Design, Fabrication and 3D Volumetric Characterization Criteria for Plasmonic Bullseye Structures

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Abstract: In this report, certain claims regarding the design criteria and characterizations of plasmonic bullseye (BE) structures are investigated. Does the matching of the grating’s periodicity to the wavelength of surface waves result in a strong collimated beam along the optical axis of the device? What are the requirements for such devices if they are set to enhance the radiative decay rate of a quantum emitter? These questions are answered by proposing and modelling a new bullseye structure, optimized based on maximization of the far-field intensity along the optical axis. Dimensions and the performance of the proposed BE are then compared to that reported by another author. The other question to be answered is the characterizations techniques. What is the best approach in characterizing such devices? How many focal points are there along the optical axis? What are the strengths and depths of focus at each point? How collimated is the output? Techniques proposed by other authors are debated and a technique based on 3D confocal microscopy is proposed and applied experimentally to the alternative proposed device.

1. Introduction
Bullseye structures are relatively simple, i.e. a single subwavelength cavity (or aperture/hole) surrounded by concentric circular gratings. Centre of a BE structure may accommodate annular rings [1], a circular hole [2] or a flat disk with no opening [3]. The cavity (or the aperture) in a BE structure may host a quantum emitter, such as a nano-diamond with NV-vacancy, to enhance it radiative decay rate (RDR) and improve the collection efficiency [4-7]. Their simple planar design also makes them ideal for integration with the exit surface of other sources such as a quantum cascade laser [8] or fibre optics [3] to collimate the output light. The role of the corrugation may vary depending on the application. I tend to agree with the definition provided by Genet and Ebessen [9] and I quote: “If the output surface surrounding the aperture is also corrugated, a surprisingly narrow beam can be generated, having a divergence of less than a few degrees, which is far smaller than that of the single apertures discussed earlier. This is because the light emerging from the hole couples to the periodic structure of the exit surface and to the modes existing in the grooves, which in turn scatter the surface waves into freely propagating light. This then interferes with the light that has travelled directly through the hole generating the focused beam [9].”

The influence of the BE geometry on the transmission efficiency has previously been investigated for a given wavelength [10], where the authors showed that the optimum groove width for maximum transmission efficiency is approximately $0.5\times\lambda_{SPP}$, where $\lambda_{SPP}$ is the
wavelength of surface plasmon polaritons (SPP). The number of corrugations where the transmission efficiency approaches saturation was found to be 6–10. Furthermore, it has been claimed that the far-field radiation pattern of a BE structure depends primarily on the corrugations surrounding the hole rather than the aperture’s geometry. It was confirmed that the distance between the aperture and its nearest groove, i.e. the radius of the first corrugation, influences the coupling between the LSPs inside the cavity and the SPPs in the corrugations, thus affecting the transmission efficiency [11].

The directionality of the emission from a NV- inside a cylindrical nano-diamond cavity surrounded by concentric circular corrugations engraved in a silver film was investigated in [6]. Such a design operates in the reflection mode, where the incident and the collection surfaces are the same. Although this design uses concentric circular corrugations to achieve collimated reflected beam, the enhancement to the RDR was reported to be poor, presumably due to the utilization of a non-resonant cavity. Yet, a more fundamental flaw in the design is the operation in reflection mode. In order to excite the single photon emitter inside the cavity, it must be positioned at the focal point of a confocal objective lens. Point-illumination/point-collection using the same objective lens is an intrinsic feature of the confocal microscopy. Moving the objective lens away from the device into the far-field zone, (hence moving the emitter away from the focal point), reduces the incident power over the dipole drastically. Although the excitation of the dipole in the cavity may be achieved by a side-illumination technique via a separate objective lens, such configurations are more complex. In this design the choice of diamond superstrate is well justified as it prevents the emission of the nano-diamond cavity (at the centre of the BE) from total internal reflections. The inclusion of the diamond superstrate allows the emission to escape the nano-diamond and since diamond crystal has low absorption (due to its large band gap [12]) most of the emitted power may be collected from the superstrate. Other authors have also based their design on collecting the dipole’s emission from optically dense materials [2,12-15]. Recent developments in nano-diamond fabrications have led to designs such as a thin diamond film (supported on a glass substrate) with BE grating etched on the surface surrounding a nitrogen vacancy to collimate its emission [5]. This device was reported to have a high collection efficiency when characterised with a confocal microscope in the reflection mode. The use of a homogenous diamond film with NV-near its surface was to avoid complications (such as total internal reflections) associated with designs where a nano-diamond is integrated with an external structure. The far-field radiation pattern was obtained using back-focal-plane imaging techniques (using Bertrand lens) that eliminated the need to move the confocal objective away from the emitter, hence maintaining a constant power over the emitter at all time. Is this practical in real-life applications? One must consider both the in-situ and in-laboratory conditions, where in-laboratory refers to the stage where a device is under investigation and in-situ referring to a stage where the device is integrated with its intended circuitry. When characterizing devices such as these (hence in-laboratory conditions), one needs to map its near- and far-field profiles. When using a Bertrand lens one has no idea on how far is the far-field. The ultimate goal behind subwavelength plasmonic devices here is miniaturization. Miniaturization of the whole confocal setup with Bertrand lens on the optical path so that the receiving antenna can benefit from the collimated beam in k-space is a very complex problem. It is only intuitive for such devices to be designed based on transmission geometry where in-situ and in-laboratory conditions are not too different. Device reported in [5] also shows that the field intensity in the air is lower in comparison to those inside the diamond film and the substrate. This may be due
to the complex interaction of NV- emission with the patterned diamond film that possesses photonic crystal characteristics (i.e. band gap, wave-guiding ... etc), and/or direct interaction of NV- with air/diamond and glass/diamond boundaries (which includes the total internal reflection and the quenching of the dipole’s radiation). It is obvious that the larger portion of the emitted power is unutilized, only trapped inside the diamond. Integrating nano-diamonds with a properly designed plasmonic structure in transmission mode may offer a solution to this problem.

2. Analysis and Design
Electromagnetic activities within the BE structure are complex. Dimensions of the cavity/aperture and the thickness of the film also have bearings on the strength of the electric field over the surface of the bullseye. The surrounding dielectric material, the distance of the first groove from the cavity/aperture, the depth, periodicity and the width of the corrugations all play a role in strengthening/suppressing scattered power and shaping the radiation pattern. In this section, a qualitative description of light-matter interactions in a plasmonic BE is provided and a BE design by another author is examined. Reliance on analytical solutions in modelling such a complex interaction in a seemingly simple structure is not fruitful. With the help of numerical solutions, however, one can optimize the design of the BE for a given wavelength that enhances the radiative decay rate (RDR) as well as producing highly directional light along its optical axis. Finally, a novel technique in harnessing the power emitted by a NV- in a diamond substrate is proposed and the performance of the BE is evaluated in such a setting. To date, the design of plasmonic BE structures has been primarily focused on utilizing a hole aperture and matching the period of its surrounding corrugations to the wavelength of the surface plasmon polaritons. Surface waves on a flat metallic film, however, are the results of evanescent electric fields normal to the surface boundaries. SPPs also carry electric fields that are parallel to the surface above and beneath a metal/dielectric interface. Components of the electric field that dominate the far-field along the optical axis, have been largely ignored in most BE designs so far. Here, it is first demonstrated that the normal and the parallel to the surface components of the electric field of light transmitted through a subwavelength resonant aperture exhibit higher amplitudes in comparison to those of a circular hole. By combining the resonant aperture with a BE structure and optimizing the corrugations to maximize the parallel components scattered into the far-field, it is shown that a highly directional beam reaching tens of microns away from the device is attainable. Furthermore, it is demonstrated that for a sufficiently thin metallic film, parallel components of the electric field from the substrate side, couple to the surface plasmon inside the corrugations resulting in further enhancements of the far-field intensity.

An investigation of the interaction between an aperture and a nearby groove via surface waves has been undertaken [16-18] and the coupling of waveguide modes to the fields inside the groove through a thin layer of metal has been demonstrated [19]. Here, a qualitative description of such interactions in 2D in terms of the \( x \) and the \( z \) components of the electric field are provided. Considering an \( x \)-polarized TM wave travelling in the \(+z\) direction and normally incident on an optically thick “flat” metallic film (laid along the \( x \)-axis) perforated with a single subwavelength resonant aperture. The role of the aperture is to utilize some of the incident power to generate the electric field, \( \vec{E}_a = E_{ax} \hat{n}_x + E_{az} \hat{n}_z \), which travels along the film’s surface.
The interaction between $E_\omega = E_{\omega x} \hat{x} + E_{\omega z} \hat{z}$ and the surface charge densities is limited only to surfaces that satisfy the condition $|\hat{n} \cdot E_\omega| > 0$, where $\hat{n}$ is the vector normal to the surface.

Corrugations surrounding an aperture, (Figure 1(a)), may be considered as a sequence of alternating vertical and horizontal surfaces. Upon arrival at a groove, $E_{\omega x}$ interacts with the surface charge density, $\sigma_x$, that leads to the accumulation of charges in the corner of the groove, Figure 1(b). This gives rise to the surface charge density, $\sigma_x$, on the vertical surfaces and, subsequently, to an $x$-directed electric field, $E_{xc}$, inside the grooves. The interaction between the already established $E_{xc}$ and the newly arriving $E_{\omega x}$ (depending on their relative phase differences), either strengthens or weakens the surface charge density $\sigma_x$. The formation of a point source with a dipole moment $\vec{p}_t$ on the upper corner of a groove positioned at $(x_i, z_0)$, is the result of the surface charge oscillations $\sigma_x(x_i, z_0)$ and $\sigma_x(x_l, z)$, (Figure 1(b)). Properly spaced corrugations provide a mechanism to intercept the $E_{\omega x}$ with a correct phase. Spacing of the corrugations also controls the shape of the scattered light by the BE. Scattered field from the gratings, therefore, must make a constructive interference with that of the aperture along the optical axis.

![Figure 1: Schematics of (a) components of a bullseye structure and (b) formation of a point source with a dipole moment $\vec{p}_t$ on the upper corner of a groove at coordinates $(x_i, z_0)$, due to the surface charge oscillations $\sigma_x(x_i, z_0)$ and $\sigma_x(x_l, z)$, (c) Electric filed components launched by a resonant cross-shaped aperture at $\lambda_0 = 700$ nm.](image)

Contributions by the $E_{\omega x}$ to these charge oscillations at $(x_i, z_0)$, however, are $180^\circ$ out of phase with those at $(x_l, z_0)$. This is simply due to the $E_{\omega x}$ being an odd function of $x$, (compare the three components of the electric field along the film surface in Figure 1(c)). Consequently, any contribution by $E_{\omega x}$ to $\vec{p}_t$ and $\vec{p}_t$ (hence to their scattered fields) leads to a destructive interference along the optical axis. This is an adverse effect given that the $E_{\omega x}$ is the strongest of the three components. In an optically thick BE structure, the corrugations should intercept the power carried by $E_{xc}$ that propagates along the surface to maximize $E_{xc}$ that propagates in the $z$-direction. For a BE structure composed of $N$ concentric corrugations, there are $2 \times N$ vertical surfaces that can potentially be utilized to maximize the $x$-component of the transmitted/scattered electric field. When drawing an analogy between the charge oscillations on the corner of a groove and a point dipole, a BE structure may be looked at as an antenna
array that is composed of a number electric point dipoles, \( p_i \), positioned at \((x_i, z_0)\), where \( i \) is an integer. In such an antenna arrays, when \( x_i - x_{i+1} < \lambda_0 \), increasing the number of hotspots, strengthens the intensity in the central lobe [20].

Garcia-Vidal et al. [21], proposed an analytical formalism to calculate the far-field radiation pattern of a slit flanked by surface corrugations using the Huyghen’s principle [22]:

\[
H(r) = \frac{1}{\mu_0 c} \sum_{i=1}^{N} E_{ai} G(r, r_i)
\]

Here, \( H(r) \), is the total \( y \) component of the magnetic field calculated at position \( r \), from the superposition of electric fields at the indentations located at \( r_i \). Note that \( E_{ai} \) is related to the \( x \)-component of the electric field at the \( i^{th} \) indentation and \( G(r, r_i) \) is the scalar Green’s function, see equation (2) in [22] for full definition. Equation (1), however, does not reveal much about the interaction between the \( E_{ai} \) and its surroundings. The angular spectrum of a single dipole positioned near a planar interface is formulated by Novotny and Hecht [23]. The dipole moment of a point source may be described as \( \vec{p}_i = e^{-i\omega t} p_{10} (a\hat{n}_x + b\hat{n}_z) \), where \( \phi_s, p_{10} \) and \((an_x + bn_z)\) describe the phase, magnitude and the orientation of the \( i^{th} \) dipole moment respectively. The total transmitted/scattered electric field measured at an arbitrary location, \( r = \sqrt{x^2 + z^2} \), in the half-space above the BE surface may then be described by the superposition of all the electric fields due to each dipole:

\[
\vec{E}(r)_{tot} = \sum_{i=1}^{N} \omega^2 \mu_0 \mu_1 \left[ \vec{G}_0(r, r_i) + \vec{G}_{ref}(r, r_i) \right] \vec{p}_i
\]

where \( r_i = \sqrt{x_i^2 + z_0^2} \) denotes the location of the \( i^{th} \) point source, \( \mu_0 \) is the permeability of free space and \( \mu_1 \) is the relative permeability of the medium within which the dipole’s radiation propagates. This is an extension to the equation (10.17) provided by Novotny and Hecht [23]. The dyadic Green’s functions \( \vec{G}_0(r, r_i) \) and \( \vec{G}_{ref}(r, r_i) \) map the direct dipole radiation and its Fresnel reflection by the film into the half-space above the BE structure. For a full description of the dyadic Green’s functions \( \vec{G}_0(r, r_i) \) and \( \vec{G}_{ref}(r, r_i) \) see equations (10.6-10.21) and D1-D5 [23]. A consequence of equation (2) is the reflection from the planar interface contributing to the total radiated field. Considering a horizontally oriented dipole moment above a plane surface, it was shown that only 50% of the power is radiated into the upper half-space directly. The other 50% is incident on the surface below which is partly reflected back into the upper half-space. The remaining portion of the reflected power interacts with the dipole, see equation 10.48 [23]. Furthermore, Equation 10.32 [23] shows that only the parallel components of a dipole moment positioned near a planar surface, make contribution to the far-field along the \( z \)-direction. For the reflection term not to vanish, however, the dipole must be positioned at a distance larger than \( \sim 10 \) nm above the metallic surface [13,23]. Strong surface charge densities formed on the upper corners of a groove satisfy this condition with respect to the bottom surface, \( z_{ref} = z_0 - h \). To increase the reflective surface area, the width of periodic corrugations must occupy a larger portion of the BE structure as whole. To determine the optimum
dimensions of a BE structure under such complex light-matter interactions, the use of equation (1) is not advisable since a previous knowledge about the amplitude and the phase of electric fields at each indentation is required. The same applies to equation (2) with respect to the magnitude and the phase of each dipole moment, not to mention the complexity involved regarding the interaction between dipoles and the film. The situation becomes even more convoluted when dealing with dimensions that are comparable with skin depths in metals. Therefore, the best approach here is to rely on numerical solutions, which have proven to be reasonably accurate all along this project.

It is obvious that $\sigma(x_i,z)$ is a function of $E_a$, however, the strength of the $E_a$ depends on the distance travelled, decaying rapidly as it propagates away from the aperture along the surface. Utilizing a resonant aperture strengthens the $E_a$ over the surface of the BE. To demonstrate this, a hole and a cross-shaped aperture were modelled in 3D. My previous simulations [24] considered a symmetric cross-shaped aperture with arm-lengths 170 ± 5 nm and arm-widths 40 nm perforated in a 100 nm silver film, laid in $z = 0$ plane, supported by a glass substrate with a refractive index $n = 1.52$. The refractive index data for silver were taken from Palik [25]. These simulations showed a peak in transmission at the target wavelength $\lambda_0 = 700$ nm, corresponding to the emission wavelength of nitrogen vacancies in nano-diamonds at room temperature [26,27]. The simulation is reproduced using an $x$-polarized TM wave at $\lambda_0 = 700$ nm, normally incident on the glass/silver interface. Full Maxwell equations were solved numerically using the finite element method (FEM). Simulations were repeated for a circular hole with a radius $R = 61.5$ nm. The radius of the hole was chosen so that it corresponds to the same surface area (or volume) as the cross. This will highlight the impact of the shape resonance associated with the cross-shape aperture on the surface fields. Electric field strengths at the silver/air interface calculated over the film surface produced by the cross and the hole apertures are depicted in Figure 2. The surface surrounding the cross, Figure 2(a)-(c), exhibits higher field amplitudes compared to that of the hole, Figure 2(d)-(f). Intensity ratios, $R_{x,y,z} = \frac{\int |E_{x,y,z}|^2_{\text{cross}}}{\int |E_{x,y,z}|^2_{\text{hole}}}$, calculated for each component over the film surface at the air/silver interface were found to be $R_x = 12$, $R_y = 27$ and $R_z = 32$. Here $|E|^2_{\text{cross}}$ and $|E|^2_{\text{hole}}$ are the field intensities associated with the cross and the circular hole respectively. My models also showed strong localization of the electric fields in the vicinity of the aperture with their amplitude decaying rapidly with the distance, see Figure 2(a)-(c). Furthermore, Figure 2(g), suggests that the separation between the LSPs and SPPs occur at $x \approx 200$ nm (see lines in brown and blue). Here $x = 0$ marks the centre of the aperture. Calculations of the time averaged radial component of the Poynting vectors $<S>_{\text{hole}}$ and $<S>_{\text{cross}}$, vs. the angular direction in the $x$-$z$ plane for the circular hole and the cross apertures, Figure 2(h), showed the power emanating from the cross (besides being stronger) is more directional in comparison to the hole. Enhancement to the overall throughput, $R = 11.3$, was calculated using $R = P_{\text{cross}}/P_{\text{hole}} = \frac{\int <S>_{\text{cross}} \cdot ds}{\int <S>_{\text{hole}} \cdot ds}$, where total powers, $P_{\text{cross}}$ and $P_{\text{hole}}$, were calculated by integrating the time averaged radial component of the Poynting vectors over the surface of a hemisphere, 2.3 $\mu$m in radius, encompassing the apertures.
Figure 2: Amplitudes of the x, y and z components of the electric field obtained from 3D models, calculated over the exit surface surrounding (a)-(c) the cross aperture and (d)-(f) the circular aperture. (g) z component of the electric field calculated in the vicinity of the cross aperture: (red) amplitude, |Ez(x)| (grey and blue) Ez(x,t) at two arbitrary times t1 and t2. (h) 3D calculations of the time averaged radial component of the Poynting vector vs. the angular direction in the x-z plane for (red) circular aperture and (blue) cross aperture.

Full 3D modelling of an aperture in the presence of corrugations is computationally expensive, particularly when parametric sweeps are performed over the geometry. To reduce the number of degrees of freedom, I have modelled the BE in 2D that exhibits strong resemblance to the 3D model as far as the radiation patterns are concerned. The radiation pattern of a 50 nm wide slit perforated in a 95 nm thick silver film on a glass substrate modelled in 2D, closely matches that of a 3D model discussed above. Corrugations were incorporated into the 2D model with depth h = 50 nm and number of grooves N = 7. The width of the periodic corrugations, was set to be a function of the periodicity, p, such that wp = p - 86 nm. This ensures that reflective surface at the bottom of the corrugations widen as the period increases. The 86 nm wide indentation was chosen to be 3.5 times the skin depth, to prevent the mixing of the charge oscillations formed on the adjacent vertical surfaces while allowing for the widest possible grooves. Note that numerical calculations set skin depth as S = 24.6 nm at λ0 = 700 nm. The r1 = 621 nm value was fixed at 0.89×λ0 [11]. The width of the first corrugation was selected so that the edge of the disk surrounding the aperture coincides with the strongest peak associated with the SPP wave, see Figure 2(g). The device was illuminated from the glass/silver side by a TM wave at normal incident travelling in the +z direction and parametric sweeps were performed over p. The transmitted far-fields were calculated over a closed arc encompassing the aperture and the corrugations. Integrating the x-component of the far-field intensity and plotting it vs. the period, revealed multiple maxima, see Figure 3(a). Although there are additional maxima beyond the range of data calculated here, to keep the device compact, a periodicity of p = 493 nm was chosen. In the case of t0 ~ S, see Figure 1(b), a new effect comes into play, namely the coupling of the x-components of the electric field from the substrate, Exg, to the LSPs inside the grooves. The increasing trend in the continuum with respect to p, seen in Figure 3(a), is attributed to the widening of corrugations with increasing p, that results in more of the incident power to leak through the film. This power leakage partially couples to
the vertical surfaces and partially is transmitted without any interaction. While the use of resonant aperture enhances the $E_a$, coupling of the $E_{xg}$ to LSPs inside the grooves presents an opportunity to further enhance $E_x$ directly. Amplitudes of the $x$ component of the electric polarization, $|P_x|$, inside the film and the total electric field, $|E_x|$, in the surrounding dielectrics in the $x$-$z$ plane calculated for $h = 50$ nm, Figure 3(c) and (inset), also confirms the formation of hotspots and the coupling between the $E_{xg}$ to the LSPs inside the corrugations.

Nominated configuration, C6, was selected with $r_1 = 621$ nm, $w_1 = 422$ nm, $p = 493$ nm $w_p = 407$ nm and $N = 7$. Configuration reported by Yi et al. [28], denoted as $C_{\text{ref}}$, was also modelled at $\lambda_0 = 660$ nm for comparison. Table 1 lists the dimensions associated with each device.

Table 1: Summary of the configurations for C6 and $C_{\text{ref}}$ configurations.

| Device | $r_1$ (nm) | $w_1$ (nm) | $p$ (nm) | $w_p$ (nm) |
|--------|------------|------------|----------|------------|
| C6     |            |            |          |            |
| $d = 50$ nm | 621        | 422        | 493      | 407        |
| $h = 50$ nm |            |            |          |            |
| $t = 95$ nm |            |            |          |            |
| $C_{\text{ref}}$ |            |            |          |            |
| $t = 300$ nm |            |            |          |            |
| $d = 300$ nm | 620        | 200        | 620      | 200        |
2.1. Excited with a normally incident light

To compare the performance of the C6 configuration in the presence of a resonant cross aperture vs. a circular hole, corrugations were incorporated into the 3D models. The number of corrugations, however, were limited to four, (instead of \( N = 7 \)), to reduce the computational resources needed. For each device, simulations were first carried out with a Gaussian incident beam \( E_x = \frac{1}{\sqrt{2\pi}} e^{-\frac{x^2+y^2}{2\alpha^2}} \), where \( \alpha \) is the standard deviation that sets the beam width. Notice that the standard deviation is eliminated from the denominator of the multiplier, i.e. first term in the RHS, to keep the incident power over the aperture constant while varying the width of the incident beam.

With \( \alpha = 199 \) nm, contributions resulting from the \( E_{xg} \) to LSP coupling are minimized, Figure 4(a)-(g). This ensures that the interaction between the corrugations and \( E_x \) is more observable, hence comparable. Simulations were then repeated with an incident beam covering the whole device to include the \( E_{xg} \) to LSP coupling, Figure 4(h). Notice the higher amplitudes of the electric field components on the surfaces surrounding the cross, Figure 4(a)-(c), compared to those of the circular hole, Figure 4(d)-(f). The cross-shaped aperture and the disk surrounding it constitute a “resonant system” that brings about further enhancement in the transmitted power and improvement to the directionality. Radiation pattern of the BE with the cross shows 5 times the intensity of the BE with the circular hole, Figure 4(g). Comparing the radiation patterns of
the cross in isolation, Figure 2(h) (line in blue), and in C6 settings, Figure 4(g) (lines in blue), transmitted power along the optical axis is enhanced by a factor of ~10, accompanied with a significant reduction in angular divergence. Radiation patterns shown in Figure 4(g) were calculated along a semi-circle, 2.3 \( \mu \)m in radius measured from the centre of the BE. However, when the incident beam waist covered the whole device, the two radiations pattern became comparable with that of the cross showing slight improvement over that of the hole, see Figure 4(h).

I must state that at the time of this exercise I was misled to believe circular holes lack resonances. Consequently, above comparison between cross-shaped apertures and circular holes must not be taken as a conclusion that cross-shaped apertures outperform circular holes at all time. I am confident with proper optimization, a better performance would be achieved by circular holes, see Appendix B for a discussion on resonance of circular holes.

However, the actual far-field zone associated with C6 extends tens of microns away from the device. Figure 5 shows calculated far-fields using 3D models for the cross and circular apertures in the C6 settings.

![Far-field radiation patterns](image)

**Figure 5**: Far-field radiation patterns. The cross aperture in the C6 setting (a) with \( \alpha = 199 \) nm and (b) with the incident beam covering the whole device. Circular hole aperture in the C6 settings (c) with \( \alpha = 199 \) nm and (d) with the incident beam covering the whole device.

Comparing the results for \( \alpha = 199 \) nm, (Figure 5 (a) and (c)), the enhancement to the far-field due to the presence of a resonant cross-shaped aperture is 1.5 times that of the hole. But when including the \( E_{xx} \rightarrow \) LSP coupling, (Figure 5 (b) and (d)), differences in far-field amplitudes and radiation patterns are barely detectable, with that of the hole showing slight improvement over that of the cross. Interestingly, Figure 5(a) is comparable to Figure 2(h)-line-in-blue and Figure 5 (c) to Figure 2(h)-line-in-red. One may even conclude that circular holes confine light to a subwavelength area more efficiently.

When illuminated with a normally incident light, it is not possible to isolate the contribution of the leaked power that couples to the vertical surfaces from that transmitted directly into the air half-space. A comparison between C6 and C_{ref}, therefore, must be carried out using an optically thick film. The film thickness in the C6 configuration therefore was set to \( t = 300 \) nm. The
aperture width of the C6 was also set to \( d = 318 \) nm in order to maintain the same aspect ratio \( d/\lambda_0 \) specified for \( C_{\text{ref}} \). Figure 6 shows the radiation patterns of (a) C6 and (b) \( C_{\text{ref}} \), with both film thicknesses set to \( t = 300 \) nm and the same \( d/\lambda_0 \) ratios. The radiation pattern associated with \( C_{\text{ref}} \) showed stronger side lobes in comparison to C6, where the radiation pattern is well confined to the central lobe. Although the radiation pattern of \( C_{\text{ref}} \) showed a longer reach along the central lobe, its intensity drops rapidly beyond 3 \( \mu \)m from the surface (in comparison to 8 \( \mu \)m observed in C6).

![Figure 6](image.png)

How do the C6 and \( C_{\text{ref}} \) configurations interact with the power emitted by a dipole placed inside the holes? Can these structures enhance the RDR of a single photon emitter while retaining their beaming qualities? What is the most efficient way of integrating a nano-diamond with the aperture? These questions are answered in the next section.

### 2.2. C6 with a nano-diamond inside the aperture

An isolated slit having a width \( d = 50 \) nm with a nano-diamond at its centre was modelled in 2D. The centre of the nano-diamond was configured with a dipole having its moment oriented along the \( x \)-axis. Total power scattered into the air was calculated at \( \lambda_0 = 700 \) nm for various nano-diamond diameters in the range of 40 nm \( \leq D \leq 50 \) nm. Maximum power was obtained for \( D = 50 \) nm when the nano-diamond makes contact with the inner surfaces of the cavity. Various positions along the slit’s axis, i.e. \( z \)-axis, revealed that the power ratio \( P_{\text{air}}/P_{\text{sub}} \) is maximized when nano-diamond is positioned at \( z = 25 \) nm where the nano-diamond makes contact with the substrate. Here \( P_{\text{air}} \) and \( P_{\text{sub}} \) are the total powers emitted into the air and the substrate respectively and the position \( z \) is measured from the centre of the nano-diamond. For convenience, lets label this source system as “50/25”, signifying a 50 nm wide slit (perforated in a silver film) with a nano-diamond that is 50 nm in diameter, positioned at \( z = 25 \) nm at the centre of the slit. Here \( z = 0 \) marks the silver/glass interface.
Positioning a 50 nm wide nano-diamond at the centre of C_{ref} (i.e. the centre of the hole that is 300 nm in diameter) did not result in a strong coupling between the dipoles radiation and the hole, Figure 7(a). Performance of C_{ref} in the presence of the 50/25 system was also inadequate, see Figure 7(b). C6 configuration, on the other hand, exhibits a highly directional light with strongest intensity along the central lobe.

Figure 7: |E|^2/1.3x10^{17} (V/m)^2 for (a) C_{ref} with 50 nm in diameter nano-diamond positioned inside the aperture, (b) C_{ref} in the presence of the 50/25 system and (c) C6 C_{ref} in the presence of the 50/25 system.

To make a fair comparison between C6 and C_{ref}, both devices were excited with the 50/25 source system. Optimum thicknesses for C6 and C_{ref} in this case were found to be \( t = \{365 \text{ and } 350\} \) nm respectively, corresponding to the second Fabry-Pérot resonance of each slit in the presence their respective corrugations [29]. Therefore, it is safe to say that the comparison between the two would highlight the impact of BE geometry in the presence of a resonant slit and in the absence of any leakage. The enhancement to the RDR, \( RDRE = P_{air}/(0.5 \times P_{nano-diamond}) \), for C6_{t=365nm} and C_{ref_{t=350nm}} were found to be 30 and 34 respectively. Here \( P_{nano-diamond} \) is the radiated power of a 50 nm nano-diamond in vacuum and \( P_{air} \) is the power radiated by the same nano-diamond into the air half-space when integrated.
with the device. The factor of 0.5 in the denominator is due to the $P_{\text{nano-diamond}}$ being calculated along the arc length of a full circle, whereas $P_{\text{air}}$ was calculated over a semicircle in the air half-space. Radiation patterns produced by C6$_{t=365\text{nm}}$ and C$_{\text{ref-} t=350\text{nm}}$ are depicted in Figure 8(a) and (b) respectively and the x-component of the electric field at the silver/air interfaces of both devices are shown in Figure 8(c).

Radiated power of C$_{\text{ref}}$ becomes extremely weak along the central lobe beyond 2 microns from the surface. C6 on the other hand showed that most of the radiation is directed towards the central lobe extending ~9 microns away from the surface. C6 configuration was also modelled with $t = 120$ nm and $h = 70$ nm in the presence of the 50/25 source system where the emission was found to be at its maximum. Figure 9(a) shows the electric field intensity scattered by the device.

![Figure 9](image.png)

Figure 9: (a) $|E|^2/1.3 \times 10^{17}$ (V/m)$^2$ scattered by C6$_{t=120\text{nm}}$ when excited by the 50/25 source system with $t = 120$ nm and $h = 70$ nm. (b) Simulated directional gain of the antenna $|E(\theta)|^2 / |E_0(\theta)|^2$ as a function of angle, $\theta$, from the optical axis. (c) 3D simulation of the $|E|^2/1 \times 10^{31}$ (V/m)$^2$ scattered by C6$_{t=120\text{nm}}$ with the cross arm-lengths $L = 140$ nm and arm-widths $W = 30$ nm. Film thickness, the corrugations, nano-diamond diameter and its position inside the aperture were set according to the 2D model.

Directional gain of the antenna $|E(\theta)|^2 / |E_0(\theta)|^2$, Figure 9(b), shows that the field intensity along the optical axis of the device is 125 times that of a free standing nano-diamond in vacuum. Here $|E(\theta)|^2$ and $|E_0(\theta)|^2$ are the electric field intensities of the nano-diamond in the C6 settings and in vacuum respectively as a function of angle, $\theta$, from the optical axis. Placing a nano-diamond inside the 3D cross-shaped aperture, however, required further investigations to identify its optimum dimensions. 3D simulations revealed that a cross with arm-lengths $L = 140$ nm and arm-widths $W = 30$ nm perforated in a 120 nm silver film has its resonance at $\lambda_0 = 700$ nm when excited with a nano-diamond (50 nm in diameter) positioned at $z = 25$ nm at its centre. Figure 9(c) shows that the electric field intensity in the 3D model agreeing with the 2D model.

Further enhancement to the decay rate, ($RDRE = 39$ for C6$_{t=120\text{nm}}$ compared to 30 for C6$_{t=365\text{nm}}$), however, is partly due to the difference in powers emanating from the slit at two different
thicknesses. To quantify the impact of the leakage alone, the loss/gain factor due to the grooves were calculated as \((1-P_{\text{air-hole}}/P_{\text{air-C6}})\times 100\%\) and was found to be -5% and +6% at thicknesses \(t = \{365, 120\}\) nm respectively. Here \(P_{\text{air-hole}}\) is the power emitted into the air by the source system in the absence of any groove. This is a total of 11% gain due the \(x\)-component of the electric field that propagates along the silver/glass interface that partially couples into the corrugations via the leakage, see Figure 10. This is a remarkable result given that the power ratio \(P_{\text{sub-hole}}/P_{\text{total-hole}} = 0.22\) indicates that an isolated 50/25 source system (with thickness \(t = 120\) nm) scatters only 22% of the total power into the substrate with only \(P_{\text{sub-surface-hole}}/P_{\text{total-hole}} = 18\%\) available on silver/glass surface.

![Figure 10: x-component of the electric field calculated for C6 with at an arbitrary time for (a) \(t = 365\) nm and (b) \(t = 120\) nm. (Red =1, blue = -1, green =0).](image)

### 2.2.1. Experimental trials

During the course of this project, nano-diamonds that exhibit anti-bunching properties were identified and characterized. The idea behind the pick-and-place approach, was to identify and characterize a single photon emitter prior to its integration with any plasmonic device followed by a post-integration characterization to detect any changes in the RDR/life-time. Positioning a nano-diamond inside a plasmonic aperture/cavity using the pick-and-place technique, however, proved to be a challenge. Attractive forces between the tip of the micromanipulator and the nano-diamond (perhaps Van Der Waals) created an undesirable scenario where the nano-diamond became permanently attached to the tip. It was also argued that the pick-and-place approach may also change the orientation of the NV- with respect to that in which it was characterized, hence voiding any measurements of its life-time/ant-bunching made prior to the integration. Furthermore, forcing the nano-diamond into the cavity, if achievable, may have some undesirable side effects such as distorting the apertures geometry, hence shifting its resonance away from the target wavelength. In the following section, a novel technique is proposed for harnessing the radiation of a dipole that may help avoiding such complications.

### 2.2.2. Surface Plasmon-Coupled-Enhanced Transmission in diamond substrates with a NV\(^{-}\) near the surface

The work represented in this section became the basis for more elaborated manuscript that introduced the concept of Surface Plasmon Coupled Enhanced Transmission (SPCET) as well as the procedural aspect concerning the optimization of the BE structure that led to a totally different configuration than C6 [30]. The main goal of this section is to apply SPCET to C6
configuration that includes skin depth for comparison appropriate for this report. Therefore, the simulations are carried out on the C6 configuration with the original $t = 100\, \text{nm}$ and $h = 50\, \text{nm}$, unless specified otherwise. Previously I mentioned the SPCE and I quote:

("Theoretical and experimental studies of surface plasmon-coupled emission have shown that fluorophores within about 10 nm of the metal are quenched. Hence surface plasmon-coupled emission occurs for fluorophores in a region 10 to 50 nm above the metal" [13]).

However, this quenching effect, is often misinterpreted due to the theoretical studies and the experimentally reported data being limited to the calculated/measured "reflected" power of a dipole near a flat metallic surface [31-34]. In fact, this “quenching” effect in the region less than ~10 nm away from the metallic surface is nothing but a strong coupling of the dipole’s radiation to the surface waves that propagate at the supercritical angle along the metal/dielectric interface [30]. Device proposed in this report was developed so that a single crystal diamond membrane with nitrogen vacancy implants near the surface [35] is used as a substrate. The C6 configuration was simulated in 2D with a diamond substrate where the incident field was replaced by the radiation of a dipole positioned at $(0, z)$, with its moment oriented in the $x$-direction. Here $z = 0$ marks the diamond/silver interface. The power ratio $P_{\text{air-C6}}/P_{\text{sub-C6}}$ vs. the $z$ is shown in Figure 11(a).

![Figure 11: (a) Power ratio vs. the $z$ calculated as $P_{\text{air-C6}}/P_{\text{sub-C6}}$ where $P_{\text{air-C6}}$ and $P_{\text{sub-C6}}$ are the total power scattered into the air and the substrate respectively. (b)-(g) $|E|^2/1.3\times10^{17} \, (\text{V/m})^2$ vs. the dipole distance from the surface. (h) weakly coupled regime occur between the surface modes and the dipole’s emission occurs at $z \leq -200\, \text{nm}$, where the film becomes reflective.

The maximum power ratio of $P_{\text{air-C6}}/P_{\text{sub-C6}} \approx 4$ is achieved for $z \approx -10\, \text{nm}$, for which the $P_{\text{air-C6}}/P_{\text{air-noFilm}} = 9$ is the enhancement due to the presence of the C6 device, where $P_{\text{air-noFilm}}$ is the total power scattered into the air in the absence of C6. And if one defines the RDRE with respect to the radiation of a free standing nano-diamond in vacuum, $P_{\text{nano-diamond}}$, the enhancement rises to $\text{RDRE} = P_{\text{air-C6}}/0.5 \times P_{\text{nano-diamond}} = 40$. Beaming profiles for various $z$ values are depicted in Figure 11(b)-(g). In the range of $0 > z > -35\, \text{nm}$, most of the power emanating from the dipole is coupled into the corrugations, via the slit, producing transmitted beams that are collimated. For $z < -35\, \text{nm}$, the coupling strength decreases and ultimately at $z = -200\, \text{nm}$, fields from the dipole decouple from the surface and the diamond/silver interface becomes reflective, Figure 11(h). In Figure 1(b) I described the formation of dipolar point sources above the metallic surface with part of their emission being reflected by the surface. When I considered the physical effects governing Figure 1(b) in conjunction to that of
11(h), intuitively it became apparent that a series of quantum emitters above a metallic surface, if positioned carefully, may harness the enhanced emission as well as the reflection by the metallic surface in producing a highly directional collimated beam. This became the foundation of a new highly directional pixel technology I proposed [36].

Further simulations were carried out for few different scenarios. A diamond substrate in the absence of any silver film with a dipole positioned at \( z = -10 \) nm, Figure 12(a), resulted in a power ratio of \( \frac{P_{\text{air-noFilm}}}{P_{\text{sub-noFilm}}} = 0.2 \). This is a clear indication that in the absence of any plasmonic structure, the larger portion of the emitted power is scattered inside the diamond. Introducing a 100 nm thick silver film on top of the diamond membrane, Figure 12(b), reduced the scattered power into the substrate by only 20% while the transmitted power became negligible. If a 50 nm wide slit perforates the flat 100 nm thick silver just above the dipole, Figure 12(c), power ratios \( \frac{P_{\text{air-Ag100nm+hole}}}{P_{\text{sub-Ag100nm+hole}}} = 6.5 \), \( \frac{P_{\text{air-Ag100nm+hole}}}{P_{\text{sub-noFilm}}} = 9.5 \) are obtained, however, the radiation pattern in this case is dispersive. So it seems that by positioning the dipole inside the substrate just 10 nm (or less) below the aperture, most of the dipole’s radiation is scattered into the superstrate via the dipole-cavity coupling. A separate model with the C6 configuration having a film thickness \( t = 200 \) nm while retaining all other parameters, resulted in \( t - h = 150 \) nm being much larger than the skin depth of 25 nm. Although the radiation pattern of \("C6Ag200nm\) is similar to that of the C6, see Figure 12(d), the power ratio \( \frac{P_{\text{air-C6-Ag200nm}}}{P_{\text{sub-C6-Ag200nm}}} \approx 0.5 \), (compared to \( \frac{P_{\text{air-C6}}}{P_{\text{sub-C6}}} \approx 4 \)) is obtained. Reduction in the power ratio is due to the increased film thickness that shifts the Fabry-Pérot resonance [29] which also reduces the coupling via the leakage. The process of quantifying each effect is not repeated here. The comparison between the C6Ag200nm and C6 is an indication of the sensitivity of the absorbed power by the substrate with respect to the film thickness. Note the higher field accumulation along the diamond/silver interface in Figure 12(d). Another scenario is when the corrugations are milled through the film, i.e. \( t - h = 0 \), Figure 12(e). In this case the power ratio \( \frac{P_{\text{air-C6-h100nm}}}{P_{\text{sub-C6-h100nm}}} = 0.92 \) (compared to 4 for \( h = 50 \) nm) is a clear indication that the film underneath the corrugations also participates in harnessing the fields from the silver/diamond interface. Strong field distributions along the film surface observed in Figure 11(b) is an indication of an underutilized power.

Fabrication of ultra-thin diamond membranes in the shape of rings is now a possibility [37]. Filling the corrugations with such a high refractive index material assists in trapping the fields that travel along the exit surface. With a properly designed BE structure, this results in further enhancements to the LSPs inside the grooves which ultimately fortifies the coupling between the substrate and the corrugations via the leakage. Figure 12(f) depicts the intensity of the electric field when the substrate and the material filling the C6 corrugations are set to diamond. In this configuration, although the power ratio \( \frac{P_{\text{air-C6}}}{P_{\text{sub-C6}}} = 2.7 \) (compared to 4 in Figure 11(c)), the transmitted beam profile shows no distinct side lobes with most of the power being redirected towards the central lobe. The enhancement to the radiative decay rate in this case was found to be \( \text{RDRE} = \frac{P_{\text{air-C6}}}{P_{\text{sub-C6}}} \approx 45 \) (compared to 40 in Figure 11(c)). With the diamond substrate and diamonds filling the grooves, the loss/gain factor, \( (1-P_{\text{air-hole}}/P_{\text{air-C6}})\times100\% \), at two thicknesses corresponding to the Fabry-Pérot resonances of the slit in the presence of the grooves were calculated as -21% and -4% for \( t = \{360, 110\} \) nm respectively, i.e. a total of 17% gain (compared to 11% in the previous section) purely due to the coupling of the x-components of the electric field via the leakage.
Figure 12: $|E|^2/1.3 \times 10^{17}$ (V/m)$^2$, for a diamond substrate with an NV- positioned 10 nm below the surface with (a) no silver film, (b) 100 nm thick silver film, (c) 100 nm thick silver film with a 50 nm wide slit, (d) C6 configuration with $t = 200$ nm, (e) C6 when corrugations are milled all the way through the film and (f) C6 having refractive index of the material filling the corrugations set to 2.41.

One can therefore design a BE structure with no aperture and purely based on the leakage. One possible design criteria may be based on the wave function:

$$
\psi = \frac{1}{\sqrt{2}} \left( \psi_a e^{ik_\alpha y} + \psi_g e^{ik_\beta y} \right)
$$

where $\psi_a e^{ik_\alpha y}$ and $\psi_g e^{ik_\beta y}$ are the surface waves travelling at the silver/air and the silver/diamond interfaces respectively. Here the probability amplitude of $1/\sqrt{2}$ implies a state with equal contributions by SPPs from both the substrate and the superstrate. In an optically thick silver film, SPP wavelengths at the air/silver and the diamond/silver interfaces were numerically calculated for $\lambda_0 = 700$ nm to be $\lambda_a = 2\pi/k_a = 667$ nm and $\lambda_g = 2\pi/k_g = 230$ nm. To satisfy the requirement for equal contributions, (hence equal probabilities of $(1/\sqrt{2})^2$ for periodicity to match SPP wavelengths from both sides of the film) the periodicity may be set to $P = (\lambda_a + \lambda_g)/2$. While SPPs launched at the air/silver interface couple to the corrugations through direct propagation along the surface, those at the diamond/silver interface may couple into the corrugations via the leakage. Setting the film thickness to $t = 100$ nm prevents mixing of the SPPs from the two sides. The width of the corrugations were narrowed to $w = 50$ nm to minimized the surface area they occupy. The interaction between surface waves at the two interfaces is then limited only to their coupling to the LSPs inside the grooves. Depth of the corrugations were set to $h = 70$ nm (hence $t_c = 30$ nm) using a trial/error approach until a strong collimated beam was obtained. Figure 13 shows the radiation patterns of one possible configuration, C$_x$, with no aperture. In summary the film thickness, periodicity, height and the width of the corrugations in C$_x$ were set to $t = 100$ nm, $P = 445$ nm, $h = 70$ nm and $w = 50$ nm respectively with the corrugations “centred” at $m \times P$, where $m$ is an integer. Eliminating the aperture and the first groove surrounding it simplifies the design to a large extent, making the device suitable for mass production fabrication techniques, such as the single step nanoimprinting lithography [38]. Note that in this chapter, 2D field plots in Figure 7, Figure 8, Figure 9, Figure 10, Figure 11, Figure 12 and Figure 13 were produced on the same scale to facilitate the comparison between devices.
Figure 13: $|E|^2/1.3 \times 10^{17}$ (V/m)$^2$ calculated for the Cx configuration that has no aperture with periodicity matching the SPP wavelength at (a) silver/air interface $\lambda_a = 2\pi/k_a = 667$ nm, (b) silver/diamond interface $\lambda_g = 2\pi/k_g = 230$ nm and (c) $P = (\lambda_a + \lambda_g)/2$. Device was excited with a NV- positioned at $z = -10$ nm from the silver/diamond interface.

Fabricating the device on a diamond membrane with a NV positioned at $z = -10$ nm is a challenge and beyond the scope of this thesis. Perhaps, in the near future, when the relevant technology matures, such a device becomes realizable. Therefore, the BE devices were fabricated on the glass substrate and characterized by simply measuring their transmission when they were illuminated with an incandescent light filtered at $\lambda_0 = 700$ nm, incident on the substrate. In the next section fabrication and characterization of one such device is reported.

3. Fabrication and Characterization

A 2 nm thick germanium film followed by a 100 nm thick silver film were deposited on a glass substrate using IntlVac Nanochrome II electron beam evaporator. The germanium layer acts as an adhesion layer between the glass and the silver film and has little impact on the overall optical properties of the device. Corrugations for the C6 were engraved on the silver film according to the target dimensions discussed above with a depth of $h = 50$ nm, using a Focused Ion Beam (FIB) (Helios NanoLab 600). The aperture was milled through the film as a symmetric cross with target arm-lengths $L = 175$ nm and arm-widths $W = 40$ nm. It is worth mentioning that the preparation of bitmap files in my previous attempts to mill the cross-shaped apertures was based on a pixel size occupying a $10 \times 10$ nm$^2$ area on the film surface. With a Gaussian beam profile and an effective beam diameter of 7.5 nm, (corresponding to the 1.5 pA ion current), milling a perfect 10 nm $\times$ 10 nm square pixel on the film surface is impractical, see Figure 14(a). Here, a new technique was employed based on a 3.25 nm per pixel resolution, while keeping the incident ion beam as above. Such a technique leads to a beam overlap between the neighbouring pixels, Figure 14(b), that produce a cross-shaped aperture with abrupt geometries, hence improve the fabrication quality.

Figure 14: a) Ion beam 7.5 nm in diameter produces a gap of 2.5 nm between the neighbouring pixels when the FIB resolution is set to 10 nm per pixel. b) With the resolution of the FIB set to half the beam diameter, i.e. 3.25 nm per pixel, ion beam overlaps between the two neighbouring pixels on the film surface.
Fabricated device dimensions are listed in Table 2 measured from the relevant SEM images, Figure 15.

![Fabricated device images](image)

**Figure 15:** SEM images for the fabricated C6 configuration. (a) Top view. (b)-(c) Close-up images for the aperture. (d) Close-up of the top view.

|   |   |   |   |   |   |   |
|---|---|---|---|---|---|---|
| C6 | $r_1$ (nm) | $w_1$ (nm) | $p$ (nm) | $w_p$ (nm) | $L_x$ (nm) | $L_y$ (nm) |
| FIB | 634±12 | 425±3 | 506±13 | 413±6 | 185±2 | 177±2 |

The film was mounted on a 3D piezoelectric translational stage within a confocal microscopy system. The glass/silver interface was illuminated with an unpolarized collimated white light source (Supercontinuum Fianium SC-450-2), band-pass filtered at $\lambda = 700$ nm with a FWHM of $10 \pm 2$ nm (Thorlabs FB700-10). Transmitted intensities were collected via a LU PLAN Nikon 100× objective with a numerical aperture of NA = 0.95 and coupled into a multimode fibre with a core diameter of 50 µm (acting as a pinhole). The light was detected using high sensitivity single photon counting modules (SPCM). The detector and the stage were controlled using a LabView program, which also collected data. Here, the surface of the film is taken to be parallel to the $x$-$y$ plane. A set of 2D images were obtained by raster scanning in the $x$-$y$ plane. The closest distance between the objective and the sample, $z = z_{\text{min}}$, was set by an algorithm that maximized the photon counts as a function of $z$. The distance was increased in steps of $\Delta z = 0.1$ µm, between each scan up to 25 µm. The 2D images were stacked to form a high-resolution 3D volumetric map of the Fresnel zone. The process was then repeated with $\Delta z = 1$ µm to map the intensity over longer distances up to 55 µm. Although $\Delta z = 1$ µm results in a lower resolution in $z$, it reduces the time required for longer range measurements where the intensity variation with respect to $z$ is relatively slow.

### 4. Results and Discussion

Figure 16(a) shows the simulated result for $|E|^2$ along the optical axis. Depth of focuses, DOF$_1$ and DOF$_2$ are the lengths over which the intensity remains greater than half the local maxima at $z_1 = 5.8$ µm and $z_2 = 27$ µm respectively. Figure 16(b) depicts the $|E|^2$ in the $x$-$z$ plane. Normalized $|E|^2$ calculated along the $x$-axis at the two maxima. Figure 16(c)-(d), reveals the lateral beam profiles where $\delta_1$ and $\delta_2$ are the corresponding FWHM.
Figure 16: Simulated results for the C6 configuration using a 2D model. (a) Maxima positions and their corresponding depth of focuses identified on $|E|^2$ along the optical axis. (b) $|E|^2$ in the x-z plane. (c)-(d) $|E|^2$ calculated along the x-axis at their maxima.

Figure 17 and Figure 18 depict the results obtained experimentally. A 3D iso-surface obtained from the volumetric data corresponding to the $0.5 \times I_{\text{max}}$, where $I_{\text{max}}$ is the maximum photon counts, exhibits a rotational symmetry about the z-axis, see Figure 17(a). Normalized photon counts along the central lobe, Figure 17(b), and a 2D slice in the y-z plane obtained from the volumetric map, Figure 17(c), show good agreement with my simulations. The experimental results for $\delta_1$ and $\delta_2$ at their corresponding maxima were obtained from the normalized photon counts in the x-y plane, $(I(x,y)-I_{\text{min}})/(I_{\text{max}}-I_{\text{min}})$, where $I_{\text{min}}$ is the background radiation, see Figure 17(d)-(e). Table 3 summarizes the results. Figure 18 depicts a series of beam profiles in x-y plane at various distances, z, from the device. From top left to right bottom, each image represents unprocessed/unnormlized photon counts in a 10 μm × 10 μm area, detected at distances ranging $5 \leq z \leq 22$ μm. Interestingly, prior to the beam evolves into a gaussian profile, it goes through various topological profiles. Note the formation of vortex, ideal for optical twiners and trapping ions, in the range of $11 \leq z \leq 14$ μm, and Bessel-like profile just before and after. Most significant aspect of this experiment was the excitation with an unpolarized light. Considering the argument I presented regarding the x-component of the transmitted electric field dominating the far-field in a 2D numerical model, what may be inferred with respect to 3D measurements obtained experimentally when the device was excited with an unpolarized light, is intuitive. I can infer with confidence that photons depicted in the x-y plane, as seen in Figure 18, are radially polarized with respect to the z-axis. This is very intuitive considering that the device has a rotational symmetry about the z-axis.
Figure 17: Experimental results for the C6. (a) 3D iso-surface corresponding to the $0.5xI_{\text{max}}$. (b) Normalized photon counts along the central lobe. (c) A 2D slice in the $y$-$z$ plane obtained from the volumetric map. (d)-(e) Normalized photon counts in the $x$-$y$ plane at $z_1 = 5 \mu m$ and $z_2 = 26 \mu m$.

$\Delta z = 1 \mu m$, distance increasing left to right

- $z = 5$ to $10 \mu m$
- $z = 11$ to $16 \mu m$
- $z = 17$ to $22 \mu m$

Figure 18: Raw data of beam profiles in $x$-$y$ plane at various distances from the devise. From top left to right bottom, each image represents unprocessed photon counts in a $10 \mu m \times 10 \mu m$ area, detected at distances ranging $5 \leq z \leq 22 \mu m$. Colors blue and white represent the regions with minimum and maximum number of detected photons respectively.

Table 3: Locations of maxima and their corresponding full width half max and depth of focuses of the beam for the C6 configuration.

| Simulation | Experiment | Simulation | Experiment |
|------------|------------|------------|------------|
| $z_1$ ($\mu m$) | $5 \pm 0.5$ | $z_2$ ($\mu m$) | $26 \pm 0.5$ |
| $\delta_1$ ($\mu m$) | $0.9 \pm 1\%$ | $\delta_2$ ($\mu m$) | $2.35 \pm 2.3\%$ |
| DOF1 ($\mu m$) | N/A | DOF2 ($\mu m$) | 55 |
| 6 | 32$\pm 0.5$ |
Note that in Table 3, the resolution error in $z$ was calculated as $\Delta z/2$. The average FWHM and the relevant error, in percentage, were calculated using $\delta_i = 0.5 \times (\delta_{ix} + \delta_{iy})$ and $\Delta \delta_i = 50 \times (\delta_{ix} - \delta_{iy})/\delta_i$ respectively. The differences between the simulated and the experimental results are attributed to the fabrication artifacts such as imperfections in the milled cross/corrugations and the surface granularity, particularly inside the grooves. The use of experimentally obtained refractive index data for bulk silver of unknown surface roughness in simulating nanostructures that are modelled with perfectly smooth surfaces, also gives rise to discrepancies between the simulations and measurements[39]. However, unique to the experimental data was a series of maxima along the optical axis of the device. This is apparent from Figure 17(c). And in fact, there were more maxima observed along the optical axis beyond $z = 55 \mu$m, see Appendix C. It seemed that the transmitted light was composed of a collimated beam superposed with a series of focal points.

Apart from the C6, I modelled, fabricated and characterized a number of other bullseye configurations, each with interesting properties and a unique beam profile. Table 4 summarizes the dimensions for the first five configurations.

Table 4: Summary of target dimensions for the BE configurations

| Device | r1 (nm) | w1 (nm) | p (nm) | wp (nm) |
|--------|---------|---------|--------|---------|
| C1     | 336     | 107     | 714    | 107     |
| t = 95 nm | d = 50 nm | h = 50 nm |         |         |
| C2     | 720     | 490     | 650    | 140     |
| C3     | 1110    | 876     | 500    | 200     |
| C4     | 1090    | 600     | 525    | 187     |
| C5     | 636     | 415     | 486    | 400     |

Figure 19(a)-(b) depicts the simulated far-field intensities, $|E_{far}|^2$ and $|E_{far}|^2 / |E_{far}^{SH}|^2$ as a function of angle, $\theta$, from the optical axis for C1-C6 and $C_{ref}$ configurations. Here, $|E_{far}^{SH}|^2$ is the power transmitted through a single hole, 300 nm in diameter, perforated in a 300 nm thick silver film. In all cases, the simulation domain, the incident power and the number of corrugations ($N = 7$) were kept constant. Therefore, Figure 19(a)-(b) may be interpreted as the antenna efficiency and the gain respectively. C1 and C2 configurations have relatively low efficiency compared to other devices. C3-C6 exhibit between 4 to 5 times higher efficiency in the far-field zone and 4 to 5 time higher gain in the central lobe compared to that of the $C_{ref}$. 
Figure 19: Simulated far-field intensities, \(|E_{\text{far}}|^2\) and \(|E_{\text{far}}|/|E_{\text{SH}}|^2\) as a function of angle, \(\theta\), from the optical axis for C1-C6 and \(C_{\text{ref}}\) configurations.

Fabrication and characterization process are described in the previous section. Table 5 summarizes the fabricated dimensions for each device.

Table 5: Summary of fabricated dimensions for BE configurations, C1-C5

| device | \(r_1\) (nm) | \(w_1\) (nm) | \(p\) (nm) | \(w_p\) (nm) | \(L_x\) (nm) | \(L_y\) (nm) | \(f=(a-b)/a\) |
|--------|--------------|--------------|------------|-------------|-------------|-------------|-------------|
| C1     | 325±13       | 106±2        | 722±8      | 105±2       | 175±2       | 175±2       | 0.015       |
| C2     | 706±13       | 496±6        | 669±19     | 140±3       | 177±2       | 175±2       | 0.021       |
| C3     | 1090±10      | 870±5        | 515±15     | 202±2       | 165±2       | 177±2       | 0.017       |
| C4     | 1080±10      | 590±10       | 511±14     | 188±1       | 175±2       | 177±2       | 0.023       |
| C5     | 650±15       | 414±5        | 513±28     | 415±15      | 178±2       | 172±2       | 0.021       |

Simulated \(|E|^2\) and the measured intensity \(I\), (\(\Delta z = 0.1 \mu m\)), associated with C1 configuration are depicted in Figure 21(a)-(b) respectively. Both the \(|E|^2\) and the \(I\) were normalized with respect to their maximum values. Figure 21(c) shows the intensity, \((I(x,y)-I_{\text{min}})/(I_{\text{max}}-I_{\text{min}})\), at \(z = 0\) plane where \(I_{\text{min}}\) is the background radiation. Notice how the field is localized near the surface (as expected) and the extent of the intensity at the central lobe is less than 1 \(\mu m\).

Figure 20: SEM image for the fabricated C1.
Figure 21: (a) the simulated $|E|^2$ and (b) the measured intensity, $I$, with $\Delta z = 0.1 \mu m$ and (c) the intensity, $I(x,y)-I_{\text{min}}/(I_{\text{max}}-I_{\text{min}})$ at $z = 0$ associated with C1 configuration.

Figure 23(a) and (b) shows the simulated $|E|^2$ and the measured $I$, ($\Delta z = 1 \mu m$), in the $x = 0$ plane for the configuration (C2). Whereas (c), show the normalized intensity at $z = 0$, i.e. the device plane. Despite the high intensity surrounding the aperture, the beam diverges rapidly with increasing $z$, Figure 23(d). The divergence was calculated by measuring the angle between the two intercepting lines drawn along the dark bands of the beam profile and was estimated to be $26^\circ$. This is in good agreement with the simulation, see Figure 23(e).

Figure 22: SEM image for the fabricated C2.
Simulated far-field intensity for C3 configuration indicated a strong central lobe. The full field calculation of the intensity, $|E|^2$, along the optical axis and in the x-z plane are depicted in Figure 25(a) and (b) respectively. The FWHM of the simulated beam profile associated with the two distinct maxima at $z = 7.7 \, \mu m$ and $z = 36 \, \mu m$ were found to be approximately $\delta_1 = 0.88 \, \mu m$ and $\delta_2 = 2.74 \, \mu m$, see Figure 25(c) and (d). Corresponding experimental results are depicted in Figure 26. Maximization algorithm set the $z_{\text{ini}}$ to $\sim 5 \, \mu m$ away from the film’s surface. The normalized photon count along the optical path, Figure 26 (a), is obtained using $(I(z) - I_{\text{min}})/(I_{\text{max}} - I_{\text{min}})$, where $I_{\text{min}}$ is the background radiation. Beam profiles in the x-y plane, Figure 26(c)-(d), represent the normalized $I(x,y)$ values at their maxima locations, $z = \{7.6 \, \mu m, 36 \, \mu m\}$. Corresponding FWHM were calculated to be $\delta \approx \{1.2 \, \mu m, 2.88 \, \mu m\}$ respectively and are in good agreement with the simulations.
Configurations C4-C5, were treated accordingly and the results are summarized in Table 6.
Figure 28: C4, simulations.

Figure 29: C4, experimental.

Figure 30: SEM image for the fabricated C5.
Table 6: Locations of maxima and their corresponding FWHM of the beam for C3-C7 configurations.

|   | $z_1 \mu m$ | $\delta_1 \mu m$ | $z_2 \mu m$ | $\delta_2 \mu m$ |
|---|-------------|-----------------|-------------|-----------------|
| Sim | Exp $\pm \Delta z/2$ | Sim | Exp $\pm \Delta \delta_1$ | Sim | Exp $\pm \Delta z/2$ | Sim | Exp $\pm \Delta \delta_2$ |
| C3 | 7.75 | 7.6 ± 0.5 | 0.88 | 1.25 ± 7.2% | 36 | 36 ± 0.5 | 2.74 | 2.88 ± 1% |
| C4 | 8.8 | 6.2 ± 0.5 | 0.88 | 1.13 ± 4.8% | 38 | 38 ± 0.5 | 2.7 | 3.15 ± 4.8% |
| C5 | 5.5 | 5 ± 0.5 | 0.8 | 1.02 ± 6.9% | 28 | 25 ± 0.5 | 2.6 | 2.55 ± 6.3% |
| C6 | 5.8 | 5 ± 0.5 | 0.84 | 0.9 ± 1% | 27 | 26 ± 0.5 | 2.7 | 2.35 ± 2.3% |
| C7 | - | - | - | - | - | 35 ± 0.5 | - | 3.15 ± 7.3% |

The C3 configuration exhibits a slight broadening of the beam. The differences between the experiment and the simulated FWHM, $(\delta_{\text{exp}} - \delta_{\text{sim}})_{1,2} = \{0.37 \mu m, 0.14 \mu m\}$, are attributed to the granularity of the inner most corrugation, the position and the geometry of the cross, with respect to the corrugations, that fails to form a perfect concentric arrangement. The aperture dimensions also influences the SPP-LSP coupling and consequently the transmitted power. This can be demonstrated by comparing the C3 and C4 configurations. In the case of C4 configuration, the ratio $|E_1|^2/|E_2|^2 = 1.2$ is obtained from the simulation, where $|E_1|^2$ and $|E_2|^2$ are...
the field intensities at \( z_1 \) and \( z_2 \) respectively, see Figure 28. This value is very close to the experimentally measured ratio \( I_1/I_2 = 1.6 \), where \( I_1 \) and \( I_2 \) are the photon counts at \( z_1 \) and \( z_2 \) respectively, see Figure 29. On the other hand, the same ratios for the C3 configuration reads \( |E_1|^2/|E_2|^2 = 20 \) and \( I_1/I_2 = 4.3 \). Clearly, in the case of C3, the discrepancy between the measured and the simulated intensity ratios is due to the defect in the geometry of the aperture, i.e. the cross aperture is not fully milled through the film, compare SEM images in Figure 24(b)-(c) and Figure 27(b)-(c). A comparison between the fabricated C5 and C6 configurations, that are very similar in geometry, reveals the importance of the geometrical symmetry of the corrugations and their influence in shaping the beam profile. The differences \( \{6.9\%, 6.3\%\} \) in the FWHM observed for C5, compared to \( \{1\%, 2.3\%\} \) in C6, are attributed mainly to the inconsistency in the milled corrugations and partly to the presence of a debris near the aperture in C5, compare SEM images Figure 30 and Figure 15. Presence of the debris also results in dampening of the resonance system. The power ratio \( I_{C6}/I_{C5} = 1.2 \) calculated along the central lobes at \( z_2 \) is attributed to the dampening by the debris.

5. Conclusion

Strong SPP formations in the vicinity of the aperture led me to the idea of resonant disk that together with the aperture form a resonant system that not only exhibits higher scattered power but also improved directionality compared to a resonant cross aperture in isolation. The role of the corrugations in my design is to further enhance the parallel components of the electric field that dominate the far-field along the optical axis of the bullseye, by combining the strong field generated by the resonant system and the coupling of the incident field to the LSPs inside the grooves, via the leakage through the skin depth. The resonant condition observed is attributed to the complex interaction between the leakage of the incident field and its coupling with the grooves, LSP oscillations inside the grooves raised due to propagating SPPs and the field emanating from the resonant system that led to a constructive interference of the parallel to the surface component of the electric field in the far-field zone. In my measurements, 3D confocal beam profiling proved to be advantageous in producing a high-resolution volumetric map of the field intensities, revealing many interesting features that would otherwise go unnoticed.

6. Conflict of Interest

The author declares no conflict of interest. In the case of the computational resources or the fabrication/characterization facilities availed to the author being interpreted as fund of some sort, the author wishes to state that the founding sponsors had no role in the inception of the ideas, the design of the study; in the collection, analysis, or interpretation of data; in the writing of the manuscript, and in the decision to publish the results.

7. Appendix A: Radiative Decay Rate Enhancement

The spontaneous decay rate of a two level quantum system where \( |i\rangle \) &\( |f\rangle \) denote the initial and final states, is given by the Fermi’s Golden rule:
\[
\gamma = \frac{2\pi}{\hbar^2} \sum_j \langle i | \hat{H} | f \rangle \langle f | \hat{H} | i \rangle \delta (\omega_i - \omega_f)
\]  
(A1)

Note that in the spontaneous emission, unlike stimulated emission, emitted photons are not limited to a particular direction/polarization, consequently, the summation is performed over all non-degenerate final states, where each mode is identified by \(\omega_k\), i.e. the frequency of the \(k\)th mode, characterized by a particular polarization and wavevector. Although spontaneous emission is purely a quantum mechanical effect, it is possible to derive a relation for the decay rate in the terms of the dipole emission. In the dipole approximation, the interaction Hamiltonian operator of a dipole located at \(r_0\), is defined by \(\hat{H} = -\mathbf{p} \cdot \mathbf{E}\), where electric field operator at \(r = r_0\) is given by \(\mathbf{E} = \sum_k \mathbf{E}_k^+ \hat{a}_k^\dagger (t) + \mathbf{E}_k^\dagger \hat{a}_k^\dagger (t)\). The creator and annihilator operators are defined as \(\hat{a}_k(t) = \hat{a}_k(0)e^{-i\omega_k t}\) & \(\hat{a}_k^\dagger (t) = \hat{a}_k^\dagger (0)e^{i\omega_k t}\), with \(\omega_k\) denoting the frequency of the \(k\)th mode. Dipole moment operator is defined as \(\hat{\mathbf{p}} = \mathbf{p} [\hat{r}^* + \hat{r}]\) with \(\hat{r}^* = |e\rangle \langle g| & \hat{r} = |g\rangle \langle e|\) where \(|g\rangle\) and \(|e\rangle\) are the orthogonal basis indicating the ground and the excited states respectively. Here the cap, \(\hat{\quad}\), signifies an operator. Expanding the Hamiltonian in terms of the quantities mentioned above results is:

\[
\hat{H} = -\mathbf{p} \cdot \sum_k \mathbf{E}_k^+ |e\rangle \langle g| \hat{a}_k(0)e^{-i\omega_k t} + \mathbf{E}_k^\dagger |g\rangle \langle e| \hat{a}_k^\dagger (0)e^{i\omega_k t} + \mathbf{E}_k^+ |g\rangle + \ldots
\]

(A2)

The initial and the final states in terms of ground, excited and photon states are given by:

\[
|i\rangle = |e\rangle |0\rangle = |e, 0\rangle
\]

\[
|f\rangle = |g\rangle |1_{\omega_k}\rangle = |g, 1_{\omega_k}\rangle
\]

Here \(|0\rangle\) denote the zero-photon state whereas \(|1_{\omega_k}\rangle\) corresponds to a state where the system has one photon associated to the \(k\)th mode. Evaluating the decay rate as stated in equation (A1) leads to:

\[
\gamma = \frac{2\pi}{\hbar^2} \sum_k \mathbf{p} \cdot (\mathbf{E}_k^+ \mathbf{E}_k^\dagger) \cdot \mathbf{p} \delta (\omega_k - \omega_0)\), where \(\mathbf{E}_k^+ = \sqrt{\frac{\hbar \omega_k}{2\epsilon_0}} \mathbf{u}_k\) & \(\mathbf{E}_k^\dagger = \sqrt{\frac{\hbar \omega_k}{2\epsilon_0}} \mathbf{u}_k^\dagger\), \(\mathbf{u}_k\) are the normal modes and \(\mathbf{p} = \mathbf{p}_u\mathbf{u}_p\) with \(\mathbf{u}_p\) being the unit vector in the direction of the dipole moment. Note that \(\mathbf{E}_k^+ \mathbf{E}_k^\dagger\) designates an outer product resulting in a \(3 \times 3\) matrix. Finally the decay rate can be simplified to [23],

\[
\gamma = \frac{\pi \alpha}{3\hbar \epsilon_0} |\mathbf{p}|^2 \rho (\mathbf{r}_0, \omega_0)\)

where

\[
\rho (\mathbf{r}_0, \omega_0) = \sum_k \mathbf{u}_p \cdot (\mathbf{u}_k \mathbf{u}_k^\dagger) \cdot \mathbf{u}_p \delta (\omega_k - \omega_0): \{\text{in the case of free space}\} \rightarrow \rho_0 = \frac{\alpha_0^2}{\pi^2 c^3} \rightarrow \gamma_0 = \frac{\omega_0^3}{3\pi \hbar \epsilon_0 c^3} |\mathbf{p}|^2
\]

(A3)

Equation (A3) shows that the spontaneous decay rate is the function of the local density of states \(\rho\). Furthermore, the density of states depends on the interaction of the dipole moment with its own secondary field arriving at the dipole’s location after being scattered by the
environment (such as a cavity surrounding it). Microscopic representation of the decay rate based on quantum mechanics, however, does not account for losses. Alternative representation, based on classical electrodynamic, correlates the spontaneous decay rate, $\gamma$, to a macroscopically observable quantity such as the power, $P$, in an inhomogeneous environment by [23]:

$$\text{decay rate enhancement} \equiv \frac{\gamma}{\gamma_0} = \frac{P}{P_0}$$

(A4)

For derivation see 8.3.3 and 8.5.2 in [23]. Here, $P_0$ is the power emitted by a dipole in a homogeneous environment (such as vacuum) and $\gamma_0$ is the FWHM of the Lorentzian line associated with the dipole’s frequency dependent emission in the same environment, see section 8.5.1 in [23] for the origin of the damping factor, $\gamma_0$. Depending on the inhomogeneous medium containing the dipole, some or all of the power emanating from the dipole maybe absorbed leading to an expression of the total power $P = P_{\text{far}} + P_{\text{abs}}$ where $P_{\text{far}}$ and $P_{\text{abs}}$ are the power detected in the far-field and the power absorbed by the medium (e.g. due to losses, surface mode excitations, quenching effects and more). Similarly, spontaneous decay rate should be expressed as $\gamma = \gamma_r + \gamma_{nr}$, where $\gamma_r$ and $\gamma_{nr}$ are the radiative and non-radiative terms [23]. Since $\gamma_r$ is proportional to $P_{\text{far}}$ the power ratio $\frac{P_{\text{far}}}{P_0}$ is a valid quantity, representing the enhancement or inhibition of the radiative part of the spontaneous decay rate, provided that $P_0$ be also measured in the far-field over the same area as $P_{\text{far}}$. The power ratio $\frac{P_{\text{far}}}{P_0}$ is used in this report to evaluate radiative decay rate.

8. Appendix B: Resonance of Circular Hole

A biperiodic array of circular holes in a silver screen immersed in optical oil with a refractive index $n = 1.52$ was modelled based on the same dimensions I reported in [40], also see section 7.2.1 of my thesis [41]. A parametric sweep over the film thickness, $h$, was performed, while keeping all other parameters unchanged. Incident field with $\lambda_0 = 790$ nm, was polarized at $\alpha = 90^\circ$ to excite the array’s (1,0)SPP mode in the $y$-direction. This is to ensure that the holes would benefit from the maximum excitation power that surface waves could offer. Consequently, any variations in $h$ and the transmitted power may be associated to the modes associated with the hole. Figure B 1(a) revealed circular or cylindrical holes do have resonances that can be explained in terms of standing waves formed along the axis of the cylinder that give rise to the Fabry-Pérot (FP) resonance at a given wavelength. The first FP resonance in this case occurs at thickness $h = 80$ nm. Figure B 1 (b) confirms that most of the incident power is transmitted through the device with minimum reflection when $h = 80$ nm, hence the resonance of cylindrical hole. This exercise may be extended to a single hole in a bullseye settings.
9. Appendix C

Figure C 1 depicts the y-z plane extracted from the experimentally obtained volumetric photon counts associated to the C6 configuration when the device was illuminated with an unpolarized incandescent light filtered at $\lambda_0 = 700$ nm. Two sets of measurements were carried out with $\Delta z = 0.1$ μm for finer resolution and $\Delta z = 1$ μm to obtain the extent of the far-field.

Figure C 1: Experimental results for the C6. 2D slices in the y-z plane obtained from the volumetric map with (a) $\Delta z = 0.1$μm and (b) $\Delta z = 1$μm

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