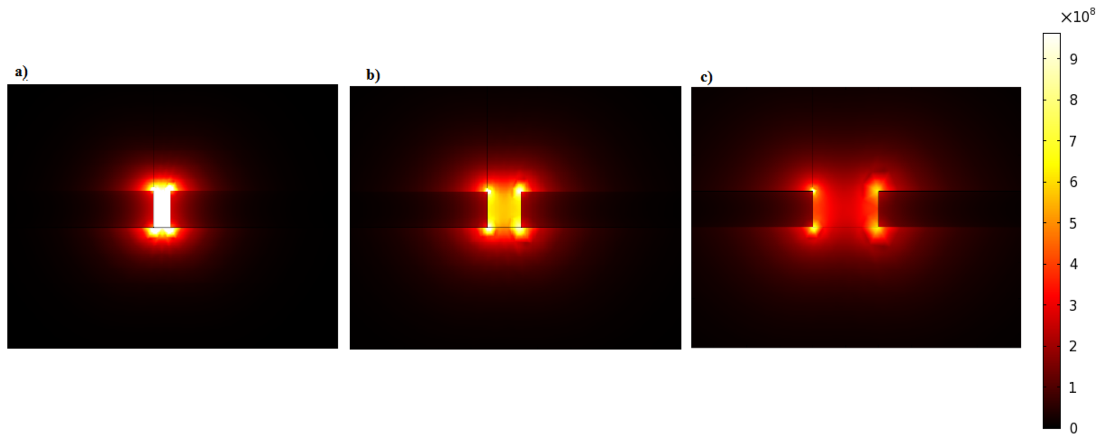

Figure (ii): Field plots of the |E| norm for symmetric GPW for (a) w=25 nm, (b) w= 50 nm and (c) w= 100 nm

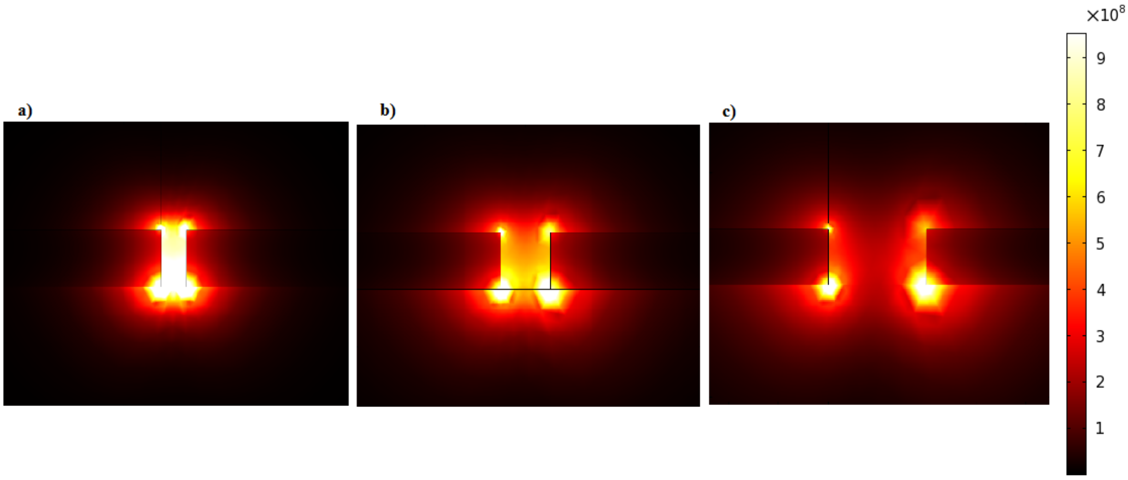

Figure (iii): Field plots of the |E| norm for asymmetric GPW for (a) w= 25 nm, (b) w= 50 nm and (c) w= 100 nm

Field of the fundamental mode is more concentrated on the edge of the symmetric waveguide for larger dimensions [refer Figure (ii)]. Fundamental field is more concentrated on the edges at metal/glass interface for asymmetric waveguides [refer Figure (iii)]. Since the QD is placed 5 nm above the edge of the waveguide, coupling between QD and plasmon mode in symmetric waveguide should be higher than that of asymmetric waveguide.

Wavenumber and propagation length of the fundamental mode for gap widths and heights for symmetric and asymmetric waveguides are shown in Figure (iv) below.

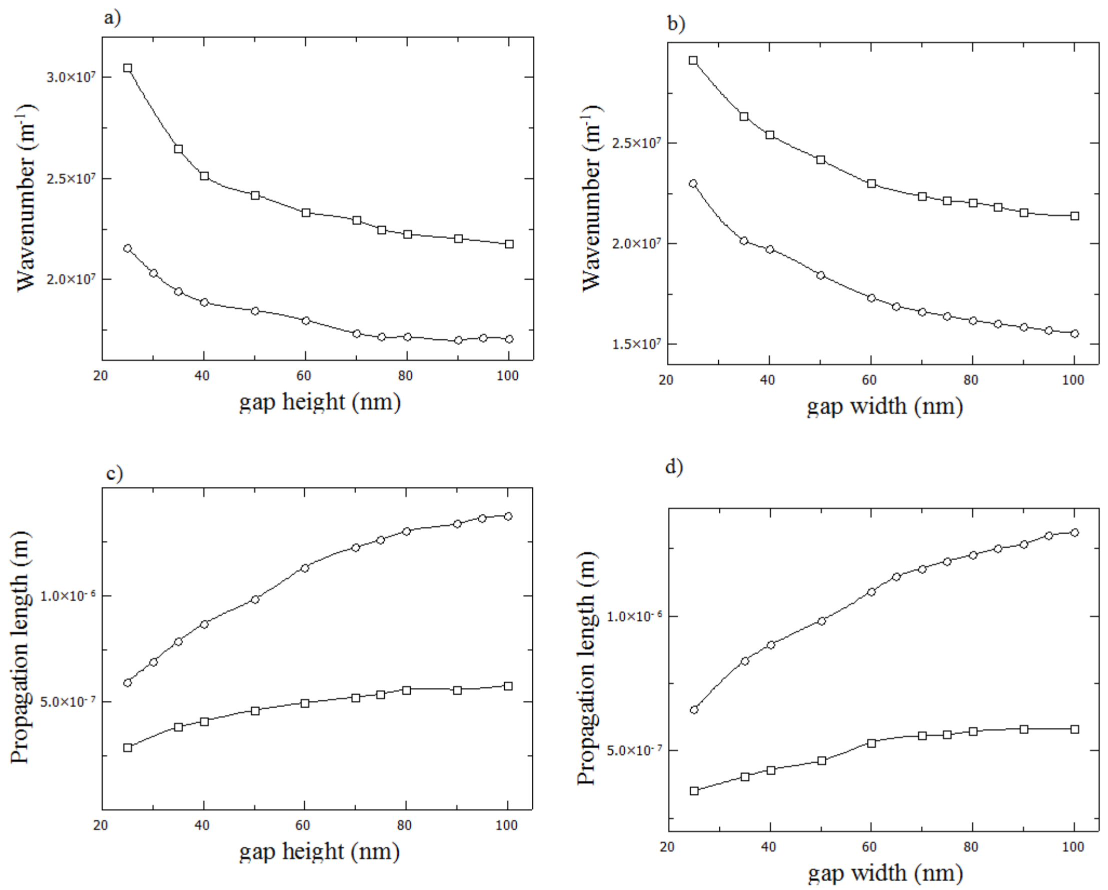

Figure (iv): Wavenumber of the fundamental mode of the GPW for (a) gap height, (b) gap width. Plots of (c) propagation length vs gap height, (d) propagation length vs gap width. Ellipse curve corresponds to symmetric GPW, square curve corresponds to asymmetric GPW.

As the gap dimensions increase, the wavenumber decreased. Larger wavenumber implies smaller group velocity which leads to larger local density of optical states (LDOS). Larger LDOS increases the coupling efficiency to the Quantum Dot in the near vicinity. Such geometric slowing down of the plasmon mode will decrease its propagation length as can be seen in Figure (iv) (c) and (d).

Normalized spontaneous emission of a QD in to a plasmon mode of the GPW is shown in Figure (v). Normalized $\hat{\gamma}_{pl}$ is higher at smaller gap dimensions for both symmetric and asymmetric waveguides. This is in agreement with the prediction made following wavenumber behavior. Coupling to plasmon mode is higher in symmetric structure than the asymmetric structure with same gap dimensions. Enhancement in SE for thinner films is due to both group velocity and mode area reduction. As seen from the field plots of the fundamental mode, field is more localized on the edges of the symmetric waveguide but field is more concentrated on the bottom edges (at metal/glass interface) than air/metal edges on top for asymmetric structures. Therefore, field strength is higher for QD at symmetric waveguide structure leading to higher coupling efficiency all the time.

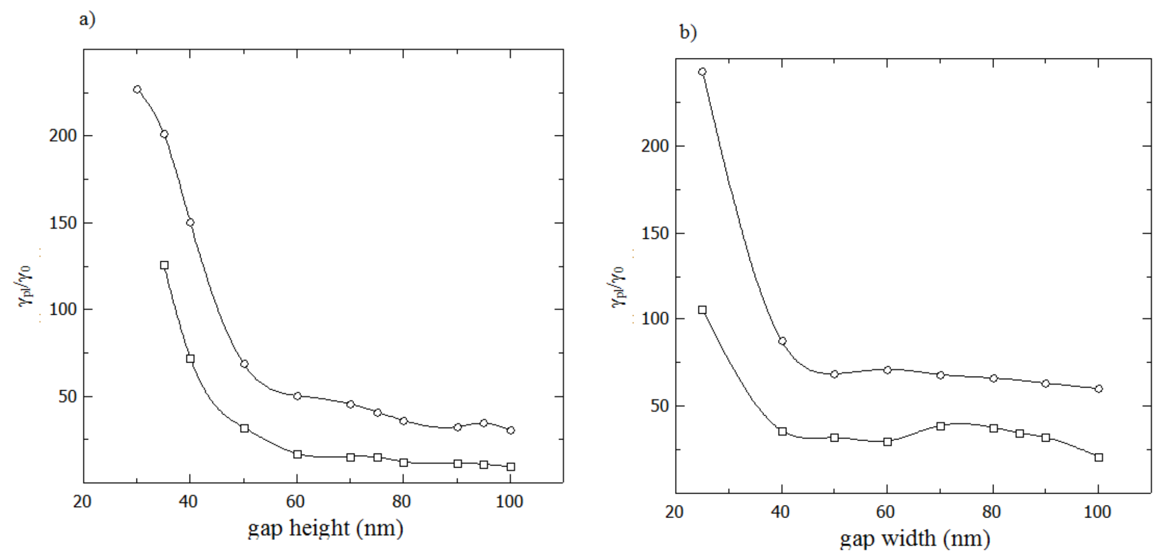

Figure (v): γ_{pl} / γ_0 for (a) gap height and (b) gap width. Ellipse curve corresponds to symmetric GPW and square curve corresponds to asymmetric GPW.

Jun *et al* have shown theoretically that decreasing width at constant height of a planar MDM waveguide with a QD placed in the middle of the slit resulted in decrease of $\hat{\gamma}_{pl}$ which is in agreement with our results [10].

To determine the probability of an excited QD in near vicinity of waveguide decaying into plasmon mode, spontaneous emission β factor must be determined. According to the equation (ii) β can be found using normalized $\hat{\gamma}_{pl}$ and $\hat{\gamma}_{tot}$. Dependence of β values with gap height and width for symmetric and asymmetric GPWs are shown in Figure (vi).

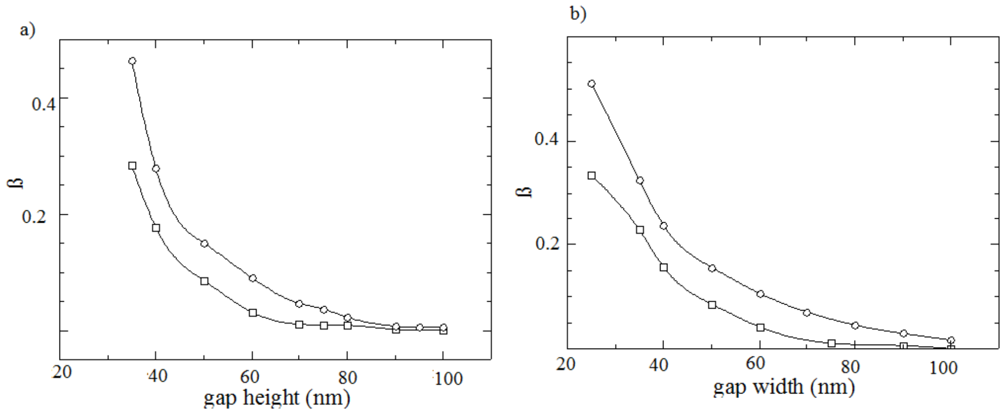

Figure (vi): Variation of spontaneous emission β factor with (a) gap height and (b) gap width. QD is placed 5 nm on top of the waveguide edge. Ellipse curve corresponds to symmetric GPW and square curve corresponds to asymmetric GPW.

Spontaneous emission β factor is higher for smaller waveguide dimensions. As explained in the normalized spontaneous emission in to plasmon mode, coupling between QD and plasmon is higher in the symmetric waveguide structure. With QD placed 5 nm on top of the symmetric waveguide edge, 45% of the excited state of the QD decay into plasmon mode.

It is said that by optimizing the position of the QD with respect to the waveguide surface, β factor can be increased by decreasing the non-radiative decay[17]. β factor is computed by varying the distance of QD from waveguide surface, Figure (vii).

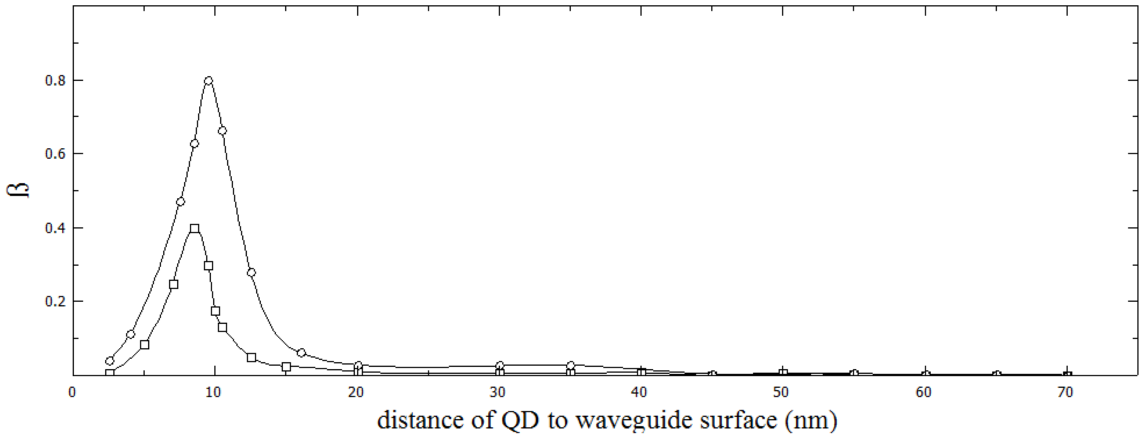

Figure (vii): Dependence of β factor with the distance of QD to waveguide surface. Ellipse curve corresponds to symmetric GPW and square curve corresponds to asymmetric GPW. Gap width and height is kept at 50 nm.

According to Figure (vii), 80% of the excited QD can couple in to the fundamental plasmon mode in a symmetric GPW when the QD is positioned around 10 nm on top of the waveguide edge. For an asymmetric structure this drops down to around 40%. β factor is smaller when the distance between the QD and the waveguide surface is smaller. This is mainly due to the fact that non-radiative decay of excited QD is significant when the QD is very close to the waveguide. When the distance is large, the coupling efficiency becomes low as the field strength is low. Since QD is positioned far away from the evanescent surface plasmon mode tail, β factor is lower at larger QD to waveguide distances.

IV. DISCUSSION:

In conclusion we have studied how to optimize QD-gap plasmon coupling for a single mode GPW. We observe symmetric GPW has a higher efficiency in catching spontaneous emission into guided plasmon mode than asymmetric GPW with same dimensions. With a QD placed 5 nm on top of the waveguide, increasing the width and the height of the GPW tends to decrease the coupling efficiency as fraction of modal power inside the metal is lower. There is a tradeoff between $\hat{\gamma}_{pl}$ and $\hat{\gamma}_{tot}$. Distance between QD and waveguide surface was varied to compromise $\hat{\gamma}_{pl}$ and $\hat{\gamma}_{tot}$. It is shown that for an optimally placed QD, 80% of the total spontaneous emission of the QD can be decay into guided plasmon mode of a symmetric waveguide. For an asymmetric waveguide this coupling efficiency is around 45%. We expect this outcome would be useful for active control of plasmon propagation in plasmonics waveguides.

It should be noted that in the process of fabrication of GPW, it is hard to control the edge sharpness precisely. As shown in Appendix A, increasing the roundness of the waveguide edges will decrease the QD emission decay rate in to plasmon mode. This is due in part to the fact that arbitrary sharp edges create over estimated light intensity and thus the QD-plasmon coupling [5, 18]. To quantitatively describe the behavior of the QD-plasmon coupling the edge sharpness should be compared to experimentally fabricated samples. The models in this paper provide an understanding of the QD-plasmon interaction for various waveguide widths and heights, and edge sharpness should be discussed in future work.

APPENDIX A:

In simulations included, we modelled 2D symmetric GPW with gap dimensions $w = h = 50nm$ and QD distance to waveguide surface at 5 nm with corners of the two semi-infinite metal film regions rounded with radius $r_s \leq h/2$ as shown in Figure viii (a). The mode exhibits a strong concentration at the edges [Figure viii (b)]. When the edge is rounded the strength of the electric field around the edge is slightly weakened. The change in $\hat{\gamma}_{pl}$ value with the curvature of the edge is depicted in Figure viii (c). We can conclude that sharpness has an effect on the QD- gap plasmon coupling. Even though it is hard to control the detailed shape of the sharp edge in the fabrication process, it is better to make the edges as sharp as possible for higher QD emission in to plasmon mode decay rate.

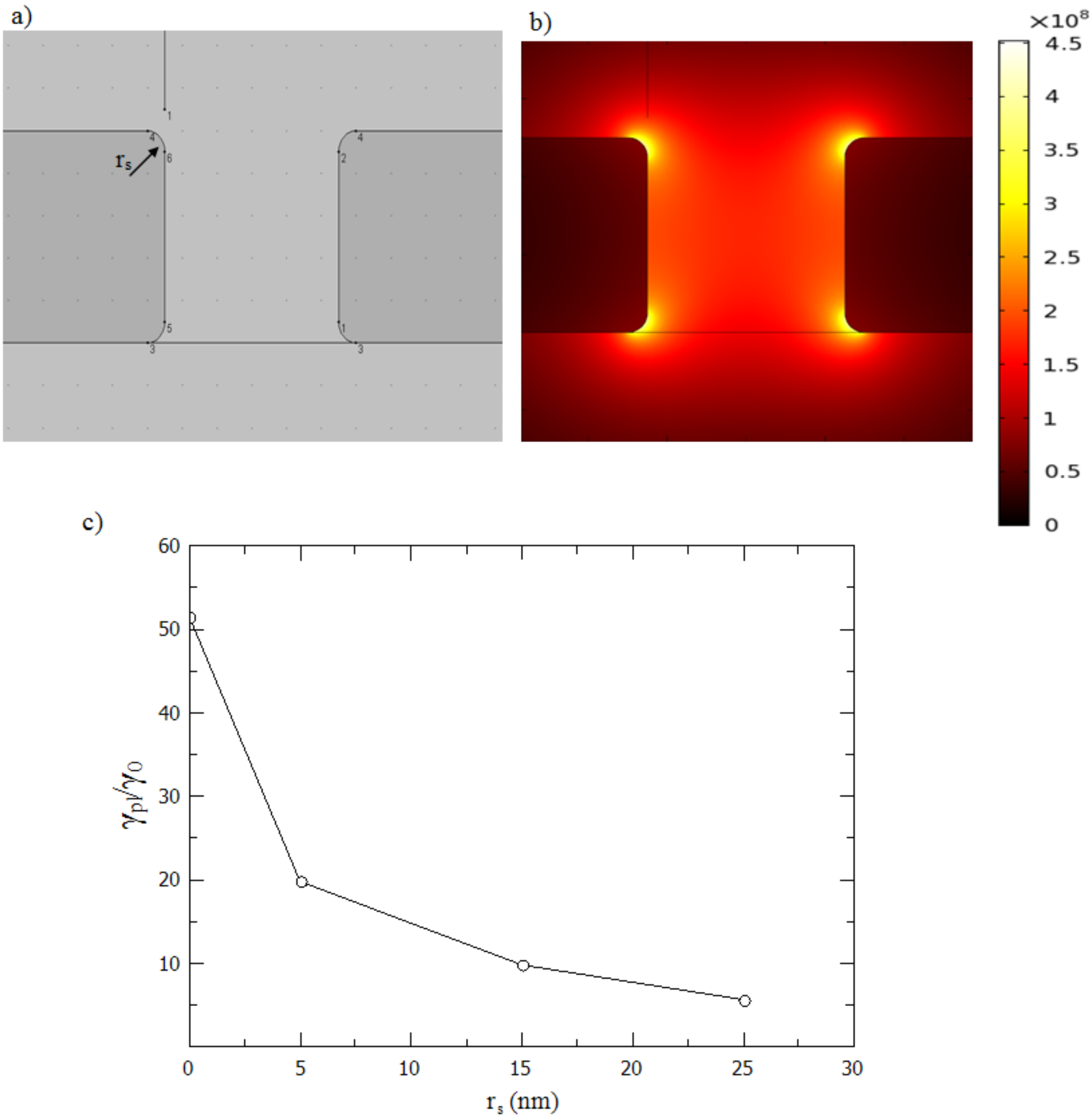

Figure (viii): (a) Schematic of a symmetric GPW with top corners of the two semi-infinite metal films are rounded with radius $0 \le r_s \le h/2$. (b) Field plot of |E| norm of the fundamental mode of the symmetric GPW for $r_s = 15nm$. (c) $\hat{\gamma}_{pl}$ vs r_s for fundamental mode of the GPW with ellipse curve corresponds to symmetric GPW and square curve corresponds to asymmetric GPW.

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References:

1. Purcell, E.M., H.C. Torrey, and R.V. Pound, "Resonance Absorption by Nuclear Magnetic Moments in a Solid", Physical Review, 69(1-2), 37-38 (1946).
2. Atwater, H.A., "The promise of plasmonics", Scientific American, 296(4), 56-63 (2007).
3. Zayats, A.V. and Smolyaninov, II, "Near-field photonics: surface plasmon polaritons and localized surface plasmons", Journal of Optics a-Pure and Applied Optics, 5(4), 16-50 (2003).
4. Atwater, H.A. and A. Polman, "Plasmonics for improved photovoltaic devices", Nature materials, 9(3), 205-213 (2010).
5. Veronis, G. and S.H. Fan, "Modes of subwavelength plasmonic slot waveguides", Journal of Lightwave Technology, 25(9), 2511-2521 (2007).
6. Chance, R., A. Prock, and R. Silbey, "Molecular fluorescence and energy transfer near interfaces", Adv. Chem. Phys, 37(1), 65 (1978).
7. Chang, D.E., A.S. Sørensen, P.R. Hemmer, and M.D. Lukin, "Strong coupling of single emitters to surface plasmons", Physical Review B, 76(3), 035420-1 - 035420-26 (2007).
8. Jun, Y.C., R. Pala, and M.L. Brongersma, "Strong Modification of Quantum Dot Spontaneous Emission via Gap Plasmon Coupling in Metal Nanoslits", Journal of Physical Chemistry C, 114(16), 7269-7273 (2010).
9. Gruber, C., A. Trügler, A. Hohenau, U. Hohenester, and J.R. Krenn, "Spectral modifications and polarization dependent coupling in tailored assemblies of quantum dots and plasmonic nanowires", Nano letters, 13(9), 4257-4262 (2013).
10. Jun, Y.C., R.D. Kekatpure, J.S. White, and M.L. Brongersma, "Nonresonant enhancement of spontaneous emission in metal-dielectric-metal plasmon waveguide structures", Physical Review B, 78(15), (2008).
11. Mertens, H., J.S. Biteen, H.A. Atwater, and A. Polman, "Polarization-selective plasmon-enhanced silicon quantum-dot luminescence", Nano letters, 6(11), 2622-2625 (2006).
12. Chen, Y., T.R. Nielsen, N. Gregersen, P. Lodahl, and J. Mørk, "Finite-element modeling of spontaneous emission of a quantum emitter at nanoscale proximity to plasmonic waveguides", Physical Review B, 81(12), 125431 (2010).
13. Veronis, G. and S.H. Fan, "Plasmonic Slot Waveguides", Plasmonic Nanoguides and Circuits, 159-187 (2009).
14. Jackson, J.D. and R.F. Fox, [Classical electrodynamics], Wiley, New York (1975).
15. Vernon, K.C., D.E. Gomez, T.J. Davis, and N. Tischler, "Simulations of the effect of waveguide cross-section on quantum dot–plasmon coupling", Journal of Applied Physics, 110, 074315 (2011).
16. Palik, E.D., [Handbook of Optical Constants of solids, Volume I, II, and III: Subject Index and Contributor index], Elsevier Science and Tech, Maryland, 286-295 (1985).
17. Chang, D.E., A.S. Sorensen, P.R. Hemmer, and M.D. Lukin, "Strong coupling of single emitters to surface plasmons", Physical Review B, 76(3), 1-34 (2007).
18. Seniutinas, G., L. Rosa, G. Gervinskas, E. Brasselet, and S. Juodkazis, "3D nano-structures for laser nano-manipulation", Beilstein journal of nanotechnology, 4(1), 534-541 (2013).