Simulations of the effect of waveguide cross-section on quantum dot – plasmon coupling

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Quantum dot - plasmon waveguide systems are of interest for the active control of plasmon propagation, and consequently, the development of active nanophotonic devices such as nano-sized optical transistors. This paper is concerned with how varying aspect ratio of the waveguide cross-section affects the quantum dot - plasmon coupling. We compare a stripe waveguide with an equivalent nanowire, illustrating that both waveguides have a similar coupling strength to a nearby quantum dot for small waveguide cross-section, thereby indicating that stripe lithographic waveguides have strong potential use in quantum dot – plasmon waveguide systems. We also demonstrate that changing the aspect ratio of both stripe and wire waveguides can increase the spontaneous emission rate of the quantum dot into the plasmon mode, by up to a factor of five. The results of this paper will contribute to the optimisation of quantum dot - plasmon waveguide systems and help pave the way for the development of active nanophotonics devices.

I. INTRODUCTION

The development of active nano-optical devices using quantum dot - plasmon waveguide systems is a hot topic of nanophotonics research. Quantum dots (QDs) enable active control over the plasmon propagation^{1,2} resulting in possible applications in the development of nano-optical amplifiers, single-plasmon sources and nano-optical transistors.³⁻⁷ The study of QD-plasmon interactions is not only of interest to the development of nano-optical devices, but has potential applications in solar cell technology⁸ and quantum entanglement^{3,4}. The coupling of plasmons and QDs has been experimentally demonstrated for QDs coated on thin metal films^{9,10}, QDs in gap plasmon waveguides⁵ and QD coated metal nanowires^{11,12}. There has been much theoretical work on

how an individual QD couples to plasmons supported by various metal waveguide structures. These works have been based on the fluorescence of dipole emitters.^{13,14} When an excited QD, that acts as a dipole emitter, is in the vicinity of a metal structure, it can either radiatively decay into bulk electromagnetic radiation, nonradiatively decay due to internal losses, heating of the metal and other such mechanisms, or decay into a plasmon mode.^{13,14} Determining the spontaneous emission rate of the excited QD decaying into a plasmon mode is imperative in the design and optimisation of QD-waveguide systems. The ratio of the spontaneous emission rate of the excited QD decaying into a plasmon to the total decay of the excited QD into all possible decay channels is known as the spontaneous emission β factor.^{3,15} A β factor of 1 indicates a 100% probability of the QD decaying into a plasmon mode. The β factor has been determined analytically for the QD-nanowire system^{1,2,6} and has been determined numerically for QDs coupled to square, wedge, gap plasmon and channel plasmon polariton waveguides.^{3,5,15,16}

Martin-Cano et al.³ have shown that the wedge and channel plasmon polariton waveguides can have larger β factors than a nanowire. Channel and wedge plasmon waveguides can be fabricated lithographically^{17,18}, and unlike chemically synthesized nanowires, nano-scale precision of the waveguide position can be obtained. Such precision bodes well for the development of complex nano-optical devices such as plasmonic and quantum circuits.³ Another waveguide which is commonly produced lithographically is the stripe waveguide¹⁹⁻²², which consists of a metal waveguide with either a square or rectangular cross-section, as schematically shown in Figure 1a,b. The β factor of a square waveguide has been determined as a function of distance between the QD and the center of the waveguide.¹⁵ However a direct comparison of this β factor to an equivalent nanowire has not been performed. Furthermore, the effect of the aspect ratio of the waveguide cross-section on the QD-plasmon coupling has not been determined.

This paper aims to optimise the wire and stripe geometries by varying the aspect ratio. The spontaneous emission β factors are determined for metal nanowires and stripe waveguides of varying aspect ratios (Figure 1). We also compare the wavenumber, propagation distance and mode

area of the fundamental plasmon mode of these waveguides, to gain insight into the physics behind the QD-plasmon coupling. The optimal QD-waveguide configuration to attain maximal coupling to the plasmon is discussed.

II. METHOD

The β factor is determined using a numerical model based on the finite element method described by Chen et al.¹⁵ and used by Martin-Cano et al.³ The method is applicable to any type of single mode plasmon waveguide, as long as the fundamental plasmon mode experiences low losses.^{3,15} These applicability conditions come from the approximations used in solving the Green's Dyadic for the plasmon field.¹⁵ Our numerical method differs from that of Chen et al.¹⁵ in that we use scattering boundary conditions rather than the matched boundary conditions. The use of scattering boundary conditions has the advantage of ensuring absorption of plane waves, but may have larger memory requirements than using the matched boundary conditions.

The β factor of a QD-waveguide system determines the probability of an excited QD decaying into a plasmon mode compared to all other decay paths. The β factor is defined as the normalised rate of spontaneous emission of a QD into a plasmon ($\hat{\gamma}_{pl}$) divided by the normalised total rate of spontaneous emission of a QD ($\hat{\gamma}_{tot}$). These values are normalised to the total decay rate of a QD in vacuum (γ_0) i.e. no metallic structure present. The β factor is defined as

$$\beta = \frac{\hat{\gamma}_{pl}}{\hat{\gamma}_{tot}} \quad (1)$$

To determine $\hat{\gamma}_{pl}$ the Green's Dyadic for the electric field for the plasmon mode is defined and solved. This is shown explicitly in Chen et al.¹⁵ and will not be repeated here. The value of $\hat{\gamma}_{pl}$ is

$$\hat{\gamma}_{pl}(x_{QD}, y_{QD}) = \frac{3\pi c \epsilon_0 |E_y(x_{QD}, y_{QD})|^2}{k_0^2 \int_{A_\infty} (\mathbf{E} \times \mathbf{H}^*) \cdot \hat{\mathbf{z}} dA} \quad (2)$$

where the dipole/QD is oriented in y , k_0 is the wavenumber in vacuum and $\hat{z}$ is the unit vector in the z -direction (see Figure 1). The dipole orientation has been chosen for maximum QD-plasmon coupling for the rectangular stripe waveguide, of $s > t$, where s is the structural dimension and t is the thickness of the waveguide cross-section (Figure 1).¹⁹ The main electric field component for the plasmon mode of this rectangular structure is the E_y -component²⁰, and thus maximal QD-plasmon coupling is expected for a QD polarisation oriented in y . In the circular wires and square waveguides the E_x -component and the E_y -component have the same magnitude.

The integral in Eq. (2) is taken over the entire computational domain transverse to the waveguide i.e. the x - y plane. Eq. (2) can be solved analytically if an analytical form for the fields of the plasmon mode is known. Otherwise, the fields can be computed numerically. In this paper we have used a 2D finite element modelling simulation of the waveguide to determine the fields of the fundamental plasmon mode. We have assumed the waveguides to be infinitely long. All the numerical calculations were carried out with the commercial finite element modelling package COMSOL Multiphysics.

According to Eq. (1), in order to calculate β we must also determine $\hat{\gamma}_{tot}$. The total normalised rate of spontaneous emission of a QD ($\hat{\gamma}_{tot}$) is related to the total power dissipation of the QD near the metallic waveguide divided by the total power dissipation of the QD in vacuum.¹⁵

$$\hat{\gamma}_{tot} = \frac{0.5 \int_V \text{Re}(\mathbf{J}^* \cdot \mathbf{E}_y) dV}{0.5 \int_V \text{Re}(\mathbf{J}^* \cdot \mathbf{E}_{0y}) dV} \quad (3)$$

where E_{0y} is the y -component of the electric field in vacuum and $\mathbf{J}$ is the current source. To determine these quantities a full 3D numerical solution is required, to take into account the radiation modes. The QD can be modelled as a line current of current $I_0 = 1$ A and line length 1 nm. By making the assumption of constant current this simplifies Eq. (3) to

$$\hat{\gamma}_{tot} = \frac{\int \text{Re}(E_y) dl}{\int \text{Re}(E_{0y}) dl} \quad (4)$$

assuming the same current source is used in vacuum as used near the metallic waveguide, and dl implies integration over the length of the emitter. The length of the line source was chosen to be sufficiently small that it can be considered a dipole source.^{3,15} Having a small length of the line source ensures that the higher order multipole moments are negligible.¹⁵

To solve Eq. (4) the solution for the electric field at the QD position needs to be determined. The waveguide must be infinitely long, or long enough that a plasmon reflected from the end of the waveguide will not reflect back and interfere with the QD. Otherwise, the 2D solution for the infinite waveguide will be invalid. In our simulations we found a waveguide of length 4 times the propagation length of the fundamental plasmon mode was sufficient, where the propagation length is defined as the distance the plasmon travels before the electric field amplitude drops by a factor of e . The scattering boundaries were placed sufficiently far from the QD-waveguide system so that reflections back to the QD were minimal.

III. RESULTS

A. Normalised spontaneous emission rate of QD into plasmon

The normalised rate of spontaneous emission of a QD into a plasmon ($\hat{\gamma}_{pl}$) is determined for the circular wire, rectangular, square and ellipsoidal plasmon waveguides (Figure 1). The results are compared to those of Chen et al. where possible.¹⁵ To facilitate this, we have chosen a QD emission wavelength of 1000 nm, gold waveguides of permittivity $-50 + 3.85i$ and a surrounding dielectric of permittivity 2. The QD is positioned 5 nm above the center of the waveguide unless stated otherwise.

The wavenumber and mode area of the fundamental mode of a waveguide provide insight into the QD-plasmon coupling. Large wavenumbers indicate a smaller group velocity and a larger

density of states, which should lead to a larger $\hat{\gamma}_{pl}$.⁶ As can be seen from Figure 2a, the wavenumber of the fundamental mode of the circular wire is similar to the wavenumber of the fundamental mode of the square waveguide, only differing for small structural dimensions. This similarity in the wavenumbers indicates a similar level of QD-plasmon coupling for the wire and square waveguide. For the wire the structural dimension is the diameter and for the square waveguide the structural dimension is the side length, Figure 1.

The mode area²¹ which is the area contained in the contour $|E_y|_{\max}/e$, is about 10 times smaller for the fundamental mode of the square waveguide compared to that of the wire waveguide. This should indicate larger fields and stronger coupling, thus a larger $\hat{\gamma}_{pl}$ for square waveguides, Eq. (2). However this is not the case, see Figure 4a. In Figure 4a, $\hat{\gamma}_{pl}$ has been plotted as a function of structural dimension for nanowires and square stripe waveguides. The values for $\hat{\gamma}_{pl}$ agree with the results from Chen et al.¹⁵ where comparisons can be made.

It is clear that $\hat{\gamma}_{pl}$ is similar for a QD coupled to a wire and square waveguide for small structural dimensions. However, at large dimensions the QD is more strongly coupled to the fundamental mode of the wire, despite the fact that the mode area of the square waveguide is smaller. This can be explained by looking at the field distribution of the fundamental mode of the square waveguide.

The fields of the fundamental modes of the circular wire and square waveguide are shown in Figure 5. For square waveguides of large structural dimensions, the field is strongly localised around the edges, not the center of the waveguide, as shown in Figure 5d. Thus having a smaller mode area than the nanowire does not necessarily mean a larger $\hat{\gamma}_{pl}$, as the $\hat{\gamma}_{pl}$ is dependent on the field strength at the position of the QD, see Eq. (2). The fact that the QD positioned above the center of the waveguide experiences low fields for waveguides of large structural dimensions, explains why the $\hat{\gamma}_{pl}$ for the square waveguide is less than the $\hat{\gamma}_{pl}$ for the wire at large structural dimensions, Figure 4a. Keeping the QD 5 nm above the waveguide and moving its position off-center to above the stripe edge, it is clear from Figure 5b,d that this will strongly affect the field strength at the QD

position for large structural dimensions. This is reflected in the results for the $\hat{\gamma}_{pl}$ shown in Figure 4a. At large structural dimensions, having the QD off-center significantly increases $\hat{\gamma}_{pl}$. For small structural dimensions, the QD will experience slightly stronger fields if the QD is positioned above the center of the waveguide (Figure 5b), explaining why the $\hat{\gamma}_{pl}$ is slightly higher for QDs positioned above the center of the waveguide for small structural dimensions, Figure 4a .

We have shown that a square waveguide will give a similar $\hat{\gamma}_{pl}$ to that of a nanowire, for small structural dimensions. This is due to the fact that both square and wire waveguides have a similar wavenumber and for small structural dimensions, a similar strength of the electric field at the QD position. The square waveguide has two distinct advantages over the wire, the ability to position the square waveguide with nano-scale accuracy using lithography and longer propagation lengths of the plasmon mode, Figure 2b. To maximise $\hat{\gamma}_{pl}$ the QD should be positioned off-center, above the square edge, for squares of large structural dimension. To further maximise $\hat{\gamma}_{pl}$ the wavenumber and localisation of the fundamental mode should be increased, as this will result in a stronger electric field at the QD position. To increase the wavenumber and localisation of the fundamental mode, the waveguide can be flattened or its thickness decreased. We have numerically explored the effect of waveguide dimensions by keeping the structural dimension s at 50 nm, varying the thickness t and thus changing the aspect ratio of the wire and stripe waveguide, making them ellipsoidal and rectangular respectively (Figure 1). As the thickness of the waveguides decrease, the wavenumber increases, see Figure 3a.

This results in an increase in $\hat{\gamma}_{pl}$ with decreasing thickness for both types of waveguide, Figure 4b. In fact flattening a nanowire waveguide so that it has a t of 10 nm and s of 50 nm results in doubling $\hat{\gamma}_{pl}$. Changing the aspect ratio of stripe waveguides also results in a significant increase in $\hat{\gamma}_{pl}$, around 3-5 times (depending on QD position) for a rectangular waveguide of $s = 50$ nm and $t = 10$ nm compared to a square of 50 nm by 50 nm. For small thickness, the ellipsoidal and rectangular waveguide have a similar $\hat{\gamma}_{pl}$ if the QD is positioned above the stripe edge. For an s of 50 nm it is beneficial to place the QD off-center, Figure 5c,d.

We have demonstrated that a square waveguide which can be fabricated lithographically, can have similar $\hat{\gamma}_{pl}$ to that of a chemically synthesised nanowire. We have also demonstrated the benefits of changing the aspect ratio of the waveguides and moving the QD position. Moving the QD position off-center to above the stripe edge, is beneficial for large structural dimension square waveguides and thin rectangular waveguides.

B. Spontaneous emission β factors

To truly determine if the excited state of the QD decays preferentially into a plasmon mode, the β factor must be determined. The larger the β factor the more likely a QD will decay into a plasmon than into any other decay mechanism. To determine β , the values of $\hat{\gamma}_{pl}$ from the previous section can be used and the values of $\hat{\gamma}_{tot}$ must be computed, see Eq. (1). The value of $\hat{\gamma}_{tot}$ can be computed for the waveguides using Eq. (4). Using the values of $\hat{\gamma}_{tot}$ and $\hat{\gamma}_{pl}$ one computes β , Figure 6.

The β factor is around 1 for small dimensions of the wire and square waveguide, Figure 6a. Both the wire and square waveguide can exhibit strong coupling between the fundamental plasmon and the QD. What is interesting to note, is that the β for the square waveguide is significantly lower for small structural dimensions if the QD is positioned off-center, above the stripe edge. The total decay rate of the QD near the stripe edge is stronger than if the QD is positioned above the center of the waveguide. There may be some benefit to placing the QD above the stripe edge for large structural dimensions, but all waveguides at these dimensions have small β .

Flattening the wires and squares has a significant impact on the β , Figure 6b. Flattening the square and making it $s = 50$ nm and $t = 10$ nm results in doubling β . For an s of 50 nm, the wire has almost double the β of the stripe waveguide regardless of stripe thickness. Furthermore, the QD position for an s of 50 nm does not affect the value of β for the stripe waveguide. Even though the $\hat{\gamma}_{pl}$ of a QD above the edge of the stripe is almost double that of the $\hat{\gamma}_{pl}$ of a QD above the center, the $\hat{\gamma}_{tot}$ is also larger for a QD above the edge. The benefits achieved by increasing $\hat{\gamma}_{pl}$ by positioning

the QD above the stripe edge, are decreased due to the greater total decay rate of the QD above the stripe edge.

As a final note, the dependence of β on distance of the QD from the waveguide surface was briefly investigated for a wire, stripe and rectangular waveguide. It is clear from Figure 7 that the β for all three QD-waveguide systems exhibit a similar dependence on distance of the QD from the waveguide surface, with a distance of 20 nm being optimal. In agreement with previous discussions, the β is quite similar for the square and wire waveguide, except at close QD-waveguide distances of 5 nm, where the wire has a significantly larger β than the square. By flattening the square and making it rectangular in nature, the β factor is significantly improved, with β values larger than the nanowire. Flattening the waveguides significantly improves β .

IV. DISCUSSION

The spontaneous emission β factors have been determined for a QD coupled to waveguides with four types of cross-sections: circular, ellipsoidal, square and rectangular. It has been shown that the β factor for a square waveguide is comparable to that of a nanowire, thus stripe lithographic waveguides can be used for creating nonlinear nano-optical devices. For small waveguide dimensions, the square and wire waveguide have similar β factors, and only at structural dimensions greater than 50 nm does the nanowire have a significantly larger β factor than an equivalent square waveguide.

Positioning the QD above the stripe edge resulted in larger values of $\hat{\gamma}_{pl}$ but also larger values of $\hat{\gamma}_{tot}$. There is a trade-off between $\hat{\gamma}_{pl}$ and $\hat{\gamma}_{tot}$ and only in cases of large structural dimensions was there a benefit to placing the QD above the stripe edge. A QD above a stripe edge has a stronger coupling to the plasmon but it also has a larger total decay rate. This compromise between $\hat{\gamma}_{pl}$ and $\hat{\gamma}_{tot}$ is important when designing QD-plasmon waveguide systems. The compromise between $\hat{\gamma}_{pl}$ and $\hat{\gamma}_{tot}$ has been seen previously for varying distance of the QD from the

waveguide surface, but we report that this compromise also occurs for horizontal movement of the QD.

The thickness or aspect ratio of the waveguide also plays a critical part in the β factor of the QD-waveguide system. Decreasing the thickness of the wire and square waveguides results in much larger β factors, by up to a factor of 5, which can be attributed to decreasing mode area and increasing wavenumber of the fundamental plasmon mode with decreasing waveguide thickness.

We have demonstrated that stripe waveguides and wires both sustain high β factors for small waveguide cross-section and that altering the aspect ratio of the stripe and wire waveguides can greatly increase the β factor. These outcomes should have a significant impact on the design of future QD-waveguide systems for active nanophotonics applications.

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Figure 1: (a) Square, (b) rectangular, (c) wire and (d) ellipsoidal waveguide. s is the structural dimension which is the width/diameter along the x axis and t is the thickness which is the width/diameter along the y axis. The QD (star) is positioned half way along the length of the waveguide, 5 nm above the waveguide surface, in the center.

Figure 2: (a) Wavenumber of fundamental plasmon mode supported by a square (square) and wire (circle) waveguide. The structural dimension is for the square width and wire diameter respectively (b) Plot of the propagation distance of the fundamental plasmon mode supported by the square (square) and wire (circle) waveguide.

Figure 3: (a) Wavenumber of fundamental plasmon mode supported by a rectangular (square) and ellipsoidal (circle) waveguide. The stripe and nanowire have a width/diameter of 50 nm, with varying thickness (b) Plot of the propagation distance of the fundamental plasmon mode supported by the rectangular (square) and ellipsoidal (circle) waveguide.

Figure 4: (a) $\gamma_{\text{plas}}/\gamma_0$ for a wire-QD system (circle), a square-QD system (square) and a square-QD system when the QD is positioned 5 nm above the waveguide corner (diamond) (b) $\gamma_{\text{plas}}/\gamma_0$ versus t for $s = 50$ nm and an ellipsoidal waveguide (circle), a rectangular waveguide (square) and a rectangular waveguide when the QD is positioned 5 nm above the waveguide edge (diamond).

Figure 5: Contour plots of the electric field distribution of the E_y component for (a) a circular wire of $s = 10$ nm, (b) a square waveguide of $s = 10$ nm, (c) a rectangular waveguide of $t = 10$ nm and $s = 50$ nm and (d) a square waveguide of $s = 50$ nm. Position of the QDs indicated by stars.

Figure 6: β for a QD coupled (a) 5 nm above the center of a wire (circle), a square waveguide (square) and 5 nm above the edge of a square waveguide (diamond), and (b) 5 nm above the center of an ellipsoidal waveguide of $s = 50$ nm and variable thickness (circle), a rectangular waveguide of $s = 50$ nm and variable thickness (square) and 5 nm above the edge of a rectangular waveguide of $s = 50$ nm and variable thickness (diamond).

Figure 7: β for a QD coupled above the center of a wire (circle), a square waveguide (square) and a rectangular waveguide (plus), for varying distance of the QD to the waveguide surface. $s = 50$ nm and thickness is 25 nm for the rectangular waveguide.

Figure 1: (a) Square, (b) rectangular, (c) wire and (d) ellipsoidal waveguide. s is the structural dimension which is the width/diameter along the x axis and t is the thickness which is the width/diameter along the y axis. The QD (star) is positioned half way along the length of the waveguide, 5 nm above the waveguide surface, in the center.

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Figure 4: (a) $\gamma_{\text{plas}} / \gamma_0$ for a wire-QD system (circle), a square-QD system (square) and a square-QD system when the QD is positioned 5 nm above the waveguide corner (diamond) (b) $\gamma_{\text{plas}} / \gamma_0$ versus t for $s = 50$ nm and an ellipsoidal waveguide (circle), a rectangular waveguide (square) and a rectangular waveguide when the QD is positioned 5 nm above the waveguide edge (diamond).

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Figure 6: β for a QD coupled (a) 5 nm above the center of a wire (circle), a square waveguide (square) and 5 nm above the edge of a square waveguide (diamond), and (b) 5 nm above the center of an ellipsoidal waveguide of $s = 50$ nm and variable thickness (circle), a rectangular waveguide of $s = 50$ nm and variable thickness (square) and 5 nm above the edge of a rectangular waveguide of $s = 50$ nm and variable thickness (diamond).

Figure 7: β for a QD coupled above the center of a wire (circle), a square waveguide (square) and a rectangular waveguide (plus), for varying distance of the QD to the waveguide surface. $s = 50$ nm and thickness is 25 nm for the rectangular waveguide.